Mid-IR Ultrashort Pulsed Fiber-Based Lasers

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(Invited Paper)

***Abstract*—We review the latest breakthroughs in ultrafast fiber laser technology in the mid-IR wavelength range $\geq 2\ \mu$m. In particular, we concentrate on two novel laser systems built around passively mode-locked Tm:fiber lasers and fiber-based Cr:ZnS lasers, generating sub-100 femtosecond pulses and frequency combs with several Watt average output powers, hundreds of kilowatts peak powers and tens of nanojoule pulse energies. The tunability in the broad wavelength range between 2 and 2.5 μm as well as a simple all-silica-fiber design makes the Tm-fiber laser a truly unique broadband light source, particularly relevant for applications in gas sensing, fine material processing of semiconductors, composite materials, glasses and plastics, as well as for brain surgery, breath analysis, remote sensing and stand-off trace gas detection, especially in oil and gas industry. We also review techniques for coherent supercontinuum generation in the mid-IR, including a novel technique of direct generation of the supercontinuum in the fiber. A competing Watt level few-optical cycle Cr:ZnS laser operating at 2.4 μm (Patent pending, ATLA Lasers, Trondheim, Norway) is distinguished by extremely short pulse duration of only 41 fs, reliability and compactness. This unique ultrashort-pulsed laser generates intrinsically coherent frequency combs, which further extends the application range to high-resolution and high-sensitivity spectroscopy and optical clocks.**

***Index Terms*—Fiber lasers, optical pulse generation, laser mode locking, laser tuning, supercontinuum generation.**

I. INTRODUCTION

THE purpose of the present paper is to review our latest works aimed at development of a novel ultrafast (sub-100 fs) fiber-based laser technology for the generation of eye-safe, high spatial quality and high-intensity (high-brightness) radiation in the eye-safe mid-IR spectral region around and above 2 μm, tunable between 2–2.5 μm, to replace existing in this wavelength range mainly solid-state and OPO-based femtosecond technology.

Until recently the targeted mid-IR wavelength range (2–2.5 μm) represented the gap not only in direct sources of ultrashort pulsed radiation, but also in sources of tunable high power continuous wave radiation, which is interesting for many material processing and gas sensing applications. The high-power ultrashort-pulsed mid-IR laser technology was so far mainly based on optical parametric devices. These technologies suffer such problems as complex external optical components (e.g., vibration is still a problem), beam alignment issues, pre-exit aperture free space beam propagation and, most importantly, thermal management issues. As to 2 μm fiber technologies, most of the R&D has been done on high-power continuous-wave Tm-fiber lasers, only very little in short pulsed femtosecond Tm-fiber lasers, and nothing between 2–2.5 μm. There has been a clear gap in broadly tunable between 2–2.5 μm high-power ultrafast fiber laser technology, both on laser market and applications sides.

This wavelength range gap is currently covered neither by hetero-junction semiconductor lasers, nor by quantum cascade laser technology, which are probably the simplest and the most cost effective sources in the mid-IR wavelength region, especially for gas sensing. The first class of lasers now covers this wavelength range, but not with the needed power levels and can be used only as continuous-wave seed sources for high power ultrashort pulsed fiber- or solid-state laser systems. The QCLs do not allow going below 3.4 μm at performance level, required for such remote applications as remote-sensing and multi-component gas sensing. This hurdle is not going to be overtaken easily in the near future due to fundamental material issues. The QCLs also provide relatively narrow tuning ranges (compare $\sim$140 cm^{-1} tuning range of a quantum cascade laser and $\sim$1800 cm^{-1} tuning range of a Cr:ZnS or Cr:ZnSe laser) and limited output power levels at room temperature (in comparison to tens of Watts level of Cr:ZnS or kW-power levels of Tm-fiber lasers). While mid-IR semiconductor or QCL lasers have a number of fundamental limitations, this makes them less attractive for fine material processing, remote imaging and sensing, as compared to fiber and solid-state lasers: limited average power at good beam quality [1], [2] and, most importantly, low energy storage capacity, which is important for high peak-power operation. The Tm-fiber lasers, but even more Cr:ZnSe lasers, on the other hand, which operate at room-temperature and have the largest relative bandwidth of $\sim$45% of the central wavelength of the laser, can provide very high power levels (up to hundred Watt level), ultrashort pulse duration of only several optical cycles (sub-100 fs) retaining the good beam quality and narrow spectral linewidth at the wide tuning range. The ultrashort pulsed Tm-fiber lasers are in many respects similar to Yb-fiber lasers with the advantage of operation in the eye-safe wavelength region, which significantly reduces requirements on the total cost of ownership of these lasers. In combination with near-infrared

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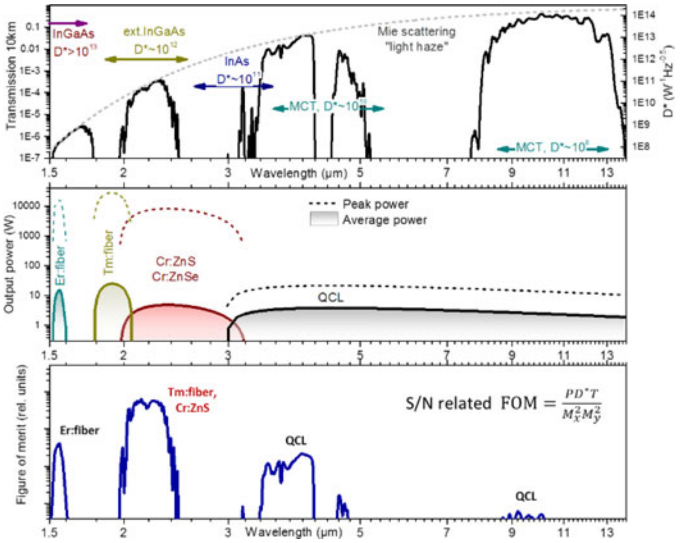

Fig. 1. *Upper graph*: Atmospheric transmission at sea level, and typical detectivity figures for thermoelectrically cooled detectors. *Middle graph*: power levels and wavelength coverage of mid-IR laser technologies. Er:fiber laser data were estimated assuming equal pump power with Tm:fiber. *Lower graph:* Synthetic figure of merit, which defines the S/N ratio at the receiver.

diode lasers as pump sources these lasers can offer stability, efficiency and compactness as well as the broad spectral coverage and tuning ranges, which are generally inaccessible for semiconductor lasers.

