Recent Breakthroughs in Solid-State Mid-IR Laser Technology

Lasers based on transition metal doped II-VI chalcogenides are coming of age for practical applications

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Availability of compact and cost-efficient mid-infrared (MIR) laser sources with a broad range of parameters is of great interest for a number of current and emerging applications. For instance, femtosecond (fs) MIR sources with high average power and broad spectral span enable high dynamic range spectroscopy, sensing, and imaging in the molecular fingerprint region. On the other hand, few-optical-cycle MIR pulses with high energy are of particular importance for the development of EUV and X-ray sources based on strong field nonlinear optics, e.g. high harmonic generation. Furthermore, high-power mid-IR lasers are of interest for processing polymers, glasses and composites, surgical, dental and cosmetology procedures, as well as numerous defense-related applications.

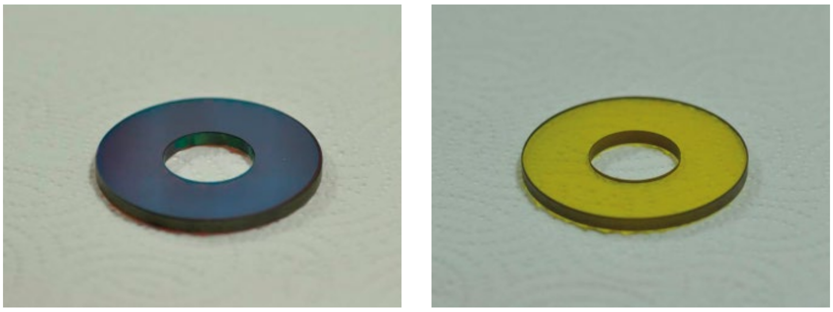

Fig. 1 Lage size polycrystalline gain elements for high power MIR lasers based on spinning ring technology: Ø 50 × 6 mm Cr:ZnSe (left) and Ø 50 × 5 mm Cr:ZnS (right)

Introduced in 1970s, the first solid-state MIR lasers were based on alkali halide crystals with color centers or oxide and fluoride crystals doped with trivalent rare earth ions and required inconvenient cryogenic cooling of the gain medium. Transition-metal doped II-VI semiconductors (TM:II-VI) were introduced as a new class of gain media in the late 1990s by William Krupke's group at Lawrence Livermore National Laboratory, Livermore, CA [1]. By their design, TM:II-VI are effective MIR laser materials. Features of II-VI semiconductor hosts (wide bandgap, low phonon cutoff, tetrahedral coordination) are very favorable for doping by TM ions. Chemically stable divalent TM dopant ions provide the 'right' multiplet structure for broadly tunable MIR lasers, including broad absorption and emission bands, high cross-sections, and absence of excited state absorption.

ZnS and ZnSe doped with Cr^{2+} and Fe^{2+} are typical and the best known representatives of the large TM:II-VI family, as reviewed in [2]. Advantages of Cr:ZnS and Cr:ZnSe lasers include room-temperature (RT) operation with close to 100 % quantum efficiency, very broad tuning over 1.9 – 3.4 µm range, and convenient pumping by reliable erbium (Er) and thulium (Tm) fiber lasers with pump conversion efficiency in excess of 60 %. Broad emission bands of Cr-doped ZnS and ZnSe are favorable for generation of ultra-short pulses; these materials are often referred to as the "Ti:sapphire of the middle IR". In many respects, Fe:ZnS and Fe:ZnSe lasers are complimentary to Cr-based sources. They are pumped in the 2.5 – 3.3 µm range and tunable over 3.4 – 5.2 µm range; they operate at RT in the nanosecond (ns) pulsed regime but require cooling to about 150 K in the continuous wave (cw) regime.

Cr (or Fe) doped ZnS and ZnSe have very similar spectroscopic and laser parameters. Physical properties of ZnS, e.g. reduced thermal-optical effects and wider bandgap are advantageous in high power laser applications, while ZnSe has higher nonlinearity. The choice between ZnS and ZnSe hosts is usually defined by the specific laser parameter requirements. Both materials are available in single crystal and in polycrystalline forms. Single-crystals of high optical quality and sufficiently high dopant concentrations are difficult to grow. However, the technology of post-

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growth thermal diffusion doping of polycrystalline materials has enabled the fabrication of laser gain elements with high dopant concentration, uniform dopant distribution, and low losses. Furthermore, this technology is scalable and quantitative, i.e. it allows for mass-production of large-size gain elements with pre-assigned parameters, as illustrated in Fig. 1. As a result, polycrystalline materials have gradually replaced single crystals in most lasers.

