Optical Engineering

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Vincent Fortin
Martin Bernier
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Dominic Faucher
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Younès Messaddeq
Réal Vallée

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Vincent Fortin
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Mohammed El Amraoui
Younès Messaddeq
Réal Vallée
Université Laval
Center for Optics, Photonics, and Lasers
Québec G1V 0A6, Canada
E-mail: vincent.fortin.5@ulaval.ca

Abstract. A growing number of applications are calling for compact laser sources operating in the mid-infrared spectral region. A review of our recent work on monolithic fiber lasers (FL) based either on the use of rare-earth fluoride fibers or on Raman gain in both fluoride and chalcogenide glass fibers is presented. Accordingly, an erbium-doped double clad fluoride glass all-FL operating in the vicinity of 3 μm is shown. In addition, we present recent results on the first demonstrations of both fluoride and chalcogenide Raman fiber lasers operating at 2.23 and 3.34 μm, respectively. It is shown that based on this approach, monolithic FLs could be developed to cover the whole 2 to 4 μm spectral band. [DOI: 10.1117/1.OE.52.5.054202]

Subject terms: fiber laser; mid-infrared; fluoride fiber; chalcogenide fiber; Raman scattering; fiber Bragg gratings.

Paper 130339P received Mar. 5, 2013; revised manuscript received Apr. 26, 2013; accepted for publication Apr. 29, 2013; published online May 29, 2013.

1 Introduction

In the last decade, significant progress has been made in the area of mid-infrared (IR) laser sources. Solid state lasers such as Ho:YAG and Er:YAG/YSGG are now commercially available and offer high power levels near 2.1 μm and 2.8 to 2.9 μm, respectively.[1–3] In addition, Cr^{2+}:ZnS/Se and Fe^{2+}:ZnSe-doped crystals can be pumped by well developed sources, i.e., fiber and solid state lasers, to deliver a broad range of long emission wavelengths between 2 and 5 μm.[4] Quantum cascade lasers are also good candidates for mid-IR laser emission in the 3.5 to 10 μm spectral region. However, the heat management of these devices is currently the main challenge that limits their power scaling.[5]

Fiber lasers (FL) present several advantageous features over other technologies: a great compactness, good beam quality, efficiency and ruggedness. Nevertheless, aside from Tm- and Ho-doped FLs emitting near 2 μm,[4,6] the number of commercial FLs having a wavelength over 2.1 μm is still limited not to say nonexistent. This limit actually corresponds to the long wavelength edge of silicate fibers transparency window. There is thus a growing need for developing FLs for the mid-IR based on other glass materials.

1.1 *Mid-IR Laser Applications*

Compact laser sources operating beyond 2 μm would benefit diverse applications. With respect to spectroscopic applications, the mid-IR contains the spectral signature of many molecules, so much that this spectral range is often labeled the molecular fingerprint region. Polymer processing applications also have a need for specific mid-IR lasers sources as most common polymers, i.e., polypropylene, poly(methyl methacrylate) (PMMA), polyvinyl chloride (PVC), acrylonitrile butadiene styrene (ABS), polycarbonate, etc., share strong absorption bands at wavelengths longer than 3 μm, notably at 3.4, 5.7, and 10.4 μm. In addition, it is well known that biomedical applications could benefit from laser sources in the vicinity of 2.94 μm, corresponding to the stretching vibration of the OH bond. Such lasers are *de facto* an ideal tool for fast and precise biological tissue ablation.[7] Mid-IR laser sources emitting inside one of the transparency windows of the atmosphere, notably 2.0 to 2.4 μm and 3.5 to 4.0 μm, also have promising applications for defense and security. Alternatively, these sources could also have a significant impact on the development of free space telecommunication systems.

1.2 *Doped FLs and Raman Fiber Lasers*

Rare-earth doped fibers possess several emission bands that can be harnessed for 2 to 4 μm laser emission. However, several of these bands cannot be used for an efficient, high power laser cavity. The most successful ones are the Tm^{3+} ($^3H_4 \rightarrow {}^3H_6$) laser transition,[8] the Er^{3+} ($^4I_{11/2} \rightarrow {}^4I_{13/2}$) transition[9,10] and the Ho^{3+} ($^5I_6 \rightarrow {}^5I_7$) transition.[11,12] They cover respectively the 1.9 to 2.1, the 2.7 to 3.0, and the 2.8 to 3.05 μm spectral ranges. Emission wavelengths of 3.22 μm (Ho^{3+}), 3.45 μm (Er^{3+}), and 3.95 μm (Ho^{3+}) have also been achieved in a ZBLAN glass host.[13–15] However, their performances were rather deceiving and the last one even required cryogenic cooling.

