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Mid-IR hollow-core silica fibre Raman lasers

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Abstract. This paper presents a brief overview of the main designs, recent advances and future prospects in the development of mid-IR hollow-core silica fibre Raman lasers.

Keywords: fibre lasers, hollow-core fibres, revolver fibre, mid-IR spectral region, stimulated Raman scattering.

1. Introduction

Fibre lasers emitting in the spectral range $3-5 \,\mu\text{m}$ are used in addressing many scientific and applied issues. Coinciding with one of the atmospheric transmission windows, this spectral range is convenient for spectroscopy and remote gas analysis [1]. It includes strong absorption lines of O–H bonds (~3 μ m), C–H bonds (3.3–3.5 μ m) and CO₂ molecules (4.2–4.3 μ m) [2], so lasers operating at these wavelengths find many applications in biomedicine, laser detection and processing of hydrocarbons and environmental monitoring. In addition, laser sources emitting in this spectral range are of interest for defence applications.

The advent of hollow-core fibres (HCFs), which offer low optical losses [3], paved the way to a new class of lasers - fibre gas lasers (FGLs) - including those emitting in the mid-IR spectral region. Such lasers are capable of combining advantages of both fibre lasers (compact design, reliability, high optical beam quality and single-mode operation) and gas lasers (wide range of lasing wavelengths, high output power and narrow emission linewidth). The gain medium of fibre gas lasers is a gas, which fills the hollow fibre core and possesses dipole-active or Raman-active transitions. The hollowcore fibre then ensures a small mode field diameter and a long interaction length of the light and gain medium. As a result, thresholds for nonlinear processes, such as stimulated Raman scattering (SRS), can be lowered by several orders of magnitude relative to nonguiding schemes [4, 5]. Moreover, hollowcore fibres may offer low optical losses even in spectral ranges where the fibre material has a strong fundamental absorption [6-9]. This fact opens up the possibility of making hollowcore silica fibre gas lasers operating at wavelengths above 3 µm [10-13].

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Received 23 October 2017 *Kvantovaya Elektronika* **47** (12) 1078–1082 (2017) Translated by O.M. Tsarev Currently, mid-IR gas-filled fibre Raman lasers are an area of rapid development. Significant advances have recently been made in this area. In particular, hollow-core silica fibre Raman lasers operating at wavelengths from 2.9 to 4.4 μ m have been demonstrated to date [10–12]. This paper presents a brief overview of recent advances in mid-IR hollow-core silica fibre Raman lasers.

In most studies, fibre gas lasers have a cavity-free, singlepass configuration [4, 14–17]. Owing to the high degree of light confinement in their core ($d \sim 5-50 \mu m$ in diameter) along the entire length of the fibre (1–10 m), active gas-filled hollow-core fibres ensure a single-pass gain sufficient for laser build-up from quantum noise. Thus, a single-pass configuration allows one to realise efficient FGLs based on both SRS [14–16] and population inversion [13]. Designing a cavity for FGLs remains a complex technical problem because there are neither fibre couplers nor analogues of fibre Bragg gratings for hollow-core fibres. Nevertheless, a few studies addressed cavity-based FGL configurations using a ring cavity made from bulk elements [13] and a Fabry–Perot cavity formed by Bragg gratings spliced to the end faces of an active hollowcore fibre [18].

Light molecular gases – light hydrogen (${}^{1}H_{2}$), deuterium (D₂), methane (CH₄) and ethane (C₂H₆) – are of interest as gain media of Raman fibre gas lasers because they have a large Raman shift (4155, 2987, 2917 and 2954 cm⁻¹, respectively). This opens up possibilities of making single-stage mid-IR Raman fibre lasers pumped by pulsed nanosecond erbium-doped fibre lasers emitting in the well-mastered spectral range around 1.5 µm.

A key component for making efficient mid-IR Raman fibre lasers is a hollow-core fibre, whose characteristics should meet certain conditions. A necessary condition for Raman lasing is that the Raman gain exceed the optical loss in the fibre. In a previous study, Bufetov and Dianov [19] introduced a quality parameter (P_f) to characterise an optical fibre as an SRS-active material:

$$P_{\rm f} = \left(\sqrt{\frac{\alpha_{\rm p}}{g_{\rm R}}}A^{\rm eff} + \sqrt{\frac{\alpha_{\rm s}}{g_{\rm R}}}A^{\rm eff}\right)^2,\tag{1}$$

where g_R is the Raman gain coefficient of the active medium; α_i (i = s or p) is the optical loss in the fibre; the subscripts p and s refer to the pump and Raman lasing wavelengths, respectively; and A^{eff} is the effective area of SRS conversion in the fibre. The physical meaning of P_f , which has the same dimensions as power (and is measured in watts), is as follows: P_f is the threshold pump power of a cw Raman laser based on the fibre under consideration placed in a high-finesse cavity. Thus, with decreasing $P_{\rm f}$, the characteristics of the fibre approach the optimal ones for the $\lambda_{\rm p} \rightarrow \lambda_{\rm s}$ SRS conversion chosen, provided the pump pulse duration is sufficiently long. It is important that $P_{\rm f}$ be substantially lower than the achievable pump power.

