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Group-velocity-matched ultrashort-pulse synthesis in a hollow fibre filled with a Raman-active gas*

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***Abstract*. Group-velocity mismatch between the pump and probe pulses is shown to limit the minimum duration of ultrashort pulses synthesised through the generation of multiple Stokes and anti-Stokes sidebands in a Raman-active medium preliminary excited by a short laser pulse. The group delay of pulses involved in short-pulse synthesis in a Raman-active gas can be reduced by using a hollow fibre with a properly chosen inner diameter, gas pressure, and waveguide modes. The number of Stokes and anti-Stokes sidebands in the nonlinear response of a Raman-active gas can be considerably increased under these conditions, opening the way to substantially reduce the duration of light pulses synthesised using this method.**

Keywords: *ultrashort laser pulses, group velocity, Raman-active medium.*

1. Introduction

Modern laser physics has approached the femtosecond borderline, and a breakthrough to the domain of attosecond pulses may be expected in the nearest future. Sub-5-fs light pulses produced by different methods in Refs [1, 2] provide an opportunity to investigate ultrafast processes in matter with an unprecedented temporal resolution corresponding to two optical cycles [3]. Sub-10-fs light pulses have already become a routine tool of laser experiments employed for high-temporal-resolution spectroscopy [4] and generation of radiation within a broad spectral range, including the water window [5].

Several possibilities of generating subfemtosecond and attosecond pulses are currently discussed. One of the ways that leads beyond the femtosecond range is based on high-order harmonic generation in gas jets [5 – 10] and plasmas created on the surface of a solid target [11]. The spectrum of harmonics produced in an intense laser field features a plateau with approximately equal harmonic amplitudes. As shown in Refs [12 – 16], sequences of attosecond pulses can be synthesised by phase-locking these high-order harmonics. The ways to solve the phase-locking problem are now extensively discussed in the literature [12 – 16]. The methods of selection of single pulses out of pulse trains synthesised under such conditions are also being explored [12, 13]. The authors of paper [17] have pointed out the possibility of producing single attosecond pulses through harmonic generation in a gas in the field of ultrashort laser pulses (with durations less than 5 fs).

Theoretical studies and recent experiments [18 – 20] devoted to stimulated Raman scattering and the methods of phase-locking Stokes and anti-Stokes sidebands in Raman-active media have shown that superintense laser fields are, in fact, not necessary for the generation of attosecond pulses. Stokes and anti-Stokes sidebands can be produced with high efficiencies even in pump fields of moderate intensities [20].

Phase relations suitable for synthesising subfemtosecond and attosecond pulses can be provided for these sidebands upon appropriate excitation of Raman modes. However, group-velocity dispersion inherent in any gas medium and the absence of adequate methods for the detection of subfemtosecond and attosecond pulses have prevented the observation of attosecond pulses in experiments so far.

In this paper, we discuss the possibilities of ultrashort-pulse synthesis through the generation of multiple Stokes and anti-Stokes sidebands in a Raman-active medium pre-excited with a short laser pulse. This method of short-pulse generation has been proposed and experimentally implemented in Refs [21 – 24].

The main idea of this approach can be described in the following way. A short laser pulse with a duration less than the vibration cycle of a Raman-active vibration in a medium is used to pre-excite such vibrations of gas molecules. The molecular vibrations thus excited modulate a probe pulse, which enters the medium with some delay relative to the pump pulse.

Amplitude and phase relations between multiple Stokes and anti-Stokes sidebands arising as a result of this process are suitable for synthesising extremely short light pulses. Important advantages of this approach to short-pulse generation are due to the fact that the preparation of a medium with a high-power laser pulse in this case is separated in time from the generation of Stokes and anti-Stokes sidebands in the field of a probe pulse with a moderate intensity. Numerous competing processes, having a detrimental effect on the

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formation of short pulses, such as self-action of laser pulses, ionisation of a medium, and broadening and shift of Stokes and anti-Stokes sidebands, can be eliminated under these conditions.