It is clear that many wavelengths simply cannot be accessed using doped FLs alone. On the other hand, Raman fiber lasers (RFL) can be made to operate on virtually any emission wavelength when they are matched with an adequate pump. In fact, using the combination of Tm-doped FLs and Er-doped FLs with Raman gain FLs, the entire spectrum from 2 to 4 μm can be covered, as shown in Fig. 1.

2 FL Components

2.1 *Mid-IR Optical Fibers*

Several key components are required to build an efficient mid-IR laser; highest quality fiber is the most critical one.

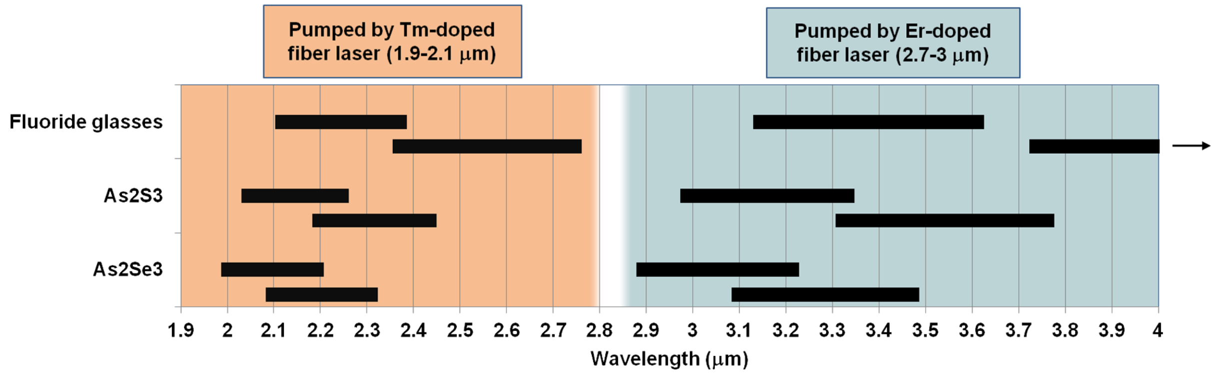

Fig. 1 Potential spectral coverage of RFLs based on either fluoride or chalcogenide fibers pumped by either Tm-doped or Er-doped FLs.

An overview of the most common mid-IR optical fiber glasses is presented in Table 1. To date, fluorozirconate glass fibers are the most technologically advanced of the nonoxide fibers available but chalcogenide-based fibers are rapidly catching up. To exhibit a good mid-IR transparency, the glass must have low phonon energy combined with ultra-low impurity content. A low attenuation coefficient is especially important for RFLs where the Stokes signal builds up over several round trips and where long fibers are often preferred. The chosen fiber must also be free of geometrical imperfections like inclusions or bubbles to sustain high power levels. In addition, the fabrication process must be mastered to produce special fiber designs such as double clad fibers for doped FLs and high numerical aperture (NA) fibers for RFLs.

2.2 *Fiber Bragg Gratings in Nonoxide Glass Fibers*

Fiber Bragg gratings (FBG) are key fiber components, providing spectral selectivity as well as mechanical stability to optical devices by avoiding the use of bulk components. Now, the inscription of FBGs through a standard UV photosensitive process has long been restricted to silica glass optical fibers. The first demonstration of FBG writing in a fluoride glass was reported in 2007 based on the use of a femtosecond pulse train at 800 nm and a phase mask.[20] More recently, the fs-writing method was successfully applied to chalcogenide fibers.[18] It was also shown that because of their large photosensitivity, inscription of FBGs in chalcogenide fibers could be performed directly through their polymer jacket. Additionally, FBG background losses associated with the fs-writing process can be minimized so as to allow them to handle high power levels. Accordingly, thermal annealing is preferably performed after inscription in order to lower their loss and to stabilize their spectral response.

2.3 *Fiber Splicing*

Fiber splices are also basic components of monolithic FLs. They are used for instance to connect FBGs to the active fiber, to launch the pump into the laser cavity, and to add end-caps on the output fiber tip. Low loss junctions need to be developed to limit intracavity losses and prevent unnecessary heating of the polymer jacket. Based on the use of filament-type optical fiber fusion splicer,[21] losses as low as 0.2 dB can be achieved between mode-matched undoped and doped single-mode fluoride fibers and losses below 0.1 dB for two identical, undoped single-mode fibers. Examples of these splices are shown in Fig. 2.