Let us estimate the $P_{\rm f}$ of a model fibre with a hollow-core diameter of $\sim 75 \,\mu m \, (A^{\text{eff}} \approx 2.4 \times 10^{-5} \,\text{cm}^2)$ filled with molecular hydrogen at room temperature and a pressure above 10 atm ($g_R \approx 1$ cm GW⁻¹). Assuming that such fibre may in principle have optical losses $\alpha_p \le 0.1 \text{ dB m}^{-1}$ (in the near-IR) and $\alpha_s \leq 1 \text{ dB m}^{-1}$ (in the mid-IR), we obtain for the quality parameter $P_{\rm f} \leq 100$ W. A peak pump power many times higher than this level can be reached using existing nanosecond solid-state and fibre lasers. It is worth noting however that the fabrication of hollow-core silica fibres with mid-IR losses within 1 dB m⁻¹ is a nontrivial issue because, in the wavelength range from 3 to $5 \,\mu$ m, the material absorption in silica glass rises sharply, from ~ 50 to $\sim 50\ 000\ dB\ m^{-1}$. Thus, a crucial role is played by the hollow-core fibre design, which should minimise the spatial overlap of the optical mode with the silica cladding.

Figure 1 shows various hollow-core silica fibre designs. One class of HCFs is hollow-core photonic crystal fibres (Fig. 1a), which allow low optical losses to be obtained in a relatively narrow spectral range. In particular, such fibres were demonstrated to have optical losses under 0.1 dB m⁻¹ in the wavelength range $3.1-3.7 \,\mu m$ [9]. At the same time, in the case of hollow-core photonic crystal fibres, it is impossible to obtain low optical losses in a spectral range wide enough to ensure near-IR to mid-IR SRS conversion. Another type of HCF is a hollow-core Kagome cladding fibre (Fig. 1b). Such fibres turned out to be well suited for making Raman lasers in the visible and near-IR spectral regions [4, 5, 16, 18, 23-25], where silica glass has low absorption. According to a theoretical analysis [26], the waveguiding regime in Kagome lattice fibres is ensured by only the first ring of capillaries at the core-cladding interface, whereas the other rings in the cladding (necessary for mechanically securing the first ring) lead to additional losses. Thus, a Kagome lattice cladding (Fig. 1b) has a complex, multiple-ring structure, which leads to an excessive overlap of the optical field with the cladding material. As a result, hollow-core Kagome cladding silica fibres are not optimal for making efficient mid-IR Raman lasers.

In a number of studies [6-8], new HCF designs were proposed, with a simplified cladding structure (Figs 1c-1e). Special mention should be given to a report by Pryamikov et al. [6], who were the first to introduce the concept of a negative curvature of the core-cladding interface and assess its

ability to reduce the optical loss in the HCF. In addition, they proposed a so-called revolver fibre design (Fig. 1c) [6] and demonstrated that hollow-core silica fibres of this type offer good transmission at wavelengths of up to ~4 μ m. Similar results were obtained for fibres whose cladding consisted of elements in the shape of ice cream cones or parachutes (Fig. 1d) [7]. Modifying the hollow-core revolver fibres (Fig. 1e) ensured an even higher degree of light confinement in the hollow core and good transmission in hollow-core silica fibres at wavelengths of up to ~8 μ m [8]. Thus, hollow-core revolver fibres with a cladding formed by a single ring of non-contacting capillaries are the most attractive fibres for mid-IR Raman fibre gas lasers.

This year, using silica revolver fibres, we have demonstrated the first mid-IR Raman fibre gas lasers [10–12, 22, 27]. For pumping, we used a pulsed nanosecond erbium-doped fibre laser emitting at ~1.56 μ m. The pump pulse duration was 3.5 ns, the peak power reached 34 kW, and the average output power was up to 3 W. At a peak pump power of ~20 kW and spectral resolution of 0.02 nm, the measured laser linewidth was 0.065 nm (9 GHz). Although at the present stage the pump laser beam was coupled into the HCF using lenses, an all-fibre Raman laser design is possible.