Advances in fabrication of polycrystalline laser materials, availability of cost efficient fiber pump lasers at 1.5 – 2 µm and innovations in laser design has enabled rapid progress of Cr:ZnS and Cr:ZnSe lasers, as illustrated in Fig. 2. Recent achievements in technology include a RT cw Cr:ZnSe laser at 2.4 µm wavelength with power in excess of 140 W, and a few-optical-cycle ultra-fast Cr:ZnS oscillator with 2 W average power and sub-MW peak power.

Spinning-ring gain element technology

A breakthrough in cw laser parameters has been achieved thanks to the use of spinning-ring gain element technology [4]. This technique mitigates the thermal optical effects in the laser medium. Obtained preliminary results allow us to expect that 0.5 kW power level in cw spinning-ring laser will be reached in the near future.

Ultrafast mid-IR oscillators

Significant improvements in the output parameters of ultrafast mid-IR oscillators in terms of average power, pulse energy, and pulse duration were achieved due to the use of optimized polycrystalline gain elements and unconventional normal incidence mounting of the gain element in the optical resonator [5]. We rely on Kerr-lens mode-locking technique as it allows for shorter pulses, higher power in comparison with, e.g. SESAM mode-locked oscillators. The developed ultrafast oscillators design is rather flexible: we obtained few-optical-cycle MIR pulses at repetition rates ranging from 79 MHz to 1.2 GHz.

Fig. 3 illustrates the parameters of a polycrystalline Cr:ZnS oscillator, which was optimized for short pulse duration at 84 MHz repetition rate. As can be seen, the spectrum of pulses completely fills the atmospheric transparency window between 2.0 and 2.5 µm. Significant optical signal at 2.6 – 2.8 µm suggests that much broader spectrum can be obtained by purging of the optical resonator. The measured 26 fs pulse duration was limited by temporal broadening of output pulses during their propagation through the substrate of the output coupler. Most likely actual pulses are shorter about two optical cycles, considering 500 nm (27 THz) bandwidth of the spectrum at half-maximum.

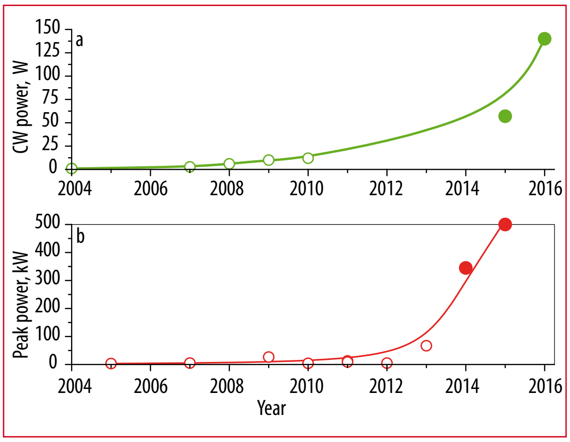

Fig. 2 The progress of room temperature MIR Cr:ZnS/ZnSe lasers in vicinity of 2.4 µm wavelength: power of cw lasers (a), peak power of femtosecond oscillators (b). The record parameters obtained to date correspond to (a) 140 W power with 62 % pump conversion efficiency in spinning ring laser; (b) 500 kW peak power in Kerr-lens mode-locked oscillator at 79 MHz repetition rate, 24 nJ pulse energy, 41 fs pulse duration. Graphics is based on the data published in references 2 and 3 and on the most current results obtained at IPG Photonics Corporation.