A hollow fibre with a length of 70 – 100 cm was used in experiments [22 – 24] to increase the length of interaction of pump and probe pulses with a Raman-active gas. In this case, the group delay of the pump and probe pulses may become one of the main factors limiting the number of excited Stokes and anti-Stokes sidebands, thereby limiting the minimum duration of light pulses. The main aim of this paper is to show that a hollow fibre may serve not only to increase the interaction length, but also to reduce the influence of group-delay effects in such experiments. The group-velocity mismatch of the pump and probe pulses can be considerably decreased in a hollow fibre with an appropriate choice of the gas pressure, the inner diameter of the hollow fibre, and waveguide modes involved in the wave-mixing process. This allows the number of Stokes and anti-Stokes sidebands to be considerably increased by using longer hollow fibres, thus providing an opportunity to substantially reduce the duration of light pulses synthesised using this approach.

2. The influence of group-delay effects on the synthesis of ultrashort light pulses

We will analyse the possibilities of synthesising ultrashort pulses through the generation of multiple Stokes and anti-Stokes sidebands using the slowly varying envelope approximation. Although such an approximation is, rigorously speaking, inapplicable for a detailed description of the evolution of ultrashort light pulses, it permits some general tendencies in the evolution of the pulse and its spectrum to be understood. We will use this approximation to examine the role of group-delay effects in the synthesis of ultrashort light pulses in Raman-active gases and to illustrate the ways of reducing the group delay due to the waveguide dispersion.

Restricting our analysis to the case of EH_{1q} modes of a hollow fibre, we represent the fields of the pump and probe pulses propagating along the z axis in a hollow fibre (Fig. 1) in the following form:

$$E_1 = {}^1/_2 f^m(\rho) A(t,z) \exp[-\mathrm{i}(\omega_1 t - K_1^m z)] + \text{c.c.}, \quad (1)$$

$$E_2 = {}^1/_2 f^n(\rho) B(t,z) \exp[-\mathrm{i}(\omega_2 t - K_2^n z)] + \text{c.c.}, \quad (2)$$

where ω_1 and ω_2 are the central frequencies of the pump and probe pulses, respectively; $f^q(\rho)$ is the transverse field distribution in the EH_{1q} mode of the hollow fibre; ρ is the distance from the axis of the hollow fibre; K_1^m and K_2^n are the propagation constants of the pump and probe pulses corresponding to the waveguide modes of the hollow fibre; and $A(t,z)$ and $B(t,z)$ are the slowly varying envelopes of the pump and probe pulses, respectively.

When the pump pulse (1) is switched off, Raman-active modes Q excited in the medium by this pulse freely decay in accordance with the following expression [23]:

$$Q(t) = Q_0 \exp\left(-\frac{t}{T_2}\right) \sin\left[\left(\Omega^2 - \frac{1}{T_2^2}\right)^{1/2} t\right], \quad (3)$$

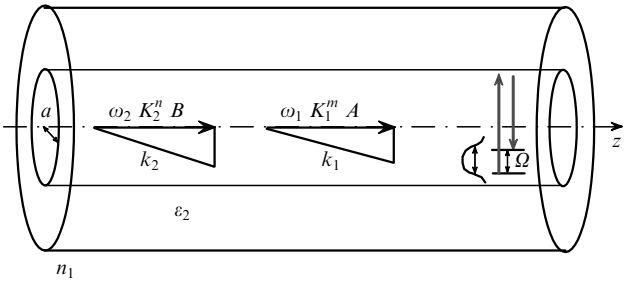

Figure 1. Propagation of light pulses in a gas-filled hollow fibre. The first pulse is used to excite Raman-active vibrations of molecules in the gas filling the fibre. The second pulse generates multiple Stokes and anti-Stokes sidebands in accordance with the approach developed in Refs [21 – 24].

where Q_0 is the amplitude of vibrations, which is proportional to the energy of the pump pulse; T_2 is the dephasing time of molecular vibrations; and Ω is the frequency of Raman-active vibrations.

