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Optical harmonic generation in hollow-core photonic-crystal fibres: analysis of optical losses and phase-matching conditions

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***Abstract*. We consider hollow-core fibres with a microstructure photonic-crystal cladding, which open a unique opportunity of implementing nonlinear-optical interactions of waveguide modes with transverse sizes on the order of several microns in the gas phase. Phase-matching conditions for optical harmonic generation can be improved in higher waveguide modes of hollow-core photonic-crystal fibres by optimising parameters of the gas medium filling the fibre and characteristics of the fibre.**

Keywords: *hollow-core photonic-crystal fibre, high-order harmonic generation, photonic crystal.*

1. Introduction

Nonlinear-optical processes in gas-filled hollow fibres are currently widely employed for the generation of ultrashort light pulses and frequency conversion by means of high-order harmonic generation and multiwave mixing. The possibility of improving the efficiency of nonlinear-optical interactions due to the increase in the interaction length in a gas-filled hollow fibre was experimentally demonstrated for the first time by the authors of paper [1], who demonstrated back in 1977 a considerable (by three orders of magnitude) enhancement of coherent anti-Stokes Raman scattering four-wave mixing process in a hollow dielectric fibre.

The authors of papers [2, 3] have shown that the use of a gas-filled hollow fibre allows the efficiency of spectral broadening of an ultrashort laser pulse due to self-phase modulation (SPM) to be improved. Laser pulses with an initial duration of 20 fs, propagated through a rare-gas-filled hollow fibre in these experiments, displayed an SPM-induced spectral broadening sufficient for subsequent pulse compression to unprecedentedly short pulse durations of 4.5 fs. Since the optical breakdown threshold for a gas filling a hollow fibre is normally much higher than the optical breakdown threshold typical of conventional optical fibres, hollow fibres permit high-power light pulses with only a few field cycles under their envelopes to be produced. The hollow-fibre-based technique of SPM spectral broadening with subsequent pulse compression proposed and implemented in papers [2, 3] is now widely employed in femtosecond laser systems [4].

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Another promising direction of using hollow fibres for the generation of extremely short light pulses involves high-order stimulated Raman scattering (SRS) [5 – 13]. High-order SRS in a hollow fibre filled with a Raman-active gas, leading to the in-phase generation of multiple Stokes and anti-Stokes Raman sidebands [8 – 12], allows, as recently reported [13], sub-4-fs light pulses to be synthesised.

Gas-filled hollow fibres appreciably increase the length of nonlinear-optical interactions and offer the ways to reduce the phase and group-velocity mismatch between the light pulses involved in frequency-nondegenerate nonlinear-optical processes, thus allowing the efficiency of nonlinear-optical frequency conversion through harmonic generation and multiwave mixing to be radically improved [14 – 21].

With an appropriate choice of parameters of a hollow fibre, gas pressure, and waveguide modes, the waveguide components of the phase mismatch and group delay partially or completely compensate for the phase mismatch and group delay due to the gas dispersion [22]. This opens the ways to substantially enhance harmonic-generation and multiwave-mixing processes (including the generation of short pulses of coherent X-ray emission) [14 – 22] and to reduce the influence of group-delay and group-velocity dispersion effects on the synthesis of extremely short field waveforms through the generation of multiple SRS components [23]. The sensitivity of coherent four-wave mixing spectroscopy of the gas phase can be also substantially improved using this approach [24 – 26].

Thus, the use of the waveguide component of dispersion is the key issue in improving the efficiency of nonlinear-optical frequency conversion and formation of ultrashort light pulses in gas-filled hollow fibres. A natural way to increase the influence of waveguide dispersion on the processes of nonlinear-optical interaction is to decrease the inner radius of the fibre [27]. Hollow fibres with smaller inner radii also allow high radiation power densities to be achieved with laser pulses of lower energies. However, hollow fibres with smaller radii are characterised by higher waveguide losses (the coefficient of optical losses for the modes of a hollow fibre is inversely proportional to the third power of the inner radius of the fibre) [28]. Therefore, most of nonlinear-optical experiments are usually performed with hollow fibres whose inner radii range from 50 up to 500 μm, which limits phase- and group-velocity-matching abilities of these fibres and prevents compensation of group-velocity dispersion effects within a broad range of physical parameters.