Fig. 1 shows the most important for remote applications mid-IR transmission windows of the atmosphere. The QCLs address 4 and 10 μm windows, while fiber and solid-state lasers address the 2.3 and 1.5 μm windows. These transmission windows are not equivalent, as there exists strong wavelengths dependence of maximum transmission due to Mie and Rayleigh scattering on long distances and because detector noise rapidly grows towards the IR. In order to analyze applicability of spectral windows and corresponding systems we define a figure of merit (FOM) in terms of laser power P, beam quality M^2, sensor detectivity D^* and atmospheric transmission T as FOM $= P{\cdot}D^*{\cdot}T/M^4$, which defines the signal to noise ratio at the receiver end. It can be clearly seen in Fig. 1 that solid-state/fiber laser technology in the 2–2.5 μm transparency window outperforms QCLs in the 4 or 10 μm windows by almost two orders of magnitude. Because the reasons for this difference are fundamental, the MWIR LIDAR systems will most likely only employ QCLs as tunable seed lasers for subsequent optical parametric amplifiers. Such laser systems are naturally more complex, bulky and expensive than direct laser sources like, e.g., a Tm-fiber or Cr:ZnS laser.

Another advantage of the 2–2.5 μm window is the broad tuning range, perfectly overlapping with Cr:ZnSe or Cr:ZnS gain media, and with the tunability range of the Tm-fiber based Raman soliton laser. This advantage is critical because it allows along with the fine material processing and conventional sensing some novel applications, e.g., a high-sensitivity and high-resolution 3-D imaging with the capability of simultaneous sensing of hazardous material on the surface of the object. The advantage is also fundamental, because Cr:ZnSe and Cr:ZnS laser media have by far the broadest tuning ranges at room temperature [4]. The competing Er:fiber source in the 1.55–1.8 μm window—an alternative eye-safe laser—offers much narrower gain bandwidth which only partially overlaps with the transparency window of the atmosphere (see Fig. 1).

In addition to being considered eye-safe, the wavelength range from approximately 1.4 to 2.6 μm may be covered using low cost InGaAs detectors operating at room temperatures, and one may utilize conventional silica fibers as well as optical components (lenses, mirrors and optical filters) for the receiver part. However, above approximately 2.5 μm more costly, cooled detectors have to be used in order to maintain the sensitivity and bandwidth relative to InGaAS detectors, as well as more costly and less well-developed fiber materials. The latter is an important consideration for the broad introduction of the fiber lasers to the markets.

In this paper we address the described above existing gap in 2–2.5 μm high-power pulsed fiber technologies and the ways to bridge this gap by applying two approaches, an all-fiber approach of Tm-fiber master oscillator power amplifier (MOPA) lasers and a hybrid fiber/solid-state approach of a novel Cr:ZnS laser. Both described here novel fiber based laser technologies bridge this gap and provide high-power (hundreds of kilowatts peak power at 100 fs pulse durations) good beam quality (high brightness) lasers and supercontinuum (SC) sources operating in the previously inaccessible wavelength range between 2–2.5 μm at several Watt level average power levels by orders of magnitude exceeding the power levels achievable by semiconductor or QCL technology. This was achieved by integration of the commercially available silica based fiber material- and semiconductor laser component technologies with innovative laser design and physical principles to produce a compact yet ultra-high power broadly tunable ultrafast fibre lasers between 2 and 2.5 μm.

The key to the new capability of the developed photonic technology both, in terms of performance as well as in terms of influence on applications, is to obtain an advanced pulsed laser performance in terms of brightness, pulse duration, peak power, broad continuous tunability and ability to select particular operation wavelength in a compact integrated all-fiber format. This unique combination, which has never been demonstrated before in this very attractive, albeit technologically challenging wavelength range, was achieved by combining the most innovative solutions offered by fiber and solid-state laser technologies. This allowed us to explore simultaneously the high efficiency and high peak power capability of Tm-fiber lasers around 2 microns and combine it with the extreme tunability and possibility to select any desirable wavelength in the targeted 2–2.5 micron range in the form of a compact all-fiber high-performance laser device. Comparable only to traditional solid-state and OPO technology, the present technology has the compactness, versatility and robustness of fiber lasers, and as such is ready for real-world applications.

The paper is organized as follows: Section I provides an introduction with an explanation of potential benefits and enabling character of 2–2.5 μm laser technology. Section II provides the literature review of some of the most important developments in ultrafast Tm-fiber lasers. In Section III, we discuss major developments in the ultrafast tunable Tm-fiber MOPA lasers. Section IV focuses on ultrafast fiber based Cr:ZnS lasers. In Section V, we provide conclusions.

II. ENABLING 2–2.5 MICRON ULTRAFAST FIBER LASER TECHNOLOGY: STATE-OF-THE-ART

Tm-fiber lasers operating around 2 microns and Cr:ZnS lasers operating around 2.4 microns are in many respects similar to Yb-fiber lasers with the advantage of operation in the eye-safe wavelength region, which significantly reduces requirements on the total cost of ownership of these lasers. Fiber laser oscillators and amplifiers scale to high output power levels limited by the onset of nonlinear effects and optical damage of the fiber. By extending the operating wavelength from 1 μm of the Yb-fiber laser system to 2 μm of the Tm-fiber laser system, the mode area is increased by a factor of 4 and the SBS threshold is increased by a factor of 2. Therefore, the output power of the Tm-fiber laser system is 8x higher than that of the 1 μm Yb:fiber system. A few years ago the breakthrough work of Goodno *et al.* [3] in thulium fiber amplifier power scaling has led to over 600 W of fundamental mode output power at 60% optical efficiency. Nowadays thulium fiber lasers produce kW-level output powers in continuous wave regime.

The ultrashort pulse generation in fiber lasers has started in early nineties competing with the corresponding developments in femtosecond solid-state lasers. The first mode-locked fiber lasers were based on Er [4] and Nd [5] fiber lasers, followed by Yb [6] and then by Tm [7] fiber lasers. The shortest pulses can be generated by the variation of Kerr-type mode-locking technique, like, e.g., using the nonlinear Sagnac interferometers [4] or nonlinear polarization rotation [8] based on the nonlinear interference between the two polarization modes. Self-starting passively mode-locked fiber lasers were also demonstrated using semiconductor saturable absorbers (SA) [9]. Although the pulse duration from fiber lasers mode-locked with SA are typically a few times longer compared to systems based on nonlinear polarization rotation due to the slower response time of SA ($>$100 fs, even for graphene), they allowed developing the passively mode-locked single polarization fiber lasers [10]. The best results in terms of pulse duration create hybrid systems comprising nonlinear polarization rotation mechanism in combination with the starting function of the saturable absorbers.

