Optik 161 (2018) 156–160

Contents lists available at ScienceDirect

Optik

journal homepage: www.elsevier.de/ijleo

Original research article

Coherent Raman scattering interaction in hydrogen gas-filled hollow core photonic crystal fibres

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ARTICLE INFO

Article history:
Received 1 November 2017
Received in revised form 22 January 2018
Accepted 3 February 2018

Keywords:
Coherent Raman scattering
Gas filled HC-PCFs

ABSTRACT

In this report, we present a numerical study of the Stokes amplification in the complex transient regime in gas filled hollow-core photonic crystal fibres (HC-PCFs). The temporal and space evolution of pump, Stokes fields are described in detail. Moreover, the complex process of population inversion and coherence are also insightfully discussed.

1. Introduction

The first experiment of stimulated Raman scattering (SRS) is proved in 1962 [1], the next studies have been conducted by solving the coupled equations for the evolution of the pump and Stokes intensities to the excitation of the Raman medium [2,3]. A full quantum mechanical solution to SRS that unifies the spontaneous emission and the spatial propagation was developed in the 1980s by Raymer et al. [4] and drew a precise picture of the Stokes amplification from the quantum zero-point motion in the Raman medium.

The characteristics of the Stokes field mainly depend on the Raman gain coefficient, dephasing rate, polarization and length of the excited Raman medium, the pump pulse duration [4–6]. The Raman scattering is indentified in three regimes: A spontaneous, steady-state and transient regimes. In which, the last one is of importance in coherent generation of higher-order SRS components. It can be achieved if the pump pulse duration is long enough that the number of pump photons is sufficiently numerous to trigger a SRS process but short enough that the collisional dephasing rate has minimum effect during the generation time of the Stokes pulse [6]. As a result, the Stokes field generated in the transient regime (or coherent high regime) is expected to maintain a high degree of molecular coherence with the initial pump field. This characteristic can be used for the generation of cascaded Raman scattering for compression of femtosecond pulses in gas-filled capillaries [7,8], thus making the transient regime a good candidate to achieve attosecond (as) pulse generation. In order to achieve the transient SRS regime in gas active medium, the ps-fs ultrashort intense pulse of had been used [3,9–11].

The discovery of HC-PCFs [12] opened the study opportunity of the SRS transient regime in a very large range of pump pulse durations of fs – hundreds of ns [13] and allows us to gain deeper into different schemes in the stimulated Raman scattering such as: precise control of transient Raman scattering using buffered H2 in HC-PCFs resulting in the effective Raman gain in a gas-filled hollow-core photonic crystal fiber can actually be significantly enhanced when a buffer gas is added [13], generation of a purely vibrational Raman comb from the vacuum ultraviolet (184 nm) to the visible (478 nm) in

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https://doi.org/10.1016/j.ijleo.2018.02.009
0030-4026/

hydrogen-filled HC-PCFs by using the pressure-tunable normal dispersion system of the "fiber + gas" [14,16–20]. HC-PCF as a microcell offers an excellent guiding structure: diffraction-free long interaction length allows light to be tightly confined during its propagation while the flexibility in designing the position of the guidance band. These make it become an excellent candidate for the investigation of light interaction with gases or vapor filled into its hollow-core.

Previous works, SRS generation is mainly performed in a steady-state regime. Thus, the coherence was not considered in detail and the population inversion was neglected. This paper will discuss these effects and the temporal and space evolution in the SRS process.

2. The equations of coherent SRS interaction

We assume the seed Stokes field E_S co-propagates with the pump field E_P in the gas filled core of HC-PCF. The envelope equations of the coherent three-wave interaction in SRS are derived in Ref. [21]. In which we assumed the contribution of Stark-shift are also ignored; the gas medium is dispersionless.