In this paper, we will demonstrate that the use of hollow-core fibres with a microstructure cladding in the form of a two-dimensional photonic crystal [29 – 38] allows the influence of the waveguide dispersion component to be increased by reducing the inner diameter of a hollow fibre without any noticeable increase in optical losses. Based on the comparison of the properties of hollow fibres with solid and photonic-crystal claddings, we will show that a periodically microstructured cladding permits the optical losses of waveguide modes in hollow-core fibres with small inner radii (on the order of several microns) to be radically lowered.

Analysis of spectral dependences of the refractive indices of argon and quartz demonstrates that hollow-core photonic-crystal fibres support the waveguide regime for high-order optical harmonics (within the wavelength range of 1 – 50 nm) due to the total internal reflection. In other words, within the short-wavelength range, such fibres are similar to conventional fibres, but the optical breakdown threshold for these fibres is much higher than the optical breakdown threshold characteristic of dielectric-core fibres. We will discuss also the possibilities of improving phase matching for high-order harmonic generation in higher waveguide modes of hollow-core fibres with a photonic-crystal cladding.

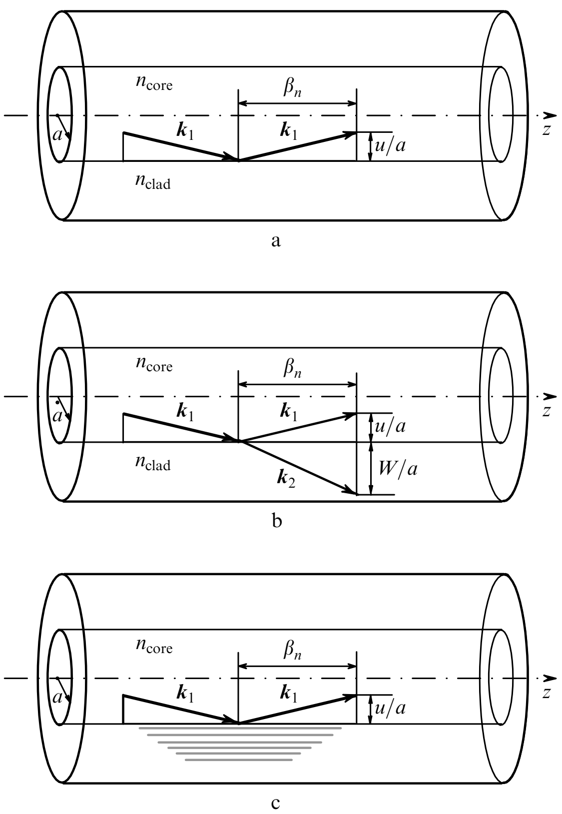

Figure 1. Waveguiding of light in optical fibres of different types. (a) A conventional fibre, which guides the light due to the total internal reflection. The propagation constant in this case meets the relations $k_1 > \beta > k_2$. (b) A hollow fibre, guiding the light in the regime of grazing incidence. The propagation constant satisfies the inequalities $k_2 > k_1 > \beta$. (c) A hollow fibre with a photonic-crystal cladding, which guides the light due to the high reflectivity of the periodically microstructured cladding within the photonic band gap.

2. Photonic-crystal cladding as a way to reduce optical losses in a hollow fibre

In this section, we will qualitatively explain the possibility of reducing optical losses in a hollow fibre by using fibres with a periodically microstructured cladding. In a general form, this possibility is illustrated in Fig. 1. As is well known [39, 40], waveguiding is achieved in conventional fibres (Fig. 1a) due to the total internal reflection from the interface between the core with the refractive index n_{core} and the cladding with the refractive index n_{clad}. The propagation constants of waveguide modes with the frequency ω under these conditions are determined from the relation $\beta = \left[k_1^2 - (u_n/a)^2\right]^{1/2}$ where $k_1 = n_{core}\omega/c, u_n$ is the eigenvalue of the characteristic equation for a waveguide mode with an index n, and a is the core radius of the fibre. The propagation constants of guided modes satisfy the inequalities $k_1 > \beta > k_2$, where $k_2 = n_{clad}\omega/c$.