Hitherto only a few ultra-short-pulse fiber oscillators based on thulium-doped fiber have been demonstrated. Oscillators with additive-pulse mode locking [7], [11] require complex cavity with bulk elements in it. Mode locking with help of SA is a promising route to manufacturing portable sub-picosecond fiber lasers [12]. Passive mode locking of Tm-doped lasers requires Sb-based SESAM to design an all-fiber ultra-short-pulse sources [13], [14]. Lately it has been demonstrated that carbon nanotube absorbers (CNTAs) inserted in the cavity allows to produce mode-locking regime of operation of Tm-doped fiber lasers (TDFLs): ring cavity thulium fiber laser mode-locked with a single-wall CNTA used in transmission produces 1.32 p.s. pulses at 1.93 μm wavelength [15], and the same idea was exploited in linear cavity all-fiber laser [16] resulting in shorter pulses of 700 fs, as well as later also using the graphene saturable absorber [17], [18]. The picosecond scale pulse solutions benefit from the compact all-fiber design and do not require stabilization and adjustment. The pulse duration that is defined by combination of dispersion, fiber nonlinearity and peak power of the pulse, in these cases was limited by fiber dispersion that was close to material dispersion of the silica glass. Particularly useful are the concepts of soliton lasers, first demonstrated by Kafka *et al*. in a fiber laser [19], and similariton lasers as demonstrated by Ilday *et al.* [20]. The pulse durations obtainable with soliton lasers are generally longer than those obtainable with dispersion compensated systems, they typically generate bandwidth-limited picosecond pulses with pulse energies of the order of 1 to 10 nJ [21]. Another promising mode-locking technique is a hybrid mode-locking using simultaneously both nonlinear polarization rotation and saturable absorber [22], [23]. This technique allows achieving 600 fs pulses at up to 300 mW output power generated directly from the oscillator. It should be noted that the same technique with exploitation of CNT also resulted in shorter pulses of 450 fs duration at lower output power of 18 mW [24].

Above the 2 μm wavelength, a femtosecond Ho-doped fiber laser has been recently reported [25]. The laser, pumped by a 1.16 μm semiconductor disk laser, produces 890 femtosecond pulses with the average power of 46 mW and the repetition rate of 15.7 MHz at the wavelength of 2085 nm. Later on, a broadband light source based on the same Ho-doped fibers has been developed [26], delivering up to 273 mW of output power, with central wavelength varying from 2.025 to 2.05 μm and full width at half maximum of 54 nm. A Ho-doped fiber amplifier was used to get nanosecond SC in the mid-IR range using all-fiber scheme, pumped by a Q-switched Er-doped fiber. The observed spectrum covers the spectral range from 2000 to 2500 nm with the power variation less than two decades, and average power of 0.4 W and pulse energy of 0.1 mJ have been demonstrated [27].

High power sources of femtosecond pulses in the spectral domain of wavelengths larger than 2 μm are useful for a number of commercial and scientific applications, including remote sensing, micromachining, THz generation and two-photon microscopy. Recently the generation of 36.7 μJ pulses of 910 fs duration in two-stage large-mode area TDFL in MOPA configuration with the use of an external bulk grating compressor were reported [28]. Raman soliton generation is an attractive alternative to a direct amplification of a seed laser. It has such advantages as tunability of the generated pulse spectral position in a wide spectral range. Tunability over 140 nm range was demonstrated in [29] where 108 fs pulses with energy of 31 nJ and average power of 3.1 W were obtained. The Er-doped fiber laser (EDFL) used in that work as a seed laser produced 400 fs pulses, which were transformed into Raman solitons in a passive fiber and after that amplified in two stages. Single-stage MOPA system utilizing the EDFL pumped TDFL seed laser was demonstrated to produce 5 nJ pulses of $\sim$100 fs duration generated directly in the amplifier with average power of $\sim$350 mW tunable up to 2.2 μm [30]. SC generation in

TABLE I
AVERAGE POWER P, SPECTRAL RANGE λ, PULSE DURATION τ, PULSE ENERGY E, SPECTRAL POWER DENSITY S

		P, W	λ, μm	τ, fs	*E*, nJ	*S*, mW/nm
TDFL	AP	0.01	1.95	350	<1 nJ	–
		1	1.95	3000	100	–
	exp.	1.27	2-2.2*	130	28	–
		2.95	2-2.2*	200**	38	–
SC	F	3	0.39-2.4	–	–	1.5
	AP	0.1	1.9-2.4	–	–	0.2
	exp.	5	1.95-2.5	–	–	10
		3.08	1.87-2.7	–	–	3.7

Legend: AdValue Photonics (AP), Fianium Inc. (F), this paper (exp.).
* tuning range.
** estimated from spectrum.

mid-infrared wavelengths range above 2 μm, and in particular in the transparency window of the atmosphere between 2 and 2.5 μm, is an important goal of applied fiber optics due to numerous applications such as LIDAR systems [31], optical coherence tomography [32], optical clocks, trace gas sensing [33] and environmental monitoring, as well as military technologies. So far, the SC generation in this wavelength range was demonstrated by various methods, e.g., pumping of a Ho-doped fiber amplifier with intense Q-switched pulses at 1.6 μm resulted in an output SC power of 0.4 W [27], by amplification of gain-switched and mode-locked Tm/Ho-doped laser pulses of a complex shape and duration on a few microseconds level in a Tm-doped amplifier resulted in 2.17 W [34], by amplification of already generated with nanosecond EDFL SC in Tm-doped fiber amplifier [35], [36] resulted in up to 2.37 W of output power in the range of 1.95–2.52 μm at –10 dB level [37]. Nevertheless, a much simpler system in a MOPA configuration utilizing a mode-locked laser seed thus ensuring the SC stability and quality and based on Tm-doped fibers only has not been yet reported and will be described in the following section.

III. MULTI-WATT FIBER LASER FOR >2 μm RANGE

Fiber lasers is a novel promising branch in mid-IR laser physics. Generally, they enable compact, robust and reliable ultra-short pulse optical sources as well as SC light sources. The problem is that it is usually difficult to realize a reliable femtosecond seed laser with pulse duration less than several hundred femtoseconds. The novel approach of Raman soliton generation with the required duration from much longer picosecond pulses allows overcoming this drawback. The advantage of this technique is that the generated short pulse is tunable in a broad spectral range. Thus, it is possible to produce not only the ultra-short pulse laser systems, but even tunable ones. Table I represents the recent state of the commercial sector as well as the experimental results reported here showing the potential of fiber laser systems.

Thulium-doped fibers provide an excellent source for generation of radiation around 2 μm. They can be efficiently pumped at 1550–1650 nm or at 770–810 nm and provide a wide lasing band (see Fig. 2) [38], [39].