3 ER-Doped Fluoride Glass FL

Er-doped fluoride FLs are the most efficient doped FLs to produce coherent radiation around 3 μm. Readily available laser diodes at 976 nm can be used to pump the Er^{3+} ions up to level $^4I_{11/2}$, the upper level of the laser transition. Still, this

Table 1 Comparison of different mid-IR optical fiber glasses.

Glass type	Composition (nominal)	Minimum loss achieved (dB/km)	Approx. mid-IR transparency (at 1 dB/m)	Raman shift (cm^{-1})	Peak Raman gain at 3 μm (10^{-14} m/W)
Fluoroindate	BaF_2, ZnF_2, InF_3, SrF_2, YF_3, GaF_3	<100 (at 3.2 μm)	5.2 μm	509 (Ref. 16)	—
Fluoroaluminate	AlF_3, BaF_2, CaF_2, NaF, SrF_2, YF_3, ZrF_4	<100 (at 2.5 μm)	4 μm	—	—
Fluorozirconate	ZrF_4, HfF_4, LaF_3, BaF_2, NaF, AlF_3	~1 (at 2.5 μm)	4.2 μm	572 (Ref. 17)	2.2 (Ref. 17)
As_2S_3	As_2S_3	95 (at 3.4 μm) (Ref. 18)	6.5 μm	340 (Ref. 19)	125 (Ref. 19)
As_2Se_3	As_2Se_3	~200 (at 5 μm)	9.5 μm	230 (Ref. 19)	890 (Ref. 19)

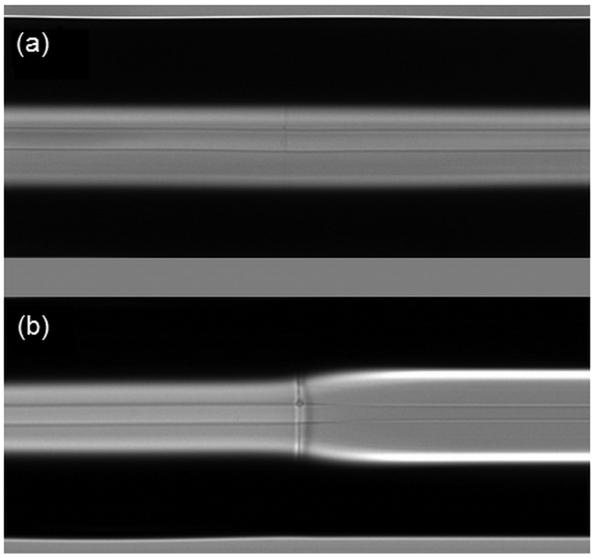

Fig. 2 (a) Splice between two single-mode undoped fluoride fibers. (b) Splice between a single-mode undoped fiber and a doped (Er^{3+}) fluoride fiber.

laser transition is self-terminating and has to be assisted with additional energy transfer processes to reach its full potential. Several strategies proved to be successful in the past. In fact, by using heavily Er-doped fibers (up to almost 10 mol%), the various energy transfer upconversion processes are enhanced, which ultimately remove the population bottleneck and lead to pump energy recycling followed by a notable increase in lasing efficiency at 3 μm.[22]

3.1 *Laser Setup*

The experimental set-up is shown in Fig. 3. The fiber used in our experiment was a 7 mol% singly Er-doped fluoride fiber provided by Le Verre Fluoré. The active fiber length ranged between 5 and 12 m, depending on the desired output power and wavelength. The truncated circular cladding had two parallel flats with a short/long diameter of 240/260 μm and a >0.46 NA. The 16 μm diameter and 0.12 NA fiber core ensured single-mode operation of the FL. A custom designed splice was used to deliver the pump from the 980 nm module, pigtailed with a multimode silica fiber, to an undoped fluoride glass fiber where the input highly reflective (HR) FBG was written. Then, a second splice was made to link the undoped fluoride fiber to the Er-doped fluoride fiber. The low reflectivity output FBG was written directly inside the doped fiber. The end of the fiber was capped to prevent premature photo-degradation of the fiber tip.