In hollow fibres (Fig. 1b), the refractive index of the core n_{core} is lower than the refractive index of the cladding n_{clad}. Therefore, the propagation constants of hollow-fibre modes, $\beta = \left[k_1^2 - (u_n/a)^2\right]^{1/2} = k_2^2 - \left(W_n/a)^2\right]^{1/2}$, where W_n is the transverse wave number of the fibre cladding mode, have nonzero imaginary parts, and the propagation of light in such fibres is accompanied by radiation losses. The coefficient of optical losses in hollow fibres scales [28] as λ^2/a^3, where λ is the radiation wavelength. Such a behaviour of the magnitude of optical losses prevents us from using hollow fibres with very small inner diameters in nonlinear-optical experiments.

The idea of lowering the magnitude of optical losses in a hollow waveguide with a periodic microstructure cladding (Fig. 1c) relative to the magnitude of optical losses in a hollow waveguide with a solid cladding is based on a high reflectivity of a periodic structure within the photonic band gap [33, 41, 42]. To qualitatively illustrate this idea, we will employ the result well known from the analysis of radiation propagation in a planar waveguide with a periodic cladding [41].

The decrease in the magnitude of optical losses in a hollow planar waveguide with a periodic cladding relative to the magnitude of optical losses in a hollow planar waveguide with a solid cladding can be quantified by determining the ratio of the logarithm of the coefficient of reflection from a periodic structure to the logarithm of the coefficient of reflection from the wall of a hollow waveguide. Around the centre of the photonic band gap in the reflection spectrum of the periodic structure in the waveguide cladding with a sufficiently large number of layers N, the coefficient of optical losses in a hollow planar waveguide with a periodic cladding α_{pbg} decreases exponentially, as shown in [42], relative to the coefficient of losses in a hollow waveguide with a solid cladding $\alpha_h$with the increase in the number of modulation periods of the refractive index in the waveguide cladding:

$$\frac{\alpha_{pbg}}{\alpha_h} \propto a \exp(-2|\kappa|Nd), \qquad (1)$$

where κ is the coupling coefficient of the forward and backward waves in the periodic structure of the waveguide cladding and d is the modulation period of the refractive index in the waveguide cladding.

Thus, hollow waveguides with a periodic cladding allow optical losses characteristic of hollow-waveguide modes to be considerably reduced. Therefore, hollow fibres with a periodic cladding offer much promise for increasing the efficiency of nonlinear-optical interactions, including self- and cross-phase modulation, harmonic generation, and wave mixing. Since hollow-core photonic-crystal fibres [33] belong to this class of waveguides, we anticipate that these fibres may be very useful for enhancing nonlinear-optical interactions. However, the structure of cladding in photonic-crystal fibres is much more complicated than the structure of a periodic multilayer considered above. Therefore, a more detailed analysis of waveguide modes of photonic-crystal fibres is necessary to quantitatively characterise the magnitudes of optical losses in such fibres.

3. Optical losses of pump radiation in a hollow-core fibre: a photonic crystal versus a solid cladding

Since the optical properties of hollow fibres in the visible and near-infrared ranges, which are characteristic of Ti:sapphire-laser pump radiation, qualitatively differ from the optical properties of these fibres in the vacuum-ultraviolet and X-ray ranges, characteristic of high-order harmonics, it seems appropriate to assess the influence of waveguide optical losses separately for pump radiation and high-order harmonics. In this section, we will provide estimates for optical losses of pump radiation propagating in hollow-core fibres with a solid and a photonic-crystal cladding.