A set of interesting opportunities arises from TM:II-VI material's com-

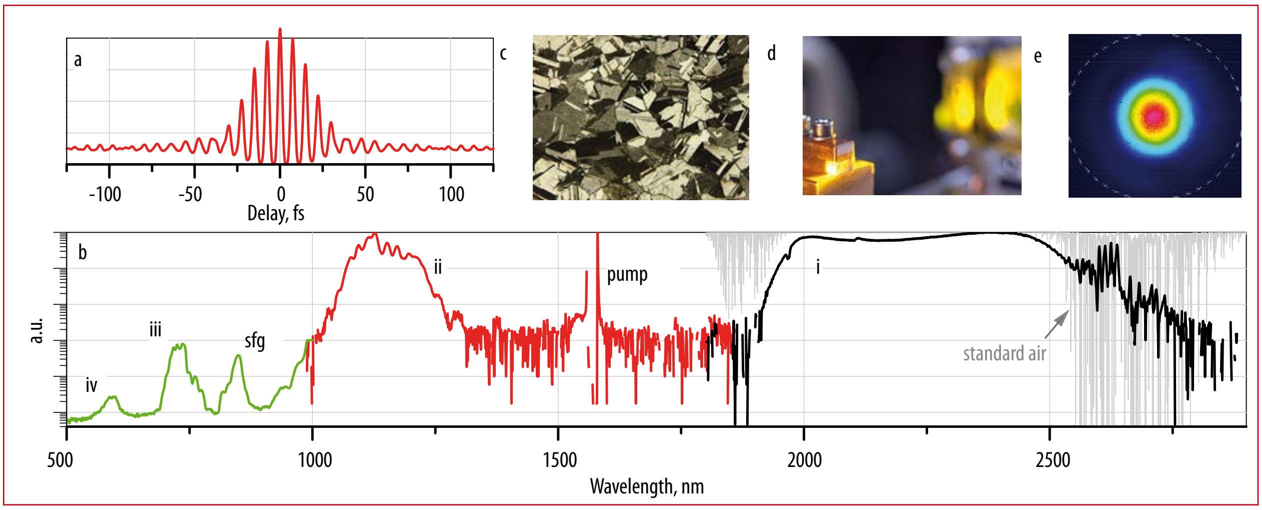

Fig. 3 TM:II-VI are the laser materials with a unique blend of parameters. Broad emission bands of Cr (and Fe) doped ZnS and ZnSe are favorable for generation of ultra-short pulses. High nonlinearity and polycrystalline structure of the material enable three-wave mixing directly in the gain medium of ultrafast laser via random quasi-phase-matching. The figure summarizes the parameters of ultrafast Cr:ZnS oscillator at 84 MHz repetition rate: autocorrelation (a), spectrum of pulses (b), microstructure of polycrystalline gain element (c), photo of the gain element of mode-locked laser (d) and output beam profile (e). The spectrum is presented in logarithmic scale including: (i) fundamental mid-IR band with 0.5 W power; (ii) SHG band with 0.1 W power, (iii) third harmonic; (iv) fourth harmonic; (sfg) sum frequency generation between fs mid IR pulses and cw pump radiation; residual pump at 1567 nm. Gray background represents transmission of standard air in 3 m long resonator. The fourth-harmonic emission at about 600 nm is sufficiently strong to be clearly visible to the naked eye as shown in the photograph (d).

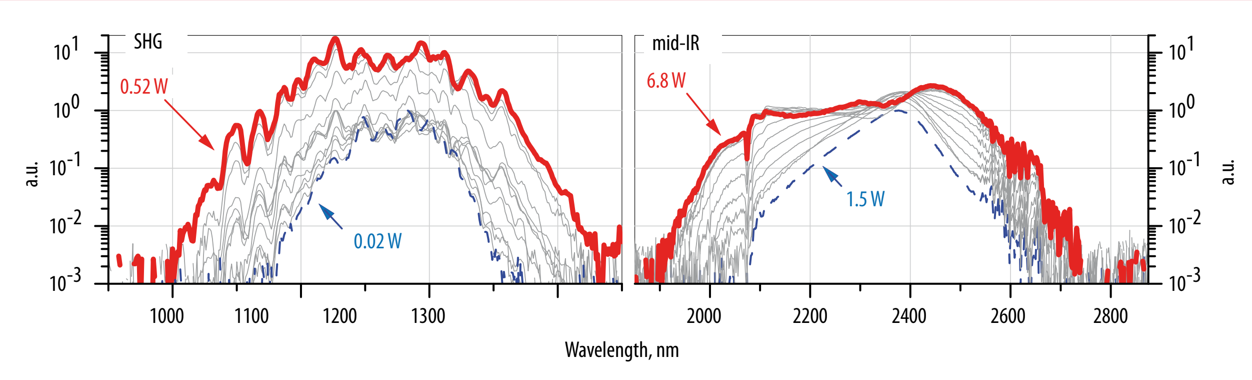

Fig. 4 Measured spectra of output pulses of ultra-fast single-pass Cr:ZnS amplifier at fundamental mid-IR (right) and SHG (left) wavelengths. Dashed lines: initial spectra obtained without optical pumping of the amplifier (normalized to unity); solid lines: the final spectra obtained at 22 W pump power (normalized to optical power); grey lines: intermediate spectra, obtained during the gradual increase of the pump power (all normalized to optical power).