The solution to the equation governing the evolution of the complex envelope of the probe pulse entering the Raman-active medium with some delay with respect to the pump pulse in the plane-wave regime under conditions when the dephasing time T_2 substantially exceeds the period of Raman vibrations and the durations of the light pulses has been found in paper [23]. In the considered case of Raman scattering occurring in waveguide modes of a hollow fibre, the solution can be written in a completely identical form:

$$B(\tau,z) = B(\tau,0) \exp\left[-\mathrm{i}\gamma \frac{\sin(\Delta K_{mn} z)}{\Delta K_{mn}} \sin(\Omega\tau + \Delta K_{mn} z)\right]. \quad (4)$$

where $\tau = t - z/v_2^n$; $B(\tau,0)$ is the envelope of the input probe pulse,

$$\Delta K_{mn} = \Omega \frac{1/v_2^n - 1/v_1^m}{2} \quad (5)$$

is the parameter characterising the mismatch of the group velocities,

$$\gamma = \frac{2\pi}{c} \omega_2 N Q_0 \frac{\partial\alpha}{\partial Q}; \quad (6)$$

N is the concentration of Raman-active molecules in the gas; and $\partial\alpha/\partial Q$ is the derivative of the molecular polarisability in the vibrational coordinate. Although we write our solution [Eqn (4)] in the form that is completely identical to the formula obtained in Ref. [23], the quantities involved in Eqn (4) are modified to include the influence of waveguide effects on the process of Raman scattering. In particular, the group-velocity mismatch involved in Eqn (4) is now defined by Eqn (5) in terms of the propagation constants related to the waveguide modes involved in the Raman process, while the coefficient γ is normalised in such a way as to include the transverse distributions of light fields in the relevant waveguide modes.

The spectrum of the probe pulse can now be represented as a superposition of equidistant spectral components $\omega_s = \omega_2 + s\Omega$ ($s = 0, \pm1, \pm2, ...$), which are separated from each other by the frequency of molecular vibrations Ω. The number of Stokes and anti-Stokes sidebands in the spectrum of the probe pulse increases with increasing propagation