We will assume in our analysis that the coefficient of optical losses in waveguide modes of a hollow fibre are small compared to the relevant propagation constants and the radiation wavelength is much less than the core radius a of the fibre [28]:

$$\frac{\omega a}{c} \gg 1, \tag{2}$$

$$\left| \frac{\beta_n(\omega)c}{\omega n_{\text{core}}(\omega)} - 1 \right| \ll 1, \tag{3}$$

where $\beta_n(\omega)$ is the propagation constant of the waveguide mode at the frequency ω.

The propagation constant $\beta_n(\omega)$ and the attenuation coefficient $\alpha_n(\omega)$ of the EH_{1n} waveguide mode at the frequency ω are then given by the following approximate formulas [28, 40]:

$$\beta_n(\omega) \approx \frac{\omega n_{\text{core}}(\omega)}{c} \left[1 - \frac{1}{2} \left(\frac{u_n c}{a \omega n_{core}(\omega)} \right)^2 \right], \tag{4}$$

$$\alpha_n(\omega) \approx \frac{1}{a} \left(\frac{u_n c}{a \omega n_{\text{core}}(\omega)} \right)^2 \times \operatorname{Re}\left\{ \frac{\varepsilon_{\text{clad}}(\omega) + n^2_{\text{core}}(\omega)}{n_{\text{core}}(\omega)[\varepsilon_{\text{clad}}(\omega) - n^2_{\text{core}}(\omega)]^{1/2}} \right\}, \tag{5}$$

where u_n is the eigenvalue for the EH_{1n} mode and $\varepsilon_{\text{clad}}(\omega)$ is the dielectric constant of the fibre cladding.

Fig. 2 presents the results of calculations performed using Eqn (5). These calculations show that the attenuation coefficient of the fundamental EH_{11} mode of a hollow fibre with a fused silica cladding and an inner radius of 7.4 μm reaches 14 cm^{-1} for radiation with the wavelength $\lambda = 0.8$ μm, which corresponds to the attenuation length of only 0.7 mm. A hollow fibre with such a coefficient of optical losses is, of course, of no interest for nonlinear-optical experiments.

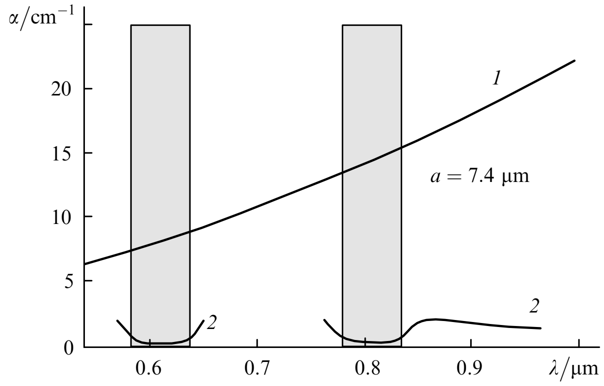

Figure 2. Attenuation coefficient α of the fundamental waveguide mode EH_{11} as a function of the radiation wavelength calculated with the use of Eqn (5) for a hollow fibre with a fused silica solid cladding (*1*) and estimated from the experimental data of [33] for a hollow-core fibre with a fused silica photonic-crystal cladding (*2*). The dashed areas show the transmission bands of the photonic-crystal hollow fibre studied in [33].

Our estimates for the coefficient of optical losses in a hollow-core fibre with a photonic-crystal cladding will be based on the experimental results obtained by the authors of paper [33], who demonstrated the existence of transmission bands for such fibres, where radiation is characterised by very low losses even when the core radius of the fibre is very small (down to several microns). The physics behind these transmission bands was discussed in the previous section. Comparison of the coefficient of optical losses calculated using Eqn (5) for a solid-cladding hollow fibre with an inner radius of 7.4 μm with the coefficient of optical losses measured in [33] for a photonic-crystal hollow fibre with the same inner radius shows (Fig. 2) that a hollow fibre with a periodically microstructured cladding allows waveguide losses of 0.8-μm pump radiation to be reduced by approximately an order of magnitude. In the following sections, we will demonstrate that photonic-crystal hollow fibres with an inner radius on the order of several microns possess expanded abilities for phase matching high-order harmonic generation.