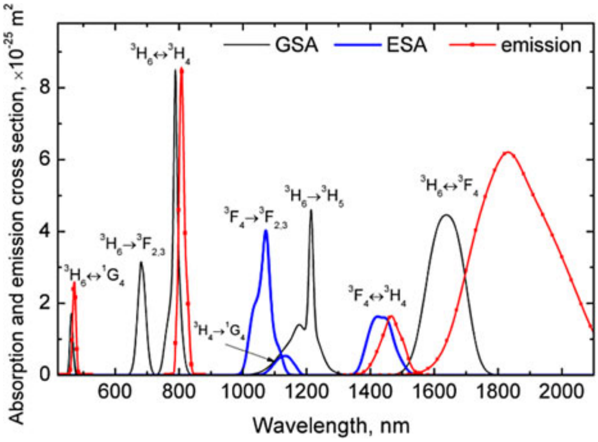

Fig. 2. Absorption and emission of Tm-doped fiber [38].

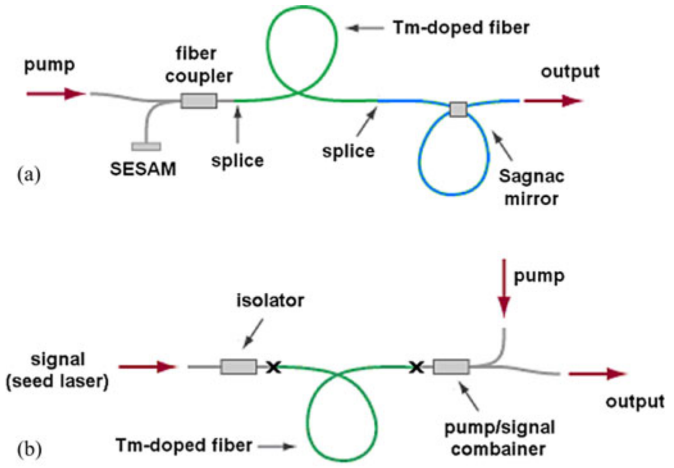

Fig. 3. Laser scheme, (a) seed laser, and (b) amplifier.

Raman soliton generation provides a significantly more simple and more powerful approach for all-fiber TDFL MOPA system which is capable of producing widely tunable Raman solitons with duration of few hundreds femtoseconds and peak power of a hundred of kilowatts level with several watts-level average output power. This simple and elegant method of producing high power broadband radiation in the mid-IR wavelength range without the need for using microstructured or specialty fibers, besides the standard commercially available silica Tm-doped fibers, has been reported in [37]. The MOPA concept with Tm/Ho-doped fiber amplifier was used in [14]; the core-pumped by an EDFL fiber amplifier demonstrated about 230 mW average power Raman soliton output of the shortest pulse duration of 150 fs at the repetition rate of 50 MHz. The corresponding pulse energy was 4.6 nJ. The SC that was also produced from the amplifier extended over 2.2. μm. The advantage of this approach is that Ho increases the gain bandwidth of the amplifier. Nevertheless, even with Tm-doped fibers it is possible to achieve high-power output in an all-fiber design. The concept is based on a laser diode pumped MOPA system utilizing a mode-locked core-pumped seed laser, Fig. 3(a) and a clad-pumped fiber amplifier, Fig. 3(b). The system uses only commercially available single mode silica-based components

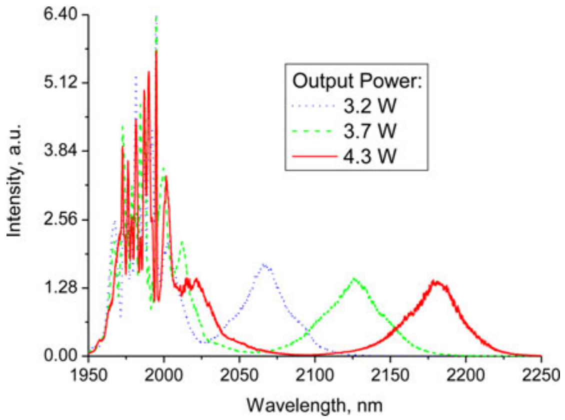

Fig. 4. Output spectra from the amplifier for different output power.

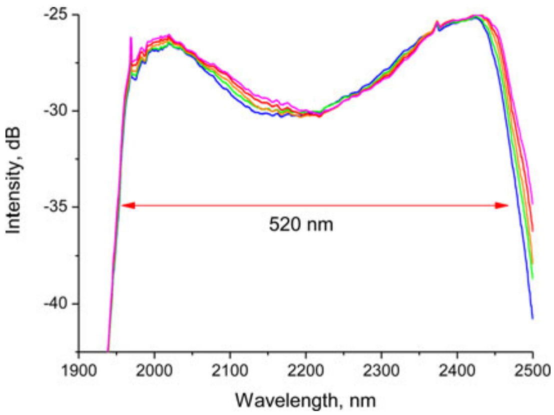

Fig. 6. Evolution of the SC spectrum with power increase from 4 to 5 W.

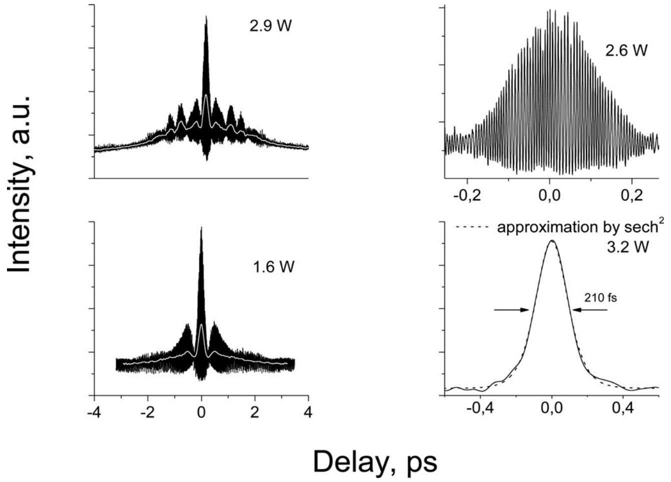

Fig 5. Autocorrelation traces (with intensity autocorrelations) and examples of the corresponding to the Raman soliton central peak typical shape and its approximation.

and can produce both, tunable solitons and super-flat SC in the region of 1.9–2.5 μm, as we shall describe below.

A. Multi-Watt Raman Soliton Pulse Generation

Raman soliton was directly produced in the Tm-doped fiber amplifier [37], [40]. In the range of 3–4 W of the output power the spectral FWHM of the soliton band consists of about 40 nm insignificantly increasing with the power increase. The average soliton power obtained from the integration under the spectral curve insufficiently changed with the spectral shift. It reaches its maximum value of 1.27 W (33% from the total output power) corresponding to the position of the soliton maximum at the wavelength of 2.125 μm starting from 0.95 W at 2.04 μm and ending with 1.25 W at 2.18 μm (see Fig. 4).