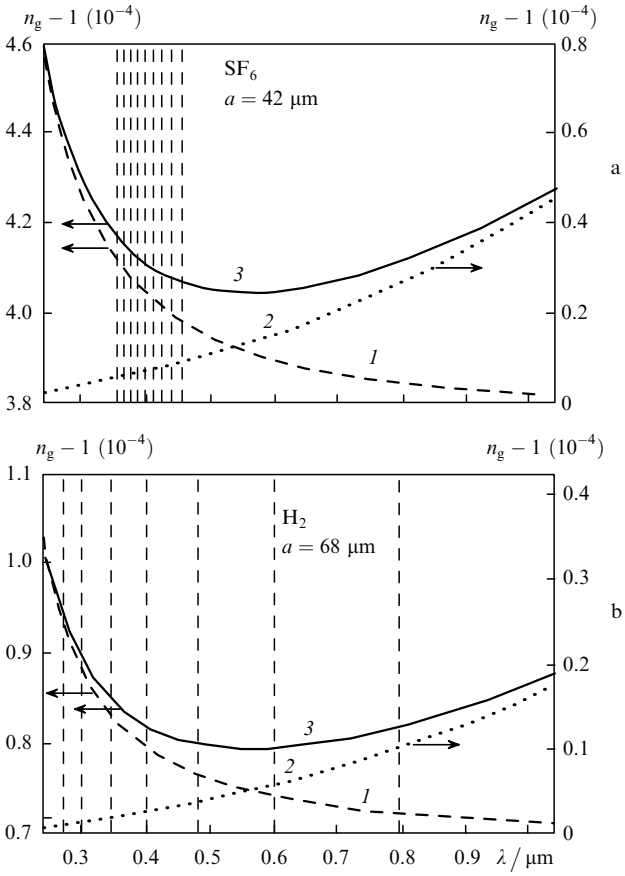

Figure 2. The group refraction index $n_g = c/v_g$ as a function of the wavelength for a gas without a fibre (based on the data from Ref. [25]) (*1*), the EH_{11} mode of an empty hollow fibre (*2*), and the EH_{11} mode of a hollow fibre filled with a gas at a pressure of 0.5 atm (*3*) in the case of SF_6, $a = 42$ μm (a) and H_2, $a = 68$ μm (b). The gas pressure is 0.5 atm. The vertical dashed lines show the wavelengths of the probe pulse (0.4 μm) and Stokes and anti-Stokes components.

Table 1.

Gas	Δk/cm^{-1}	ΔK_0/cm^{-1}	a/μm	L_a	N
SF_6	775	0.049	42	30	22
N_2	2330	0.087	54	63	8
O_2	1555	0.07	49	45	11
CO_2	1388	0.094	40	25	12
H_2	4160	0.1	68	115	4

Notation: Δk is the frequency of Raman-active vibrations (from Ref. [25]); ΔK_0 is the group-velocity mismatch for pulses of 800- and 400-nm radiation in the gas in the absence of a hollow fibre calculated for a gas pressure of 0.5 atm with the use of the data from Ref. [25]; a is the optimal inner radius of a hollow fibre allowing group-velocity matching to be achieved for pulses of 800- and 400-nm radiation in the fundamental mode of the hollow fibre filled with a gas at a pressure of 0.5 atm; L_a is the attenuation length of 800-nm radiation in the fundamental mode of a hollow fibre with the optimal inner radius a corresponding to the group-velocity matching of 800- and 400-nm radiation pulses; and $N = (2c\Delta k\tau')^{-1}$ is the minimum number of Stokes and anti-Stokes components necessary to generate a pulse shorter than 1 fs ($\tau' = 1$ fs).

length. Under these conditions, the mismatch of the group velocities of the pump and probe pulses may be a serious problem for many gases (the dashed lines in Figs 2a, 2b), restricting the interaction length to the characteristic walk-off length $l_{mn}^{g} = \pi/(2\Delta K_{mn})$, which can be understood as the length where the group delay of the pump and probe pulses becomes equal to half the period of molecular Raman-active vibrations.

In particular, for pulses of 800- and 400-nm radiation propagating in an SF_6 gas at a pressure of 0.5 atm, the group-velocity mismatch ΔK_0 in the absence of the waveguide dispersion component (Table 1) calculated using the experimental data from Ref. [25] is approximately equal in its absolute value to 0.049 cm^{-1}. This estimate shows that group-delay effects may become the main factor limiting the minimum pulse duration for the schemes synthesising subfemtosecond and attosecond pulses with the use of high-frequency Raman-active vibrations (see the data presented in Table 1 and in Fig. 2b). Two to three Stokes and anti-Stokes sidebands due to such Raman vibrations would be sufficient, as shown in Ref. [24], to synthesise a subfemtosecond light pulse.

3. The ways to reduce the group-velocity mismatch in a hollow fibre

The group-velocity mismatch under the above-specified experimental conditions can be reduced by using the dispersion of waveguide modes. Physically, this opportunity is associated with the fact that the group velocity of a light pulse propagating in a gas-filled hollow fibre (the solid lines in Fig. 2),

$$v_{pq} = \left(\frac{\partial K_{pq}}{\partial \omega}\right)^{-1}, \tag{7}$$

where K_{pq} is the propagation constant corresponding to the relevant waveguide mode of a hollow fibre with mode indices p and q, differs from the group velocity of a light pulse in the same gas, but in the absence of a waveguide (the dashed lines in Fig. 2),