4. Propagation of high-order harmonics in a photonic-crystal hollow fibre

In this section, we will analyse the propagation of high-order harmonics of 0.8-μm pump radiation in an argon-filled hollow fibre with a fused silica photonic-crystal cladding. To understand the qualitative differences in optical properties of a hollow fibre with a fused silica cladding in the VUV/X-ray and the visible/IR spectral ranges, we should keep in mind that the relation between the refractive indices of the core and the cladding of a fused silica hollow fibre in the short-wavelength range (Fig. 3) corresponds to a conventional (Fig. 1a) rather than a hollow-core (Fig. 1b) fibre (the spectral dependences of the refractive indices of argon and fused silica presented in Fig. 3 were calculated

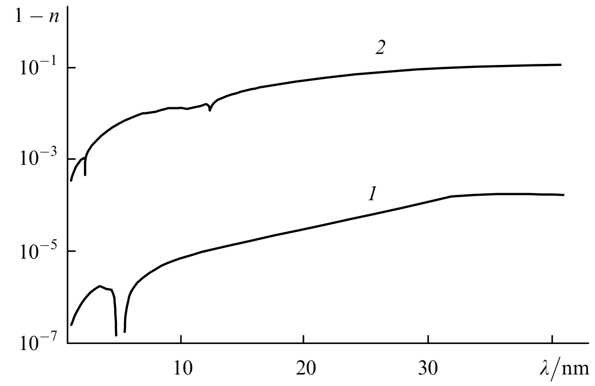

Figure 3. The refractive indices of (*1*) argon at the pressure of 1 atm and (*2*) fused silica as functions of the wavelength within the spectral range characteristic of high-order optical harmonics (based on the data from [43]).

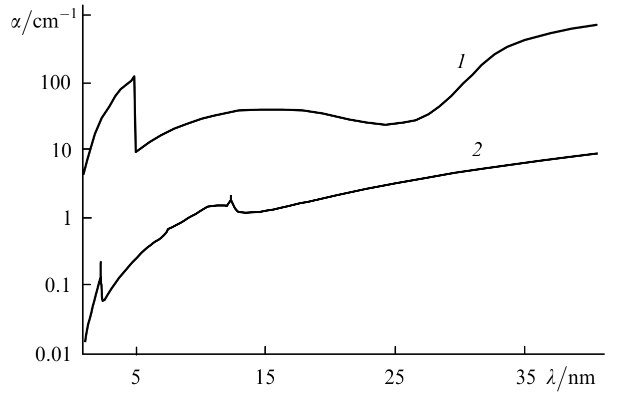

Figure 5. Spectral dependences of (*1*) the absorption coefficient of short-wavelength radiation in argon at the pressure of 1 atm and (*2*) the attenuation coefficient of the EH_{1n} waveguide mode with $n = 16$ in a hollow fibre with an inner radius of 7.4 μm and a fused silica cladding with no gas inside.

with the use of the reference data from [43]). Thus, in contrast to pump radiation, high-order harmonics are guided due to the total internal reflection.

To provide an approximate analysis of waveguiding of high-order harmonics in a photonic-crystal hollow fibre, we will neglect the structure of the cladding. To justify the use of this rough approximation for a qualitative analysis of phase matching in nonlinear-optical interactions, we estimated the penetration depth of high-order harmonics in the cladding of a fused silica hollow fibre (Fig. 4). Due to the high refractive-index step between the core and the cladding of the fibre at the frequencies of high-order optical harmonics (see Fig. 3), this penetration depth, $\sigma_{\text{clad}} = \left[\text{Re}\left(\beta_n^2 - k^2\varepsilon_{\text{clad}}\right)^{-1/2}\right]^{-1}$, is much less than the pitch of the structure in the cladding even for higher waveguide modes. In particular, for $n = 16$, the penetration depth of the EH_{1n} mode of radiation in a fused silica fibre cladding never exceeds 12 nm within the range of wavelengths from 5 to 40 nm (see Fig. 4), which is substantially less than the typical periods of the photonic-crystal structure in the fibre cladding.