In previous work [10, 22], use was made of a silica revolver fibre whose theoretical transmission spectrum is shown in Fig. 2. The fibre length was 15 m and the mode field diameter was 45 μ m. Filling its hollow core with D₂ molecular deuterium (partial pressure of 28 atm) containing ¹H₂ molecular light hydrogen impurities (partial pressure of 2 atm) made it possible to obtain Raman lasing at wavelengths of 2.9, 3.3 and $3.5 \,\mu m$ (Fig. 3). The peak power of the strongest spectral components in the mid-IR spectral region was 0.25 kW at λ = 2.9 μ m and 0.37 kW at λ = 3.5 μ m, which corresponded to an average power of 23 and 37 mW, respectively. Conversion quantum efficiency was 10% (at $\lambda = 2.9 \ \mu m$) and 6% (at $\lambda =$ 3.5 μ m), with the possibility of further optimisation. Note that adjusting the ¹H₂ and D₂ partial pressures and pump power enabled predominant lasing at a wavelength of 2.9 or $3.5 \,\mu\text{m}$ to be obtained.

In a number of studies [11, 12, 27], a silica revolver fibre was used whose design was modified so as to shift its transmission spectrum to the $\sim 4 \,\mu m$ range (Fig. 4). The mode field diameter was 56 μm . Filling the hollow core with $^{1}H_{2}$ molecular hydrogen at a pressure of 30 atm, we demonstrated the first Raman lasing at a wavelength of 4.4 μm (Fig. 5) [11, 12]. Using the single-mode output of the Raman laser, we measured the loss in the revolver fibre at this wavelength, which was found to be 1.13 dB m⁻¹, in good agreement with numeri-



Figure 1. Cross-sectional electron-microscopic images of hollow-core optical fibres; (a) hexagonal cladding [20] and (b) Kagome lattice [21] photonic crystal fibres; simplified fibre designs with a negative curvature of the core–cladding interface: (c) revolver fibre with contacting capillaries [6], (d) fibre with parachute-shaped cladding elements [7] and (e) revolver fibre with noncontacting capillaries [22] (configuration proposed by Kolyadin et al. [8]).



Figure 2. Optical loss spectrum of a hollow-core fibre designed for FGLs operating in the range $2.9-3.5 \,\mu m$ [10, 22].



Figure 3. Output emission spectrum of a Raman fibre gas laser [22] with a gain medium consisting of a mixture of molecular deuterium and light hydrogen at room temperature and partial pressures of 28 and 2 atm, respectively. The peak pump power at a wavelength of 1.56 μ m was 14 kW.

cal simulation results (0.92 dB m⁻¹) [27]. For comparison, note that the material absorption in silica glass at this wavelength is ~4000 dB m⁻¹. The use of a 15-m length of hollow-core revolver fibre ensured Raman lasing with a quantum efficiency of ~15%, and the average power at a wavelength of 4.4 μ m was 30 mW [12].

Raman lasing at a wavelength of 4.4 µm was analysed theoretically by numerically solving a system of coupled wave equations for vibrational SRS in ${}^{1}\text{H}_{2}$ molecular hydrogen. In doing so, use was made of measured optical losses of 0.04 and 1.13 dB m⁻¹ at wavelengths of 1.56 and 4.4 µm, respectively (Fig. 4), and of the Raman gain coefficient $g_{\text{R}} =$ 0.43 cm GW⁻¹ calculated for 1.56 \rightarrow 4.4 µm wavelength conversion using available data on the linewidth and scattering cross section of the Q(1) vibrational transition of ${}^{1}\text{H}_{2}$ molecules [28–30]. The theoretically evaluated optimal Raman laser length is ~3.5 m, which is substantially shorter than the hollow-core fibre length used in the experiments (15 m).

One interesting result reported by Astapovich et al. [27] is the possibility of maintaining steady-state SRS when fibre gas lasers are pumped by nanosecond pulses. It is known [28]



Figure 4. Calculated optical loss spectrum of a hollow-core revolver fibre (dashed lines) [12], near-IR loss spectrum measured using a supercontinuum source (solid line) and optical losses measured at wavelengths of 1.56 and 4.4 μ m using narrow-band laser sources [27] (asterisks).



Figure 5. Output emission spectrum of an HCF filled with ${}^{1}H_{2}$ light hydrogen at room temperature and a pressure of 30 atm. The launched peak pump power is 18 kW [11, 12]. In addition to unabsorbed pump light (1.57 µm), there are rotational (1.72 µm) and vibrational (4.41 µm) components.

that, if the pump pulse duration (τ_p) and the transverse relaxation time of optical phonons (T_2) meet the relationship $\tau_p \leq$ $20T_2$, SRS conversion is a transient process, in which the Raman gain coefficient decreases. For the Q(1) vibrational transition of molecular hydrogen at room temperature and a pressure of ~10 atm, the transverse relaxation time is $T_2 \approx$ 0.64 ns and, as a consequence, pump pulses lasting a few nanoseconds should lead to transient Raman lasing. At the same time, T_2 can readily be controlled by varying the gas pressure, because the collision frequency of molecules rises with increasing pressure, which leads to more frequent changes in the phase of molecular vibrations and, hence, to a reduction in T_2 . This effect was observed by Astapovich et al. [27] as the hydrogen pressure was varied in the range from 10 to 70 atm and ensured an increase in the output power of a Raman laser (Fig. 6) pumped by 3.5-ns pulses.