$$v = \left(\frac{\partial k}{\partial \omega}\right)^{-1} = c\left[n\left(1 + \frac{\omega}{n}\frac{\partial n}{\partial \omega}\right)\right]^{-1}, \tag{8}$$

where $k = n\omega/c$ and n is the refractive index of the gas. This difference in group velocities defined by Eqns (7) and (8) is due to the fact that the wave number k in free gas differs from the propagation constant for the waveguide mode in a gas-filled hollow fibre $K_{pq} = \left(k^2 - h_{pq}^2\right)^{1/2}$, where the quantity h_{pq} can be found from the characteristic equation for the waveguide mode of a hollow fibre (the relevant wave-vector diagram is shown in Fig. 1).

In particular, the propagation constant for an EH_{1m} mode of a hollow fibre is given by Ref. [26]

$$K_l^{1m} \equiv K_l^m \approx \frac{\omega_l n_1(\omega_l)}{c}\left\{1 - \left[\frac{u_l^m c}{a\omega_l n_1(\omega_l)}\right]^2 \left[\frac{1}{2} + \frac{\mathrm{Im}\,\mu(\omega_l)}{a\omega_l}c\right]\right\}, \tag{9}$$

where u_l^m is the eigenvalue for the EH_{1m} mode; a is the inner radius of the hollow fibre;

$$\mu(\omega_l) = \frac{\varepsilon_2(\omega_l) + n_1^2(\omega_l)}{2n_1^2(\omega_l)[\varepsilon_2(\omega_l) - n_1^2(\omega_l)]^{1/2}}; \tag{10}$$

and $\varepsilon_2(\omega_l)$ is the permittivity of hollow-fibre walls at the frequency ω_l ($l = 1, 2$).

Differentiating Eqn (9), we arrive at the following expression for the group velocity of a light pulse with a transverse field distribution corresponding to the EH_{1m} mode of a hollow fibre:

$$\left(v_l^m\right)^{-1} = (v_l)^{-1}\left\{1 + \frac{1}{2}\left[\frac{u_l^m c}{a\omega_l n(\omega_l)}\right]^2\right\}, \tag{11}$$

where

$$v_l = c\left\{n(\omega_l)\left[1 + \frac{\omega_l}{n(\omega_l)}\frac{\partial n}{\partial \omega}\bigg|_{\omega_l}\right]\right\}^{-1}$$

is the group velocity of the light pulse in the gas in the absence of a hollow fibre.

The group-velocity mismatch in a gas-filled hollow fibre can be then represented as a sum of two terms:

$$\Delta K_{mn} = \Delta K_0 + \Delta K_{mn}^{\mathrm{w}}, \tag{12}$$

where ΔK_0 and $\Delta K_{mn}^{\mathrm{w}}$ are the components of the group-velocity mismatch due to the gas and waveguide dispersion, respectively.

An important conclusion that follows from the fact that the group-velocity mismatch of short light pulses propagating in a gas-filled hollow fibre can be represented as a sum of group-velocity mismatch components related to the gas and waveguide dispersion [Eqn (12)] is that the influence of group-delay effects on nonlinear-optical wave mixing in a hollow fibre can be reduced with an appropriate choice of the sort and the pressure of the gas filling the fibre, the inner radius of the fibre, and the waveguide modes involved in the nonlinear-optical process. In particular, the waveguide component of the group-velocity mismatch, as it follows from Eqns (11) and (12), is inversely proportional to the square of the inner radius of a hollow fibre, scaling as $\Delta K_{mn}^{\mathrm{w}} \propto a^{-2}$. Physically, this circumstance implies that larger group-velocity mismatches can be compensated in hollow fibres with smaller inner diameters.

Dispersion curves for the group refractive indices of SF_6 and H_2 Raman-active gases at a pressure of 0.5 atm are shown by the dashed lines in Figs. 2a and 2b. The dotted lines in the same figures represent the dispersion curves for the group indices of the EH_{11} modes of hollow fibres with inner radii of 42 μm (Fig. 2a) and 68 μm (Fig. 2b). The resulting dispersion curves of the group indices including the waveguide dispersion are shown by the solid lines in these figures. These dependences show that an appropriate choice of hollow-fibre parameters provides the compensation for the group-delay mismatch within a sufficiently broad spectral range.