Importantly, the waveguide component of optical losses for high-order harmonics is much less than the magnitude of optical losses due to the absorption of harmonics in the gas filling the fibre (see Fig. 5). Absorptive losses of high-order harmonics should be, therefore, taken into account when

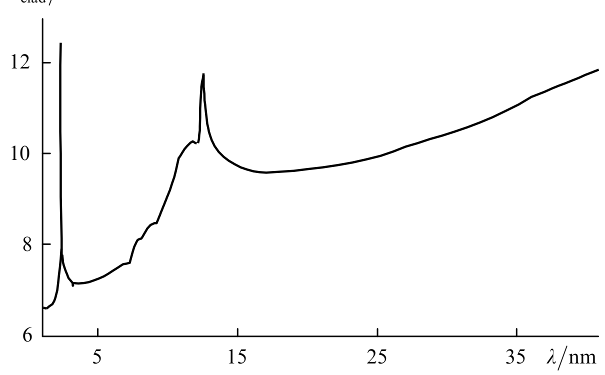

Figure 4. The penetration depth σ_{clad} of short-wavelength radiation in a fused silica cladding of a hollow fibre filled with argon at a pressure of 1 atm for the EH_{1n} waveguide mode with $n = 16$.

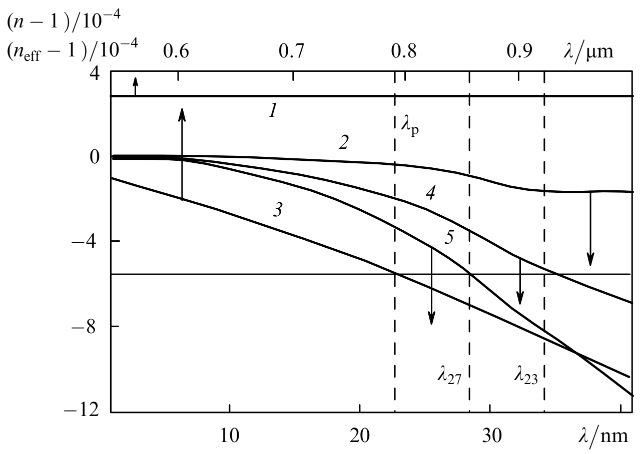

Figure 6. Spectral dependences of the refractive index of argon at the pressure of 1 atm (*1, 2*), effective refractive index for the fundamental EH_{11} mode of an argon-filled hollow fibre (*3*), and effective refractive indices for high-order waveguide modes EH_{1n} with $n = 12$ (*4*) and 16 (*5*). The upper and lower abscissa axes show the pump and harmonic wavelengths, respectively. The fibre is filled with argon with a pressure of 1 atm and has a fused silica cladding and a core radius of 7.4 μm. Positions of pump wavelength (λ_p) and optical harmonic wavelengths ($\lambda_{23}, \lambda_{27}$) are marked by dashed lines.

choosing the optimal gas pressure necessary to phase match high-order harmonic generation (see the following section).

5. Phase matching high-order harmonic generation in a photonic-crystal hollow fibre

To achieve noticeable efficiencies of harmonic generation, we have to provide phase-matching conditions. In the case of harmonic generation in optical waveguides, these conditions require the equality of propagation constants of the harmonic waveguide mode and the nonlinear polarisation induced in the gas. Ignoring nonstationary effects, giving rise to the time dependence of the phase of nonlinear polarisation (see, e.g., [22, 44]), we represent the phase mismatch $\Delta\beta$ for the process of qth-harmonic generation as

$$\Delta\beta = \beta_m(q\omega) - q\beta_n(\omega), \tag{6}$$

where $\beta_m(q\omega)$ and $\beta_n(\omega)$ are the propagation constants of the mth waveguide mode of the qth harmonic and the nth waveguide mode of pump radiation.