bination of superb ultra-fast laser capabilities and their high nonlinearity and microstructure. Polycrystalline materials consist of a multitude of microscopic single-crystal grains. The broad distribution of grain sizes and orientations results in so-called random quasi-phase-matching (RQPM). According to a number of recent reports, the RQPM process is well suited for nonlinear conversion of spectrally broad fs pulses directly in the laser gain medium [5]. A significant fraction of the laser output can be converted to second harmonic (SHG). Apart from strong SHG we observe third and fourth optical harmonics as well as signals that correspond to the sum frequency generation between fs mid-IR pulses and cw pump radiation. SHG power levels of 0.33 W and 0.52 W were obtained in a Cr:ZnS oscillator and amplifier respectively, which corresponds to 6 – 8 % single pass conversion in 5 – 9 mm long polycrystalline samples.

A great demand for spectrally broad mid-IR sources with high average power, short pulse duration, and high repetition rate stimulate our efforts on power scaling of mid-IR oscillators. Single-pass fs laser amplifiers based on polycrystalline Cr:ZnS and Cr:ZnSe, as proposed in [6], are very appealing due to their simplicity, compactness and robustness. Recent improvements in the design of polycrystalline Cr:ZnS and Cr:ZnSe MOPAs has allowed us to realize multi-W mid-IR fs sources with three-optical cycle pulse duration [7].

Fig. 4 illustrates spectra of output pulses of the single-pass Cr:ZnS amplifier at fundamental mid-IR and SHG wavelengths. The amplifier was optically pumped by an off-the shelf cw erbium fiber laser. Initial spectra obtained without optical pumping are shown by dashed lines. Final spectra obtained at maximum pump power (22 W) are shown by thick solid lines. Numbers near the spectra correspond to measured average power. As can be seen, cw optical pumping of the single-pass amplifier results in simultaneous amplification of mid-IR fs pulses (like in conventional laser amplifier) and their spectral broadening. Remarkably, the spectra of pulses became broader with an increase of cw pump power (e.g. an increase of the amplifier's gain). Thus, gain narrowing in the amplifier is overwhelmed by the nonlinear interaction of pulses with the amplifier's gain medium. As a result, we obtained mid-IR pulses with 6.8 W average power and 450 nm broad spectra (at full width half maximum, FWHM); the spectral span is about 600 nm at −10 dB. Obtained autocorrelations show that amplification of mid-IR fs pulses was accompanied by a decrease of pulse duration from 40 fs to about 30 fs. Most likely, significantly shorter pulses can be obtained with better dispersion control of the amplifier, considering their 23 THz bandwidth. Conversion of low-cost cw pump radiation at 1.5 µm to few-cycle fs pulses was as high as 26 % (32 % of the absorbed pump power). There is little doubt that the output characteristics of the amplifier can be substantially further improved.

Summary and outlook

In summary, we demonstrate that MIR lasers based on Cr:ZnS and Cr:ZnSe have come of age, and are now arguably the most effective route to practical lasers in 2 – 3 µm spectral range. Availability of high power, reliable fiber lasers for optical pumping, improvements in the optical quality of the laser materials, and innovations in the laser design have allowed us to achieve 140 W cw laser power at 2.4 µm with the expectation of 0.5 kW level in near future. Another important achievement is high (in excess of 60 %) conversion efficiency of low-cost near-IR fiber laser radiation to tunable 2 – 3 µm MIR laser emission. It opens an avenue for the use of high power cw Cr:ZnS and Cr:ZnSe lasers in real-world applications, which require cost-efficiency.

We also demonstrate that Cr:ZnS and Cr:ZnSe laser media are very well suited for generation of ultra-short optical pulses in the 2 – 3 µm mid-IR range. Over the last few years, SESAM mode-locked fs lasers with ~100 mW average power and ~100 fs pulse duration were surpassed by robust Kerr-lens mode-locked oscillators with multi-Watt few-optical-cycle output. Femtosecond MIR lasers based on Cr:ZnS and Cr:ZnSe represent an appealing alternative to complex and inefficient ultra-fast mid-IR sources based on down-conversion of near-IR lasers, e.g. synchronously pumped optical parametric oscillators, optical parametric amplifiers and difference frequency generation setups.

Current research efforts include development of ultrafast MIR amplifiers with high pulse energy, and extension of ultrafast laser oscillations to the 3 – 10 µm spectral range. Another important direction of our work is further power scaling of ultrafast Cr:ZnS and Cr:ZnSe laser sources. The high gain of spinning-ring power amplifiers potentially enables femtosecond MIR Cr:ZnS and Cr:ZnSe MOPAs with an average power approaching the 100 W level.

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