$$\frac{\partial E_P}{\partial z} + \frac{1}{v_P}\frac{\partial E_P}{\partial t} = i\kappa_2\left(\frac{\omega_P v_P}{\omega_S v_S}\right)\rho_{12}E_S - \frac{\gamma_P}{2}E_P \tag{2.1}$$

$$\frac{\partial E_S}{\partial z} + \frac{1}{v_S}\frac{\partial E_S}{\partial t} = i\kappa_2\rho_{12}^* E_P - \frac{\gamma_S}{2}E_S \tag{2.2}$$

$$\frac{\partial n}{\partial t} = \frac{1}{2}i\kappa_1 E_P^* E_S \rho_{12}^* - \frac{1}{2}i\kappa_1^* E_P E_S^* \rho_{12}^* - \frac{(n-n_0)}{T_1} \tag{2.3}$$

$$\frac{\partial \rho_{12}}{\partial t} = \frac{1}{2}i\kappa_1^* n E_P E_S^* - \frac{\rho_{12}}{T_2} \tag{2.4}$$

where $\omega_{P,S}$ are the pump and Stokes frequencies; $\kappa_{2,1}$ are the Raman coupling constants; $v_{S,P}$ are the phase velocities of dispersive medium and $v_P \approx v_S \approx c$ (light velocity in vacuum) in gas medium; T_1 is called the relaxation time of population inversion; T_2 is the relaxation time of molecular coherence; n is the population inversion, n_0 is its thermal equilibrium value n_0 in a time of the order of T_1; $\rho_{12}$is the molecular coherence; $\gamma_P$and $\gamma_S$represent the linear loss of the medium for pump and Stokes frequencies, respectively.

In order to solve the above equations, we used a moving frame with retarded time $\tau = t - z/c$, t is the laboratory time and z is the propagation distance along the HC-PCF. By using moving frame coordinates, we were able to simplify this system of complex coupled partial differential equations to the ordinary differential equations depending on z or τ only [22]. Using we a midpoint numerical method, the dynamics of the SRS process are shown in Section 3.

3. Generation of coherent stokes in HC-PCF filled with H_2 gas

The calculated model is described as following: we assumed the Gaussian seed Stokes pulse E_S of 15 ns co-propagates with respect to a long pump pulse E_P of 15 ns with the molecular coherence of ρ_{12} in HC-PCF filled with H_2 gas. This pump pulse duration satisfy well the condition for highly transient SRS regime [15]. HC-PCF's linear loss of Stokes and pump waves are ultralow $\gamma_S = \gamma_P \approx 0.01$ dB/m [23]; the relaxation time of population inversion $T_1 = 1000$ ns; the relaxation time of molecular coherence $T_2 = 5$ ns; gas pressure = 1 bar filled inside the fibre's hollow core; a normalized number of molecules N = 1; $n_0 = -1$. In our study, the HC-PCFs consist of a core with the core's radius of 5 μm and length of 4 m, its transmission window only supports for pump and 1st Stokes frequencies. The propagation constants of Stokes and pump waves $\beta_S = 5.5 \times 10^6 m^{-1}$; $\beta_P = 5.9 \times 10^6 m^{-1}$ respectively. The seed amplitude is quite weak ($E_S = 0.01.E_P$). The Raman strength of $\kappa_1^* = 7.4 \times 10^{-8}\left(m^2/v^2\right)$. The orthogonal-$H_2$ gas density is 62% of the total gas density at the room temperature [16,24].

Energy exchange between the pump and Stokes fields are shown in Fig. 1. At the initial position z = 0, the pump energy is maximum (normalized to be 1); Stokes energy is zero. Then, the Stokes field is slowly amplified until the position is about 0.5 m. This period can be considered as a uniform amplification. When z > 0.5 m, the exchange process is robustly happened, in which the pump energy is almost transferred into the Stokes field and its small part is leakage by the fibre loss. At the of the fibre z = 4 m, the Stokes energy is about 76% of energy. There is an efficient generation of Stokes frequency from the pump one.

Fig. 2 shows the temporal evolution of three envelopes (pump – Stokes – Coherence –what parameter in Eqs. (2.1) ÷ (2.4) at the different positions 0.5 m, 1 m, 2 m, 2.5 m, 3 m and 4 m along the HC-PCF. The Stokes envelope's increasing with pump envelope's decreasing performs along the fibre. However, the coherence initially builds up and gets the maximum one around the position the Stokes signal to be well generated (see Fig. 3). When pump beam keep propagating, the coherence will gradually decrease with the fibre's distance increasing. This is caused by the multipeak phenomenon in SRS. The multi-peak appearance can be understood by considering the energy flow among the pump, Stokes and coherence. As Stokes and coherence fields grow, the pump field tends to be depleted (toward) down to zero and forming an energy flow back into the pump field, its phase is possible to be shifted-π. The growth of pump field causes the depletion of Stokes and coherence fields at Stokes' trailing edge [24].