Table 1 presents the values of the group-velocity mismatch ΔK_0 for light pulses of 800- and 400-nm radiation in various gases with intense Raman-active modes at a gas pressure $p = 0.5$ atm. The walk-off length for pulses with such wavelengths in an SF_6 gas at a pressure of 0.4 atm, which was employed in experiments [22–24], is approximately 40 cm under these conditions. Group-delay effects may have a considerable influence on the generation of Stokes and anti-Stokes sidebands in such a situation, imposing limitations on the duration of light pulses synthesised in this way. The group-velocity mismatch can be completely compensated, on the other hand, for light pulses of 800- and 400-nm radiation by choosing the inner radius of a hollow fibre a equal to 47 μm and using the fundamental waveguide mode to propagate each of these pulses (for comparison, the inner diameter of a hollow fibre used in experiments [22] was 250 μm). The group-velocity mismatch of the pump and probe pulses can be compensated in a similar way also for other gases (see Table 1).

4. Generation of multiple Stokes and anti-Stokes sidebands and group-delay-free synthesis of ultrashort pulses

If the group-velocity mismatch ΔK_{mn} is small, Eqn (4) for the envelope of the probe pulse can be rewritten as

$$B(\tau, z) = B(\tau, 0)\sum_{s=-\infty}^{\infty} J_s(\gamma z)\exp(-\mathrm{i}s\Omega\tau), \tag{13}$$

where $J_s(x)$ is the sth-order Bessel function.

One can see from Eqn (13), that multiple Stokes and anti-Stokes components arise in the spectrum of the probe pulse as this pulse propagates through an impulsively pre-excited Raman active gas and becomes dressed with Stokes and anti-Stokes sidebands. The number of these sidebands increases with increasing pump energy (leading to the increase in the parameter γ) and the interaction length. The minimum pulse duration that can be achieved by compensating the chirp of the pulse described by Eqn (13) is determined by the number M of Stokes and anti-Stokes components generated in the process of pulse propagation: $\tau \approx (2c\Delta k M)^{-1}$ (see also Table 1). Due to the properties of Bessel functions, the maximum value of M, in its turn, is determined by the parameter γL (where L is the length of the Raman-active medium), $M \approx \gamma L$.

Thus, large interaction lengths are crucial for synthesising ultrashort light pulses. The increase in the interaction length is limited by group-delay effects (Fig. 2) and group-velocity dispersion (Fig. 3). However, one can see from Figs 2 and 3 that, with an appropriate choice of hollow-fibre parameters, the waveguide dispersion component, reduces the group delay and group-velocity dispersion for Stokes and anti-Stokes components. This circumstance is especially important for gases with high-frequency Raman-active vibrations. In particular, in the case of molecular hydrogen at a pressure of 0.5 atm, the lengths corresponding to the group delay of Stokes components and a probe pulse with $\lambda = 400$ nm equal to half the period of molecular vibrations are estimated in the absence of a hollow fibre as 37, 21, and 16 cm for the first, second, and third Stokes components, respectively. The use of a hollow fibre with an inner radius $a = 68$ μm under these conditions (Figs 2b, 3b) would allow these characteristic walk-off lengths to be increased to 57, 47, and 5×10^3 cm for the first, second, and third Stokes components, respectively.