The broad emission band of Er^{3+} ions inside fluoride fibers, shown in Fig. 4, made it possible to tune the laser wavelength over essentially 400 nm, i.e., from 2600 to 3000 nm. Accordingly, laser operation was demonstrated up to 3005 nm, even if the emission cross section was extremely low when compared with the peak emission cross-section around 2.72 μm. To force laser operation at such an extreme edge of the erbium emission spectrum, a high finesse cavity provided by a pair of high reflectivity FBGs was used.[23]

3.2 *Results*

Several Er:ZBLAN laser cavities have been assembled over the last few years with emission wavelengths of 2825 nm,[9] 2940 nm,[10] and 3005 nm.[23] The highest output power achieved in this study—and ever reported to date—is 27 W at 2825 nm, shown in Fig. 5. This particular laser also displayed a high stability level of <0.3% root mean square (rms) when operated for several minutes at high power. The efficiency of these laser cavities was typically around 30% with respect to the absorbed pump power. By using FBG-based cavities, the center wavelength of the spectrum was very stable with an almost negligible spectral shift when transitioning from low to high power levels.

We have also demonstrated that it is possible to operate such Er:ZBLAN FLs in quasi-cw mode (QCW) by modulating the current of the pump diodes.[24] In this regime, the

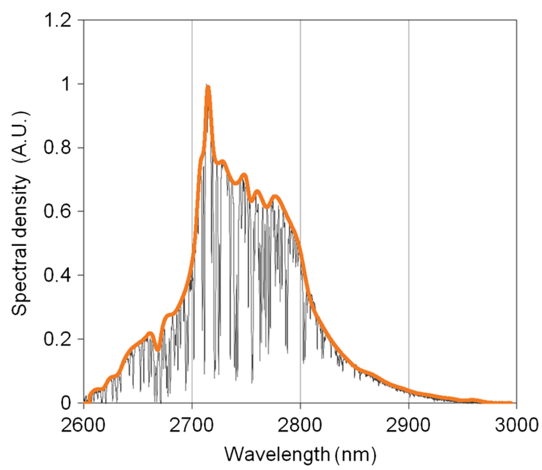

Fig. 4 Measured fluorescence spectrum of erbium ions near 3 μm (as transmitted at the fiber end).

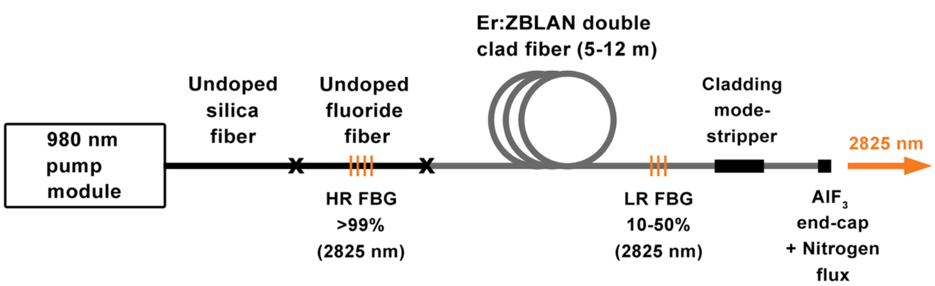

Fig. 3 Typical monolithic Er-doped fluoride glass FL setup for operation around 3 μm.

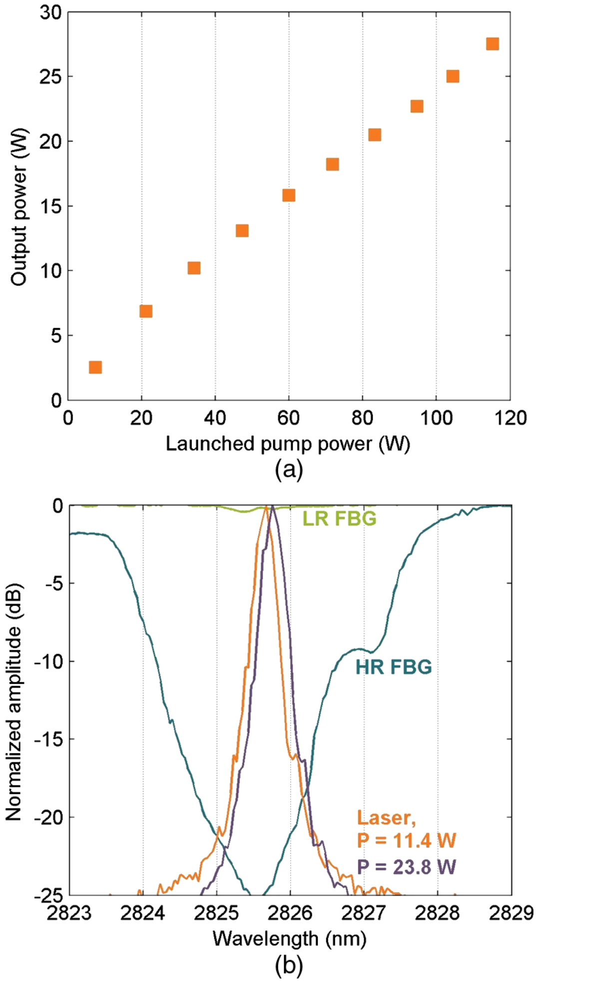

Fig. 5 (a) Output power of a high power Er-doped fluoride FL. (b) Transmission spectra of the FBGs and spectra of the laser at two power levels.