The duration of the corresponding bandwidth-limited soliton amounted to 120 fs (see Fig. 5). The FWHM of the intensity autocorrelation (210 fs) corresponds to the pulse duration of 130 fs. This duration corresponds well to the Raman soliton temporal width estimated from the spectra for a transform-limited pulse.

While we did not observe soliton break-up, we expected some distortion of the soliton spectral shape with power increase because components with lower intensity experience less spectral shift. However, the spectral shape of the Raman soliton remained practically unchanged (see Fig. 4). For the average optical power of 1.27 W and repetition rate of 46 MHz the energy of a single pulse is equal to 28 nJ and its peak power under assumption of sech^2 profile of the pulse is as high as 190 kW.

Even higher output powers were obtained for a similar MOPA with another seed laser of higher pulse repetition rate of 77 MHz. When pumped at higher optical powers of up to 30 W, its output reached 10 W. Qualitatively, this laser showed a similar behavior to the previous one. At high pump powers the part of the average power corresponding to the Raman soliton obtained from spectra integration also amounted to approximately 1/3. It reached its maximum of 2.95 W for 8.85 W of output power with the soliton spectral peak at the wavelength of 2.15 μm. The corresponding pulse energy amounted to 38 nJ. The spectral FWHM of the soliton consisted of 25 nm. The duration of a transform-limited pulse of such bandwidth and sech^2 shape amounts to 200 fs, thus the predicted peak intensity is on the same level of magnitude as in the previous case. This result demonstrates the average power scalability and feasibility of obtaining ultrashort high-energy pulses with multi-Watt average power.

B. SC Generated Directly in the Amplifier

Increase of the pump power leads to the emergence of a Raman solitons series and further increase leads to the whole spectrum smoothening and formation of an optical SC (see Fig. 6). This results in a 5 W optical SC covering the range of 1.96–2.48 μm (520 nm) with 5 dB flatness, without the need for spectral broadening in a nonlinear microstructured or other specialty fiber [41]. The average spectral power density was as high as 10 mW/nm. The average output power of the forward-pumped fiber amplifier reached 5.06 W whereas the average power of the seed laser producing a stable pulse train at the wavelength of 1960 nm with 44 MHz repetition rate and pulse duration of 2 p.s. amounted to 6.7 mW. Even higher output power, up to 6.8 W, was obtained in a similar configuration with spectral flatness of 8 dB [42]. The average spectral power density was as high as 14 mW/nm.

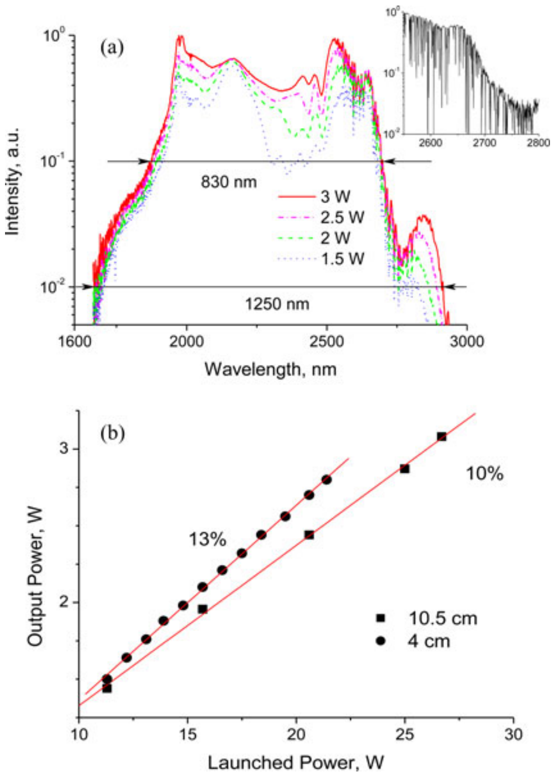

Fig. 7. Output spectrum at maximum power (a) and average power (b) from the germanate fiber. The spectrum with 0.5 cm^{-1} resolution is given in the inset.

The advantage of the technique is that it is possible to generate a broadband spectrum emission directly in a silica fiber without need for special fibers. Because the spectrum expansion is driven by Raman gain, the expansion stops in the area of a high optical loss overcoming Raman gain. This peculiarity naturally prevents radiation from expansion in a high-loss area and allows us to realize high-power multi-watt optical SC sources even with silica-glass based fibers.

C. Germanate Fibers for SC Generation

The nonlinear germanate fiber with a solid splice to the amplifier produced even broader spectrum [43], [44]. The spectra were also stronger expanded in the blue direction because of a positive dispersion in the fiber below 2 μm. The resulting bandwidths at –10 level reached 830 nm (1.87–2.7 μm) and 1250 nm (1.66–2.91 μm) at -10 and -20 dB levels, respectively (see Fig. 6). The spectrum had a complex structure. It can be associated with the change of the dispersion sign, thus linear extrapolation from the available data gives the zero dispersion value in the vicinity of 2.5 μm [45].

Dependence of the average output power versus pump power launched into the amplifier is shown in Fig. 7(b). For this fiber it was possible to achieve the SC power of 3.08 W. The slope efficiency at high pump power of the laser system as a whole amounted to 10%.

The expanded spectrum allowed observation of the narrow atmospheric absorption lines (see Fig. 7(a), inset) as the radiation propagated 1.65 m in air before entering FTIR, the cavity of which has not been isolated from the outer environment as well. The observed features correspond to absoption of atmospheric H_2O and CO_2.The advantage of such nonlinear fibers is that their application allows to keep all-fiber structure of the laser systems, with reliable solid splices. This provides the highest level of the reliability that cannot be reached with other commonly used non-linear fibers.

IV. Fiber-Based Watt- Level Cr^{2+}–Lasers

Though the advantages of the all-fiber laser systems are well known, there are certain areas where they still cannot compete with the solid-state lasers, the most important aspects being the pulse duration and pulse quality in the mode-locked regime.

In the particular mid-infrared spectral region there exists an important group of materials experiencing a lot of attention in the last decade—the chromium-doped chalcogenides. Since the Cr^{2+} ion refers to transition metals, it exhibits wide absorption and emission bands. Cr:ZnSe, the most known representative of the chalcogenides doped with Cr, has spectroscopic and thermal properties similar to the well-established Ti:sapphire laser crystal. For that reason it is often called a “Ti-sapphire of the infrared”.