The phase-matching condition (6) requires the equality of the effective refractive indices $n_{\rm eff} = c\beta_n(\omega)/\omega$ at the wavelengths of pump radiation and the qth harmonic. Fig. 6 displays the results of approximate calculations of the effective refractive indices for higher waveguide modes of harmonics and the fundamental waveguide mode of pump radiation. Propagation constants of optical harmonics guided in the fibre were calculated with the use of expressions from the elementary fibre theory (see Section 2). The propagation constant of the fundamental waveguide mode of pump radiation was calculated in accordance with Eqn (4) for a hollow fibre with a solid cladding.

A similar approximation, employed earlier in paper [35], allowed the dispersion properties of waveguide modes in microstructure fibres with a fused silica core to be qualitatively understood. Within the framework of this approximation, we, in fact, restrict our consideration to grazing-incidence modes of a hollow fibre.

In the case of a planar waveguide with a periodic cladding, the use of such an approach for the estimate of propagation constants of waveguide modes has been substantiated in [42]. However, the family of modes of a photonic-crystal hollow fibre cannot be reduced, of course, to the family of modes of a hollow fibre with a solid cladding. Therefore, this approach may provide only an approximate understanding of phase-matching conditions. More accurate analysis of phase matching for high-order harmonic generation in photonic-crystal hollow fibres requires numerical simulations of the dispersion of waveguide modes in a microstructure hollow fibre, which can be carried out using the methods described in papers [45–47].

One can see from the results of calculations presented in Fig. 6 that optimisation of parameters of the gas and the fibre permits phase matching to be achieved for the harmonic-generation process in high-order EH_{1n} waveguide modes. In particular, Fig. 6 demonstrates the phase matching for the generation of the 27th optical harmonic with $n = 16$ and the generation of the 23rd harmonic with $n = 12$. The wavelength of pump radiation was set equal to 0.78 μm in these calculations. Note that a fine tuning to phase matching can be performed by smoothly changing the gas pressure.

6. Conclusions

The analysis performed in this paper shows that hollow-core fibres with a microstructure cladding in the form of a two-dimensional photonic crystal open a unique opportunity of implementing nonlinear-optical interactions of waveguide modes with transverse sizes of several microns in a gas medium. Such waveguide regimes of nonlinear-optical interactions cannot be implemented in hollow fibres with solid claddings, where the magnitude of optical losses rapidly grows with the decrease in the inner radius. In particular, the attenuation coefficient of the fundamental mode in a hollow fibre with a fused silica solid cladding and an inner radius of 7.4 μm reaches 14 cm^{-1} for radiation with a wavelength of 0.8 μm, which corresponds to the attenuation length of only 0.7 mm. Hollow fibres with a photonic-crystal cladding are characterised by much lower optical losses as compared with hollow fibres with a solid cladding because of the high reflectivity of the periodic structure of the cladding within the photonic band gap. Due to this property, hollow-core photonic-crystal fibres offer much promise for the generation of ultrashort light pulses and increasing the efficiency of nonlinear-optical interactions, including self- and cross-phase modulation, optical harmonic generation, and wave mixing.

Our qualitative analysis of the dispersion of photonic-crystal hollow fibres reveals qualitative differences in optical properties of such fibres in the VUV/X-ray and visible/IR spectral ranges. These differences stem from the fact that the relation between the refractive indices of the core and the cladding of a fused silica hollow fibre in the short-wavelength range corresponds to a conventional rather than a hollow-core fibre. Thus, in contrast to pump radiation, high-order harmonics are guided in fused silica hollow fibres due to the total internal reflection. Our analysis has shown also the possibility of improving phase matching for optical harmonic generation in high-order waveguide modes of photonic-crystal hollow fibres by optimising fibre parameters and the characteristics of the gas filling the fibre.

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