repetition rate can be adjusted from dc to 100 Hz with pulse durations down to 1 ms.

3.3 *Thermal Management and Fiber Tip Degradation*

The thermal load of every component has been carefully managed to warrant long-term operation, i.e., over 1000 h demonstrated, of the Er-doped FL at 2.83 μm. First, the doped fiber was passively cooled on an aluminum mandrel whereas the FBGs and splices were glued to aluminum V-grooves using re-coating polymers specially engineered for their low absorption near 3 μm. During the assembling process, the temperature was also monitored in real-time using a high resolution thermal imaging camera. Finally, among the various hurdles inherent to laser operation around 3 μm, the problem of fluoride fiber tip photo-degradation had to be specifically addressed.

It is well known that OH contaminants absorb laser radiation around 3 μm, which causes local heating. A recent study on the degradation of the fiber tip showed that it is possible to predict its maximum lifetime based on Fick's laws.[25] In fact, the heating of the fiber leads to an increase of the OH diffusion process, which then increases the temperature even more and ultimately leads to the fiber tip destruction, as shown in Fig. 6. It is possible to slow down this phenomenon by adding an end-cap at the output of the fiber or by blowing a flux of nitrogen to lower the presence of moisture in the atmosphere. Moreover, the end-cap can be made of materials that are more resistant than fluorozirconate glasses to this OH diffusion process. Both AlF_3 or lead germanate based glasses are generally good options. Note that it is not always possible to completely prevent this effect from occurring when high powers are involved. In fact, it was shown that fiber tip lifetime was inversely proportional to the square of the incident 2.83 μm radiation power.[25]

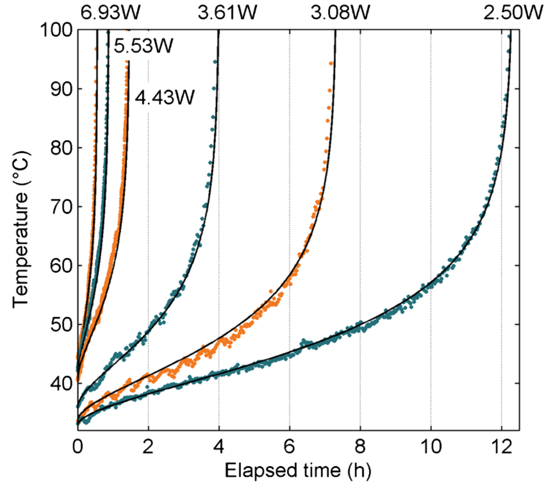

Fig. 6 Fluorozirconate fiber tip temperature versus time at different 2.83 μm radiation incident powers.

4 Fluoride Glass RFL

Fluorozirconate glasses benefit from an unrivaled low attenuation between 2.1 and 2.7 μm. This attribute makes them an ideal choice for a Raman gain laser cavity. In this section, we review our work on a nested cavity fluoride glass RFL, emitting at 2132 nm and delivering a multi-watt power level.[26]

4.1 *Laser Setup*

Figure 7 shows the experimental setup of the nested cavity fluoride RFL. It basically consists of a Tm:silica double clad fiber butt-coupled to an undoped single-mode fluoride glass fiber (6.7 μm, 0.23 NA core). A 791 nm laser diode module is used to pump the Tm-doped fiber. A first cavity, acting as the Raman pump, oscillates at 1981 nm and is bounded by a chirped HR FBG written in a mode-matched double clad undoped silica fiber and by another HR FBG at the end of the fluoride fiber. The Raman laser cavity is located inside the fluoride fiber and is completely enclosed within the pump cavity. Two additional FBGs with respective reflectivities of >99% and 93% were written to form this first order Raman cavity at 2231.4 nm. The use of concurrent pump and Raman cavities lowers the laser threshold due to higher pump intensity circulating inside the fiber.