After the first report on the Cr:ZnSe laser action in 1996 [46] a number of important milestones were reached before the material matured enough to demonstrate the femtosecond laser pulses. Among them one can mention the demonstration of tunable CW laser action [47] including the impressive 1400-nm range of continuous tunability [48], the realization of diode-pumping [49] and pumping through the charge-transfer bands [50], [51], the high-power CW laser action in ceramic material [52], etc. The femtosecond era for the Cr:doped chalcogenides began in 2006, when the first mode-locked Cr:ZnSe laser was reported [53]. Kerr-lens [54], [55] and graphene mode-locking [56] were demonstrated subsequently. To date, output power up to 300 mW, pulse energy up to 2.3 nJ [57], pulse duration as short as 80 fs [58] were achieved for Cr:ZnSe-based lasers.

These spectacular achievements are largely due to the availability of the appropriate pump sources. The first mode-locking experiments with Cr:ZnSe were performed using such “exotic” pump sources as NaCl:OH$^-$ [59] and Co:MgF$_2$ [60], [61] lasers and resulted in tens of mW of output power at picosecond durations. All femtosecond results have been achieved using either Er-fiber or Tm-fiber pump sources that have become available commercially later. Femtosecond oscillator is very sensitive to beam quality and power stability (long-term as well as short-term) of the pump source, and fiber lasers perfectly match these requirements.

The advantages of the fiber-laser pumping can be even better utilized by another laser material of the same family, Cr:ZnS. As shown in Fig. 8, the absorption band of the Cr:ZnS material ideally fits to Er-fiber laser emission range. Given the excellent thermooptical and nonlinear-optical properties of Cr:ZnS [50], one can consider the Er-fiber laser and Cr:ZnS material as a perfectly matched pair for high-power femtosecond generation in the mid-IR.

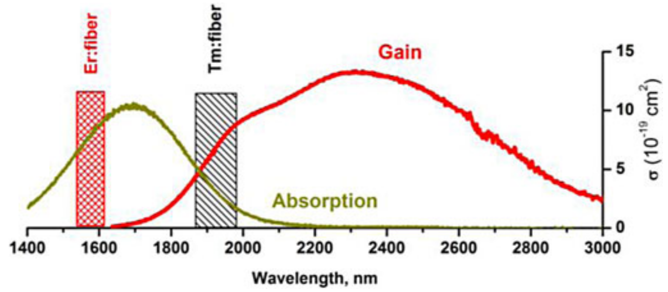

Fig. 8. Absorption (yellow) and emission (red) spectra of Cr:ZnS; emission spectra of Er:fiber (red bar) and Tm:fiber (grey bar). Adapted from [62].

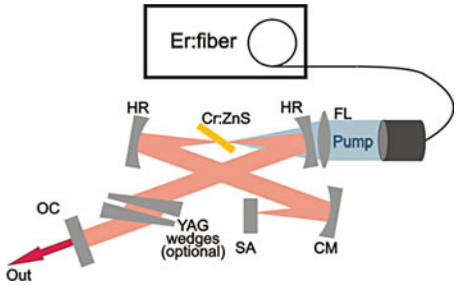

Fig. 9. Experimental setup of the fiber-based graphene mode-locked Cr:ZnS laser: FL is the pump focusing lens, HR are the highly-reflective concave mirrors, CM is the concave chirped mirror, SA is the carbon-based saturable absorber mirror, OC is the output coupler. Patent pending, ATLA Lasers, Trondheim, Norway.

The first Cr:ZnS laser was demonstrated in 2002 [63], [64], but for a certain period remained rather underinvestigated due to the material availability issues. The picosecond passive mode-locking was reported in 2006 [65] and femtosecond SESAM-initiated mode-locking was obtained in 2011 with 130 mW 130 fs pulses [66], [67].

The availability of good-quality and single-crystalline samples allowed the Er-fiber—Cr:ZnS system to unleash its real potential. At the moment, Er-fiber based Cr:ZnS lasers hold the lead in almost every respect—from commercially available tens of Watts of tunable continuous-wave operation [68] to few-cycle femtosecond pulses, demonstrated recently [69], [70]. Kerr-lens mode-locked Cr:ZnS laser produced 69-fs pulses with pulse energy of 3.8 nJ and average output power of 550 mW. The spectral bandwidth of the laser emission at 2.39 μm reached 91 nm. Laser pulses were close to transform-limited with the time-bandwidth product of 0.335, and the beam quality close to $M^2 = 1$. This result initiated a series of works on SC generation in step-index chalcogenide and germinated fibers. The stable coherent SC with spectral bandwidth reaching 800 nm was demonstrated [71], [72]. Furthermore, also in 2012 the environmentally protected delivery method of femtosecond pulses from Cr:ZnS laser through the soliton formation in commercially available ZBLAN fiber was demonstrated [73]. That result is very important for practical implementation of the broadband mid-IR sources usually suffering from the atmospheric absorption.

Year 2013 brought the remarkable progress for Cr:ZnS laser. Though Kerr-lens mode-locking is rather developed and widely commercialized technology, it still requires external starting mechanism and operation near the edge of the cavity stability region. Implementation of a saturable absorber as a starting mechanism allows better controlling of the mode distribution and easier power scalability. Realization of the graphene-mode-locking [74] and, very recently, mode-locking by the carbon nanotubes [75] allowed greatly improving the performance of Cr:ZnS laser. The generalized setup of the fiber-based passively mode-locked Cr:ZnS laser is shown in Fig. 9. The system showed itself to be rather flexible platform which could be tuned to optimize the pulse duration, the pulse energy, or the average output power.

These impressive results have been achieved using single-crystalline Cr:ZnS and Cr:ZnSe. There has also been a continuous effort to achieve mode-locked operation with polycrystalline Cr:ZnSe and Cr:ZnS materials. The first tunable continuous-wave and diode-pumped ceramic Cr:ZnSe was reported in [76] and was actively mode-locked a year later [55]. However, it took a few more years to achieve the femtosecond regime, with the first ceramic Kerr-Lens mode-locked Cr:ZnSe laser to be demonstrated in 2010 [77] and a similar Cr:ZnS laser in 2013 [67]. Moreover, the remarkable success with the single-crystalline Cr:ZnS laser led us to the development of an even more powerful ceramic Cr:ZnS laser producing 140-fs pulses with an average output power exceeding 1 W [78], ready for the real world applications.[1]