Thus, the compensation for the group-velocity mismatch of the pump and probe pulses due to the use of the waveguide dispersion of a hollow fibre allows the efficiency of synthesising ultrashort pulses through the generation of multiple Stokes and anti-Stokes sidebands in a Raman-active medium to be considerably improved. It should be mentioned here that the increase in the length of a hollow fibre inevitably leads to the increase in the magnitude of optical losses for leaky modes of a hollow fibre, which are always characterised by substantially nonzero attenuation coefficients (the characteristic attenuation lengths L_{a} are

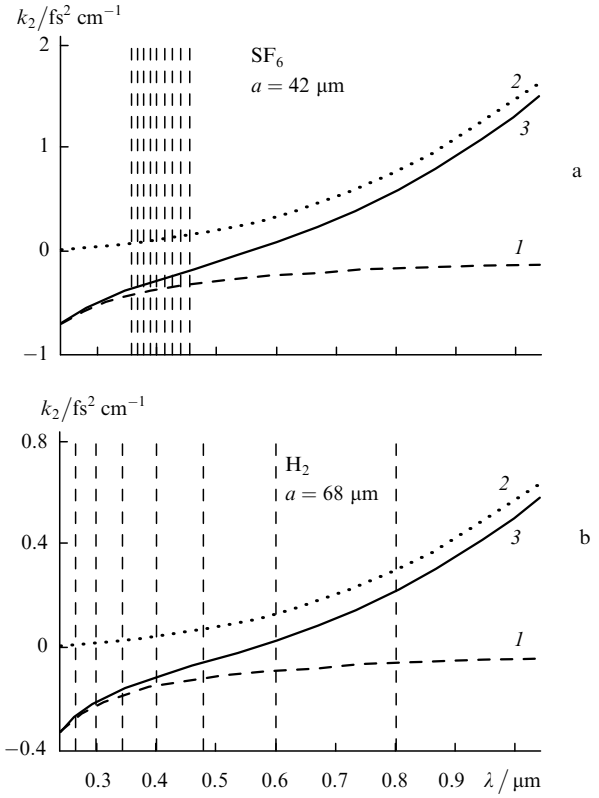

Figure 3. The group-velocity dispersion as a function of the wavelength for a gas without a fibre (based on the data from Ref. [25]) (*1*), the EH_{11} mode of an empty hollow fibre (*2*), and the EH_{11} mode of a hollow fibre filled with a gas (*3*) in the case of SF_6, $a = 42$ μm (a) and H_2, $a = 68$ μm (b). The gas pressure is 0.5 atm. The vertical dashed lines show the wavelengths of the probe pulse (0.4 μm) and Stokes and anti-Stokes components.

summarised in Table 1). One of the promising ways to solve this problem is to employ hollow-core fibres with a cladding having a structure of a two-dimensional photonic crystal—the so-called holey (or photonic-crystal) fibres [27–31]. The presence of a photonic band gap in the transmission spectrum of the cladding of such fibres permits optical losses characteristic of the leaky modes of hollow waveguides to be substantially reduced [32, 33].

5. Conclusions

Analysis performed in this paper shows that the group-velocity mismatch of the pump and probe pulses may limit the minimum duration of ultrashort pulses produced through the generation of multiple Stokes and anti-Stokes sidebands in a Raman-active medium pre-excited with a short laser pulse. The use of hollow fibres in such experiments allows the efficiency of ultrashort-pulse synthesis to be considerably improved not only due to the increase in the length of nonlinear-optical interaction of light pulses in a Raman-active gas, but also due to the possibility of reducing the group-velocity mismatch of ultrashort pulses by using the waveguide dispersion of a hollow fibre. Having represented the group-velocity mismatch of short light pulses propagating through a gas-filled hollow fibre as a sum of components related to the gas dispersion and the dispersion of waveguide modes, we demonstrated that the influence of group-delay effects on stimulated Raman scattering in an impulsively excited gas can be considerably reduced with an appropriate choice of the sort and the pressure of the gas filling the fibre, the inner diameter of the hollow fibre, and waveguide modes involved in the impulsive excitation of the medium and the Raman-scattering process. The number of Stokes and anti-Stokes sidebands can be considerably increased under these conditions, which opens the way of decreasing the minimum duration of light pulses synthesised by this method.

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