4.2 *Results*

As shown in Fig. 8, a launched pump power threshold of 8 W was observed with a maximum Raman FL slope efficiency of 15% with respect to the laser diode power at 791 nm. The maximum Stokes output power recorded was 3.7 W. One

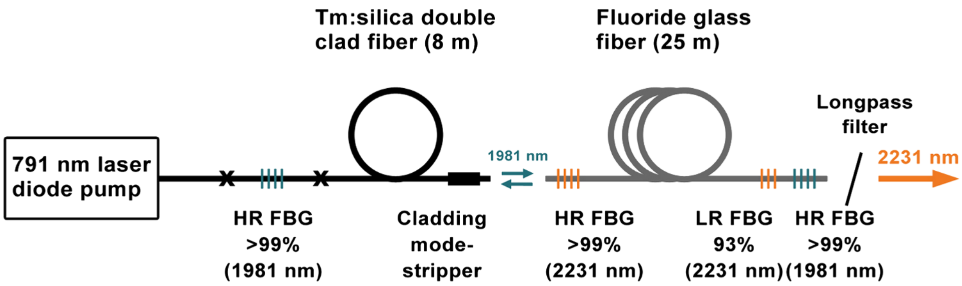

Fig. 7 Nested cavity fluoride glass RFL.

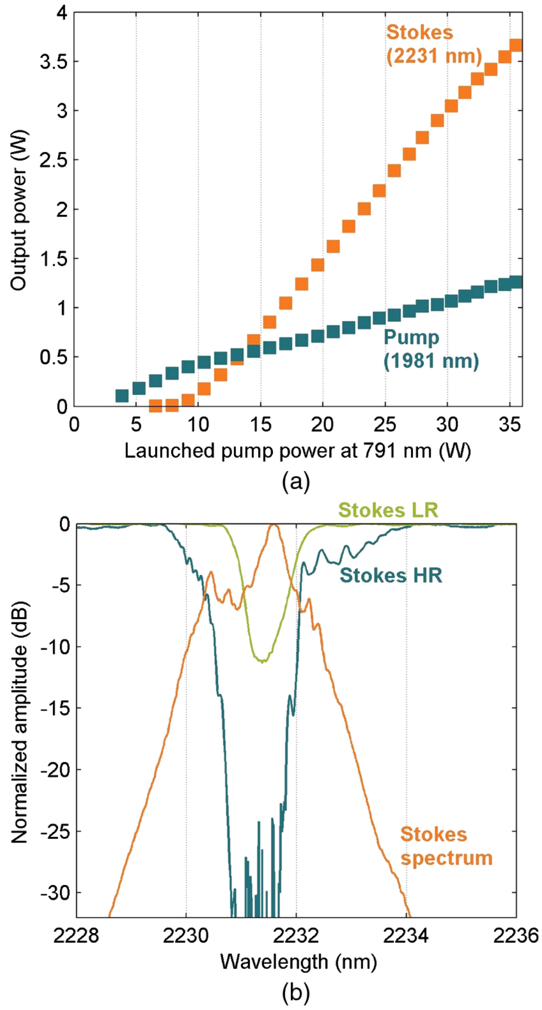

Fig. 8 (a) Output Stokes power and residual pump power of the fluoride glass RFL. (b) FBGs spectrum and output Stokes spectrum at the maximum power available.

also notes in Fig. 8(b) that the Stokes signal was strongly affected by intracavity spectral broadening. Numerical modeling of this nested cavity RFL revealed that the output coupler reflectivity was perfectly optimized to obtain the maximum output power at 36 W of launched pump power, the maximum available.[26]

The main challenge in setting up the laser cavity was with respect to the butt-coupling alignment between the silica and fluoride fibers. The use of Peltier coolers helped to stabilize the temperature of the alignment mounts and consequently, the output power. Peak to peak fluctuations of less than 0.5% were measured at 3.3 W during a 10 min test.

5 Chalcogenide Glass RFL

Arsenic sulfide glass (As_2S_3) has long been identified as an ideal candidate for mid-IR active fiber media because of its high Raman gain coefficient and its extended spectral window, spanning from 1 to 6.5 μm. Yet, no demonstration of Raman gain FL based on chalcogenide fibers had been reported until very recently.[23] This first demonstration of a chalcogenide based RFL, which relies on the use of a specially designed As_2S_3 fiber presenting optical losses below 100 dB/km at 3.4 μm, paves the way for all-FLs operating between 3 and 4 μm.