A. Pulse Duration

Pulse duration of 41 fs was reached in graphene-mode-locked Cr:ZnS oscillator [79] with a nearly-200-nm (FWHM) spectral bandwidth directly from oscillator. Such pulse duration corresponds to just over 5 optical cycles at 2.4 μm and is equal to sub-15-fs pulses on the emission wavelength of Ti:sapphire laser. These are the shortest pulses generated in the mid-IR, as well as the shortest pulses obtained with the graphene saturable absorber to-date. The important step in realization of such short pulses was the implementation of the YAG wedge pair inside the cavity. In contrast with the prism pairs traditionally used in the mode-locked lasers to compensate the dispersion, they exploit the material dispersion of YAG. It is an important feature of ZnS that its dispersion in the mid-IR, like for all the other chalcogenides, has the opposite sign in comparison with the dispersion of many common optical materials. That fact allows the all-material dispersion compensation [53], [67], [80], [81], leaving only the third-order dispersion to be corrected by, e.g., chirped mirrors [58], [66], [69]. The wedges were designed for minimal distortion of the cavity mode and allowed fine tuning the dispersion in order to extract the maximal bandwidth of the laser emission. The laser spectrum, experimental autocorrelation trace, and autocorrelation trace derived from the laser spectrum (autocorrelation of the chirp-free pulse) are shown in Fig. 10. The material gain bandwidth can support pulse durations down to a single optical cycle, but that requires redesigning of the chirped mirror to minimize the third order dispersion.

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[1] Patent pending, ATLA Lasers AS, Trondheim, Norway.

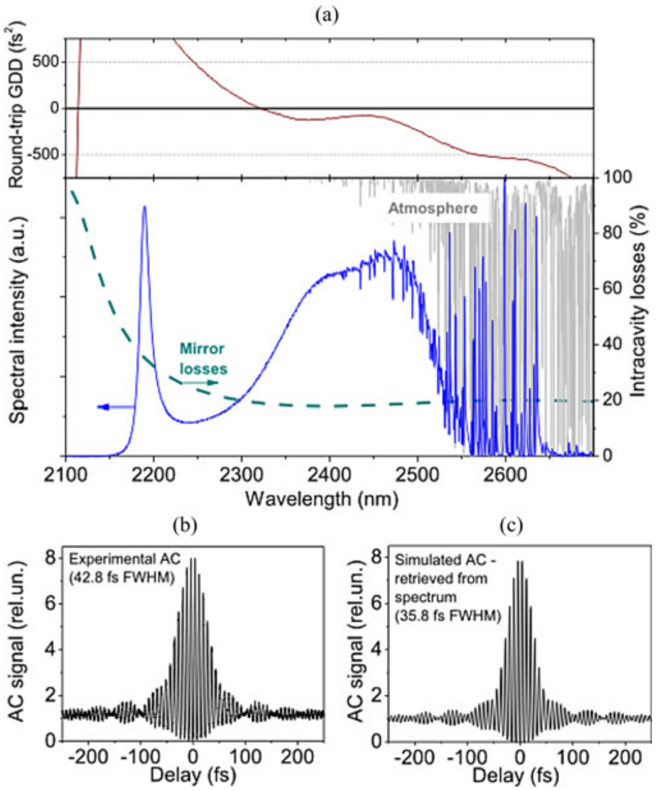

Fig. 10. (a) Laser spectrum of graphene mode-locked Cr:ZnS laser, cavity round-trip GDD and atmospheric absorption; (b) experimental autocorrelation trace of the shortest laser pulses; (c) simulated autocorrelation trace of the laser output retrieved from laser spectrum.

B. Pulse Energy

Scaling the pulse energy of the femtosecond laser usually requires advanced solutions. One of the methods is generation of the optical dissipative soliton in a so-called chirped pulse oscillator (CPO) [82]. This method has been realized in a number of laser systems, both solid-state and fiber, including Cr:ZnSe and Cr:ZnS laser [66], [83]. Recent realization of the Cr:ZnS CPO mode-locked by graphene-based saturable absorber mirror allowed to expand the cavity and obtain pulse energy as high as 15.5 nJ directly from the oscillator [84]. This system is the first reported graphene-mode-locked CPO. The pulses with duration of around 870 fs were extracavity compressed down to 180 fs without the substantial energy loss (see Fig. 11). The pulse energy of 15.5 nJ, being the highest pulse energy directly generated from the oscillator in the mid-IR, is further scalable subject to fine dispersion adjustment. The pulse energy in CPO systems is limited by the occurrence of the chaotic mode-locking regime, which is very sensitive to the cavity dispersion [85]. At the same time the pulse energy limit set by the third order nonlinearity of the active element [79] is shifted to microjoule level in the CPO systems. For the average power of 1 W available from the mode-locked Cr:ZnS laser [83], [86], pulse energies up to 30 nJ could be obtained without increase of the pump power. Subject to more powerful pump and sufficient cooling, approaching the microjoule level seems feasible.

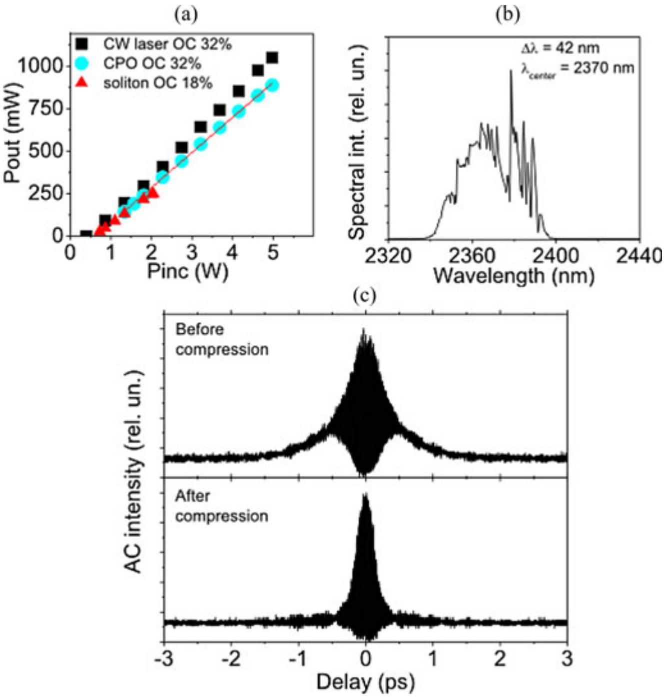

Fig. 11. The output characteristics of the graphene-mode-locked Cr:ZnS chirped-pulse oscillator [83]: (a) comparison of the input-output curves of CW laser, CPO and soliton mode-locked oscillator (in the single-pulse regime); (b) the spectrum of the laser emission at 15.5 nJ pulse energy level; (c) interferometric autocorrelation of the laser pulse before and after extracavity compression.