Figure 6. Experimentally determined output pulse energy at 4.4 μ m as a function of light hydrogen pressure in the hollow core [27]. The pump pulse duration is $\tau_p = 3.5$ ns. The vertical dashed line represents a pressure of 38 atm, at which the transverse relaxation time of molecular hydrogen vibrations is $T_2 = 0.175$ ns, thus meeting the relation $\tau_p = 20T_2$.

Optimising the fibre length and the hydrogen pressure in the hollow core, we were able to demonstrate Raman generation of nanosecond pulses at a wavelength of 4.4 μ m with an average power of ~250 mW and quantum efficiency of 36%. In this process, the rotational components at wavelengths of 1.72 and 1.91 μ m were significantly suppressed (Fig. 7) [27].



Figure 7. Calculated (lines) and measured (data points) average output power of spectral components as a function of average launched pump power for the Raman fibre gas laser. The solid line represents numerical simulation results for the unabsorbed pump power at $\lambda_p = 1.56 \,\mu\text{m}$ and the dashed line represents the Stokes power at $\lambda_s = 4.4 \,\mu\text{m}$. The measurements and calculations were made at the optimal fibre length (3.2 m) and hydrogen pressure (50 atm) [27].

In discussing the factors that limited the efficiency of the Raman laser to $\sim 36\%$, it should be noted that the experimentally determined output power at a wavelength of 4.4 µm essentially coincides with results of theoretical calculations made with allowance for the optical loss in the fibre (Fig. 7). This fact means that, at the present stage, the efficiency of the Raman laser is limited by the level of optical losses at the Stokes wavelength ($\sim 1 \text{ dB m}^{-1}$). At the same time, it is seen in

Fig. 4 that the level of losses at wavelengths under 4 μ m is an order of magnitude lower (0.1–0.2 dB m⁻¹). This spectral range is suitable for making more efficient Raman fibre gas lasers based on the already existing silica revolver fibres. The rise in optical losses at wavelengths above 4 μ m is caused by the sharp increase in material absorption in silica glass (as well as by the reduction in its refractive index). New solutions, capable of further minimising the overlap of the optical mode field with the silica cladding, are needed to reduce the optical loss in this spectral region. Note also that the use of higher peak power pump lasers may improve the efficiency of the mid-IR Raman fibre gas lasers because the effect of optical losses can then be reduced by using shorter lengths of hollow-core fibres.

To date, the peak power of pulsed nanosecond Raman fibre gas lasers emitting in the range $3-5 \,\mu$ m has been demonstrated to reach ~2 kW [27]. This parameter is limited not by any characteristics of hollow-core revolver fibres but by the pump power achievable with solid-state erbium-doped fibre lasers. Recent work [31, 32] has demonstrated Raman fibre gas lasers with a peak output power of 400 and 150 kW at wavelengths of 1.55 and 1.9 μ m, respectively. The use of such lasers as pump sources for gas-filled hollow-core silica fibres paves the way to efficient Raman fibre gas lasers generating nanosecond pulses with a peak power of ~100 kW in the spectral range $3-5 \,\mu$ m. Moreover, such mid-IR lasers can be realised due to two-stage SRS in a given revolver fibre segment filled with one or a few gases.

The near-IR ($\lambda = 1.56 \,\mu$ m) to mid-IR ($\lambda = 3-5 \,\mu$ m) SRS conversion is known to be accompanied by a large quantum defect, which may hinder obtaining a high average power at the Stokes wavelength. At the same time, in a recent study concerned with fibre gas lasers based on population inversion, Xu et al. [17] demonstrated efficient lasing at a wavelength of 3.1 µm under pumping at $\lambda_p = 1.53 \,\mu$ m. Despite the large quantum defect, comparable to that in Raman lasers, they reached a cw output power above 1 W. This result suggests the possibility of high average power of fibre gas lasers, including Raman lasers, in the mid-IR spectral region.

In conclusion, it should be noted that mid-IR optical fibres, which can be produced using the widespread, mature silica glass processing technology, have opened up new possibilities for utilising the mid-IR spectral region. In addition, the simple design and easy fabrication of hollow-core revolver fibres allow many research groups to participate in the development of such fibres and related lasers. The recent advances in lasing at wavelengths between 3 and 5 μ m clearly demonstrate that Raman fibre gas lasers can be efficient mid-IR laser sources. Moreover, they have great potential for raising the average and peak output powers and will no doubt find a variety of applications in biomedicine, gas analysis and materials processing.

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