5.1 *Laser Setup*

A schematic diagram of our RFL is shown in Fig. 9. The laser is pumped by a QCW Er:ZBLAN FL at 3005 nm based on a similar architecture as the one shown in Fig. 3. QCW operation of the pump reduces the thermal load on the As_2S_3 fiber input facet and helps to prevent thermal drift of the launching efficiency. In addition, this specific pump wavelength is used to move the first order Stokes shift inside the low loss spectral window of the As_2S_3 fiber, which begins at about 3250 nm. A free-space launch setup was used to deliver the pump to the chalcogenide fiber core. This setup consisted of two aspheric lenses ($f_1 = 12.7$ mm,

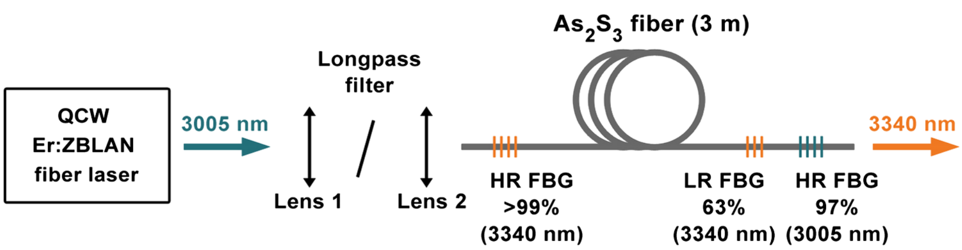

Fig. 9 QCW chalcogenide glass RFL.

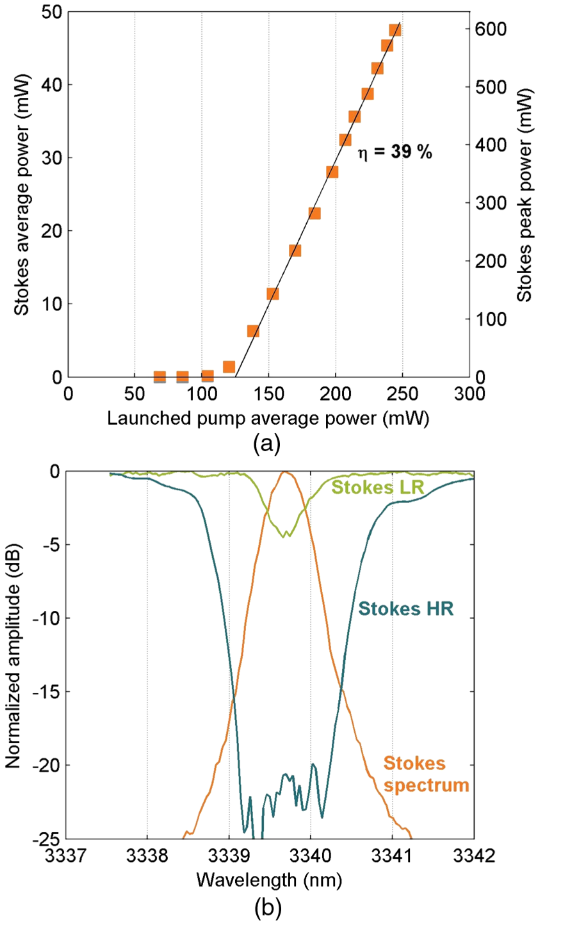

Fig. 10 (a) Output power of the chalcogenide glass RFL. (b) Transmission spectra of the Stokes FBGs and output spectrum at the maximum launched pump power.

$f_2 = 4$ mm), a long-pass filter to isolate the desired spectral content, and precision alignment mounts.

The 3-m long As_2S_3 single-mode fiber had a core diameter of 4 μm, a NA of 0.36, and an attenuation coefficient of 95 dB/km at 3340 nm. Three FBGs were written inside this fiber: the first two FBGs (>99% and 63%) delimited the first order Stokes cavity while the third one (97%) was used to recycle the residual pump. To optimize the spectral overlap between the pair of Stokes FBGs, the output FBG was placed on a special mount that allowed control of the mechanical strain applied to it.

5.2 *Results*

A maximum output peak power of 0.6 W was obtained with a lasing efficiency of 39% with respect to the launched pump power, shown in Fig. 10. The 3340 nm emission wavelength is, to the best of our knowledge, the longest output wavelength ever produced by a RFL. The average power proved to be stable, as less than 0.3% rms fluctuations were measured during 5 min at the maximum power level. On the other hand, high frequency laser noise was observed in each pulse, arising from the Er:ZBLAN pump laser. The precise origin of those instabilities is still under investigation but we currently believe it might be linked to the wavelength of the pump, located on the extreme long edge of the erbium emission band.