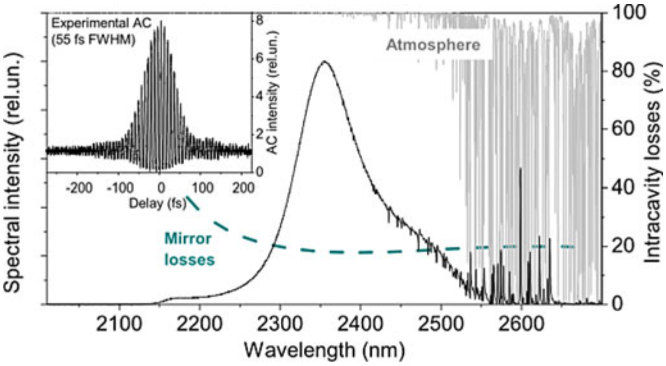

Fig. 12. Laser spectrum and of the high-average-power graphene-mode-locked laser oscillator. Inset: experimental autocorrelation trace of the laser pulses.

C. Average Output Power

Some applications require high average powers in femtosecond regime. Kerr-lens mode-locked Cr:ZnS laser with output powers around 1 W has been already reported [83], [86], but it operated either in double-pulse or in chirped-pulse regime. Realization of the graphene mode-locked Cr:ZnS laser allowed to obtain stable single-pulse output power around 850 mW with pulse duration of about 55 fs (see Fig. 12). The spectral bandwidth of the laser emission was measured to be about 100 nm FWHM. More than 1 W of output power could be obtained with a pulse duration about 140 fs [78]. Table II summarizes the parameters of all Cr^{2+}-based ultrashort-pulsed systems demonstrated to-date.

TABLE II
OUTPUT PARAMETERS OF CR:CHALCOGENIDE LASERS

Active medium	Pulse duration, fs	Average output power, mW	Pulse energy, nJ	Reference
Cr:ZnS	**42.8**	75	0.31	[79]
Cr:ZnS	870	700	**15.5**	[84]
Cr:ZnS	55	**815**	3.3	[83]
Cr:ZnSe	**80**	80	0.44	[58]
Cr:ZnSe	121	165	**1.8**	[87]
Cr:ZnSe	100	**300**	1.5	[54]

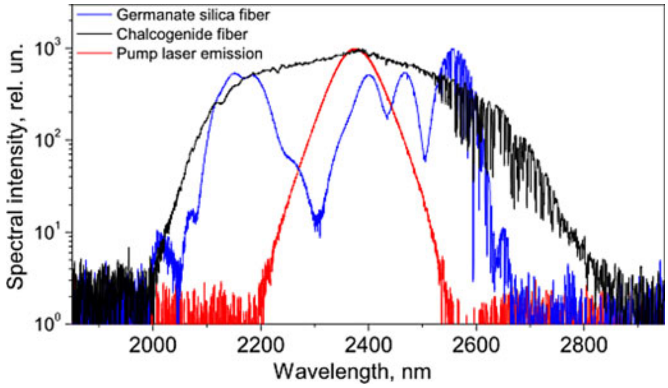

Fig. 13. Spectrum of the femtosecond Cr:ZnS laser emission (red), and the emission outcoming of 1.2-m chalcogenide fiber (black), and 65-cm germanium-codoped silica fiber (blue). The noise-like features on the spectra above 2450 nm originate from the atmospheric water vapor absorption lines in the air.

D. SC Generation

As discussed above, the success in development of the Cr:ZnS femtosecond laser stimulated series of works on SC generation in fibers. Since the pump source was capable of providing high pulse energy, most attention was drawn to non-PCF step-index fibers. The best results were achieved with chalcogenide and germanium-doped silica fibers [71], [72]. The spectra are plotted in Fig. 13.

From the two fibers, the chalcogenide fiber possess much higher nonlinearity, while the germanate fiber was pumped closer to the zero dispersion wavelength. The coupling efficiency of the pump emission was rather poor, resulting in the launched pulse energy lower than 1 nJ for both. The SC reaches 800 nm bandwidth for chalcogenide fiber and 600 nm for germanate fiber (at −20 dB level). The spectral shape of the SC generated in silica-based germanate fiber is modulated due to self-phase modulation effect and suppressed on the red wing by the phonon-assisted absorption in silica. To the contrary, the SC in the chalcogenide fiber is stable and maintains clear symmetrical shape that is advantageous for the spectroscopic applications.

V. CONCLUSION

Ultrafast fiber based lasers in the mid-IR wavelength range above 2 μm came out of age. Their robustness, compactness and reliability combined with the advanced performance—multi-Watt operation at hundreds Watt peak power and tens of femtoseconds pulse duration offer an unprecedented level of utility for many industrial applications, including fine material processing of plastics, composites and semiconductors as well as all kinds of sensing and imaging.

The new techniques include, but are not limited to generation of the Raman soliton of more then 1 W of optical average power produced in the active fiber of the Tm-doped amplifier. Remarkably, the soliton spectral position is tunable in the range of 2–2.5 μm by changing the pump power. The highest average power was achieved for the soliton with the spectral maximum at 2.125 μm and reached 1.27 W, corresponding to the pulse energy of 28 nJ. The corresponding soliton duration estimated from the autocorrelation traces amounted to 130 fs. Notably, up to 3 W of average power corresponding to the pulse energy of 38 nJ was obtained for a Raman soliton of 25 nm spectral bandwidth centered at 2.15 μm with a seed laser operating at the wavelength of 1986 nm with 77 MHz repetition rate and the average power can be scaled further up.

We also proposed and realized a novel simple and cost-effective concept for continuum generation based on a MOPA all-silica-fiber laser. The one-stage silica-based Tm-doped fiber amplifiers generated SC in the range of 1.95–2.5 μm with 5 and 8 dB flatness with 5.06 and 6.83 W average powers from 27.7 W of pump power at 46 and 77 MHz pulse repetition rates, respectively. This is the highest reported SC power with the superb spectral characteristics of SC for all-fiber TDFL system, making this SC generation technique attractive for practical applications, especially lidars.

The results show a large potential of the Raman soliton generation technique for the development of simple, compact, and practical high-power ultra-short pulse fiber lasers. However, Er-fiber based Cr:ZnS lasers hold the lead in almost every respect connected with the pulse duration and its quality, stability and spectral coherence—from commercially available tens of Watts of tunable continuous-wave operation to few-cycle 41-fs femtosecond pulses.

This breakthrough in mid-IR high power and high efficiency tunable ultrashort pulse fiber laser technology and performance opens new research avenues that could lead to novel and practical lasers capable of addressing applications in many fields, including sensing and fine materials processing.

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