6 Conclusion

We have shown that efficient and powerful monolithic FL can be produced around 3 μm based on erbium-doped double clad fluoride fibers. The availability of such FL sources along with those already developed around 2 μm can be used as the backbone for the development of compact and reliable coherent sources over the entire 2 to 4 μm band based on Raman gain in either fluoride or chalcogenide low-loss fibers. We have demonstrated here the first Raman gain fluoride FL operating at 2.23 μm as well as the first RFL based on the first Stokes shift in chalcogenide fiber, operating at 3.34 μm.

Acknowledgments

This research was supported by the Natural Science and Engineering Research Council of Canada (NSERC), Canada Foundation for Innovation (CFI), Fonds de recherche du Québec—Nature et technologies (FRQ.NT), the Canada Excellence Research Chairs (CERC), and CorActive High-Tech, Inc.

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Vincent Fortin received his BS in physics and mathematics with first class honors (summa cum laude) from University of Ottawa, Ottawa, Canada, in 2006, and his MS degree in physics from the Center for Optics, Photonics, and Lasers (COPL), Laval University, Quebec, Canada, in 2009. He is currently pursuing his PhD in physics at the same university, where he studies infrared Raman lasers based on non-silicate glass fibers. His other research interests include supercontinuum generation and rare earth-doped fiber lasers.

Martin Bernier received the PhD degree in physics from Laval University, Quebec, Canada, in 2010. Currently, he is a researcher at the Center for Optics, Photonics and Laser at Laval University. His research interests involve FBGs, FLs, diffractive optics, and femtosecond sciences. He also performed a technical position at National Optical Institute, Quebec, from 1999 to 2001 and worked on diffractive optics technology.

Nicolas Caron received his bachelor's degree in engineering physics from Université Laval, Québec, Canada, in 2006 and his MSc in physics from the same institution in 2008. His research interests include FLs, fiber optics components packaging, and high power infrared lasers.

Dominic Faucher received his BS in physics from Laval University, Quebec City, Canada in 1999 and earned his MS degree in physics from the Center for Optics, Photonics and Lasers (COPL) at Laval University in 2002. His research interests are mainly in rare earth-doped fluoride glass FLs. He now works at TeraXion, Quebec City, Canada.

Mohammed El Amraoui received his bachelor's and master's degrees in material science from Université Montpelier 2, France, in 2005 and 2007, respectively, and his PhD in physics from Université de Bourgogne, France, in 2010. His PhD research was about the development of highly nonlinear optical fibers based from chalcogenide glasses. After completing his PhD, he was a postdoctoral researcher with Prof. Younès Messaddeq at Laval University at Québec for two years. He is currently a researcher in the Center for Optics, Photonics, and Lasers (COPL), Laval University, Quebec, Canada. His main research interest is about the fabrication of high pure exotic glasses/fibers and their infrared applications.

Younès Messaddeq, who holds a PhD in solid state chemistry (1990) and a bachelor's degree in solid state chemistry from the Université de Rennes 1, France, is one of the world's most accomplished researchers in materials development for optics and photonics. In his academic research, he has published over 290 papers in refereed journals and conferences, and presented several invited talks. His H-index is 31, and has well over 3500 citations of his published works. In 2010, he was appointed to one of the 19 prestigious Canada Excellence Research Chairs (CERC), providing 28M$ in research funds at Université Laval, Québec, Canada. He has been a visiting professor at the Physics Institute of São Carlos, Brazil; the Université de Bourgogne, France; and the University of Münster, Germany. He has also served as a visiting researcher and fellow at the National Institute for Inorganic Materials in Tsukuba, Japan.

Réal Vallée received his PhD from Université Laval in 1986. He was a postdoctoral fellow at the Laboratory for Laser Energetics, at the University of Rochester from 1987 to 1988. Since 2000, he has been the Director of the Center for Optics Photonics and Lasers (COPL), the Québec photonics cluster involving six universities and over 300 scientific members. His research interests are optical fiber components and their applications, FLs, nonlinear short pulse propagation in fibers, and waveguide and Bragg grating writing. He has authored over 150 refereed papers, holds eight patents and has supervised 65 graduate students since 1990. He successfully spearheaded a grant application to the Canadian Foundation for Innovation for the construction of a state of the art building entirely dedicated to optics and photonics research on the campus of Université Laval. He is a fellow of the Optical Society of America.