Graphene mode-locked Cr:ZnS chirped-pulse oscillator

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Abstract: We report the first to our knowledge high-energy graphene mode-locked solid-state laser operating in the positive dispersion regime. Pulses with 15.5 nJ energy and 42 nm spectral bandwidth with 0.87 ps duration were obtained at 2.4 µm wavelength. The output can be compressed down to 189 fs. The graphene absorber damage threshold was established at fluence approaching 1 mJ/cm^2.

OCIS codes: (140.4050) Mode-locked lasers; (140.3070) Infrared and far-infrared lasers; (140.3580) Lasers, solid-state.

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1. Introduction

Monolayer graphene is a unique material possessing unique optical properties, allowing it to act as a wavelength-independent saturable absorber with very fast decay times, perfectly suitable to initiate the mode-locked operation of ultrafast solid-state lasers. Successful implementations range from 800 [1] to 2500 nm [2] on the wavelength scale. Pulses with durations down to 41 fs [3] have been obtained.

Power scaling of the ultrafast femtosecond crystalline oscillators has a natural limit, set by the soliton pulse breakup due to high third-order optical nonlinearity of the active medium. Increasing the output power over the certain limit results either in multiple-pulsing or harmonic mode-locking regimes. In order to overcome that, the intracavity power density inside the active medium should be reduced. That could be done by using the technique of chirped-pulse-oscillator (CPO or ‘dissipative soliton’) that allows reducing the intracavity pulse peak power, while retaining the possibility of extracavity recompression of the pulse back to sub-100-fs level. This technique is well-established in fiber [4] and Ti-sapphire lasers [5,6], and has recently been demonstrated with the Cr:YAG [7] and Yb-doped thin-disk lasers [8,9]. For Cr:ZnSe the analytical theory [10] predicts pulse energies up to 0.5 μJ [11] and initial demonstration of CPO technique has already been performed for Cr:ZnSe as well as Cr:ZnS lasers [12]. In the mid-IR, pulses with 8 nJ energy have been demonstrated recently in a KLM mode-locked Cr:ZnS laser [13]. This system was not self-starting and required certain readjustment during the power scaling, since the KLM technique is by itself very sensitive to fluctuations of the transverse mode due to thermal lensing, pump mode degradation, etc. Implementing of the graphene saturable absorber to such a system would allow unleashing the potential of the chirped pulse oscillator concept towards easy pulse energy scaling in the wide range.

2. Experimental setup

The basic experimental setup is shown in Fig. 1. We used the linearly polarized CW Er-fiber laser as a pump source. The laser provided up to 5 W of polarized pump emission at 1.61 μm. We have chosen the common X-folded astigmatically compensated four-mirror cavity as a basic cavity design concept. The nearly-symmetrical cavity had a total length of about 2.9 m. 2.5-mm thick passively-cooled Cr:ZnS active element was positioned at Brewster angle between the concave cavity mirrors M1 and M2 having 50 and 75 mm ROC, respectively. The cavity mode was additionally focused to graphene saturable absorber by ROC = 150 mm M3 concave mirror providing the mode diameter of about 120 μm. The compensation of the group-delay dispersion was achieved by a single reflection from chirped HR mirror resulting in a few hundred fs^2 of positive GDD per cavity roundtrip. The output coupler was set in a folding position in order to ensure only one reflection from the chirped mirror CM. The laser thus emitted two beams, namely Out1 and Out2, and the measured output power is a sum of both beams.

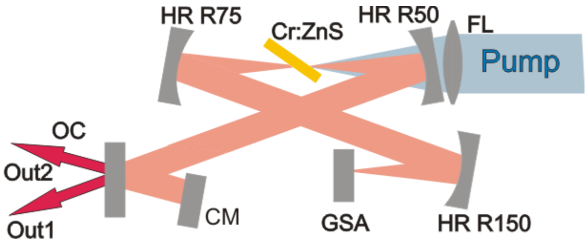

Fig. 1. Experimental setup of the graphene mode-locked Cr:ZnS CPO. FL is the pump focusing lens (f′ = 40 mm), HR are the highly-reflective concave mirrors, CM is the flat chirped mirror, GSA is the graphene-based saturable absorber mirror, OC is the output coupler.

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The commercially ordered CVD-grown graphene saturable absorber (Graphene supermarket®) has been deposited on the surface of broadband dielectric high-reflector mirror to form the graphene-based saturable absorber mirror, with single-, double-, and triple-layer structures deposited on the same mirror [3]. There have been few reports on similar absorber design [14,15]. Though positioned at the end of the laser cavity, the dielectric mirror consist number of layers with a graphene layer on the top, thus moving the absorber out of the electric field node. The calculated single bounce absorption of the device is 5.5%. The details on fabrication and properties of saturable absorber could be found elsewhere [3].

All measurements were performed in the open air, with relative humidity ~30%. The spectrum was analyzed by a commercial FTIR spectrometer at 1 cm^{-1} resolution. The pulse duration was measured using a home-made autocorrelator based on a two-photon absorption in an amplified Ge photodetector.

3. Results and discussion

3.1. Output characteristics

The laser provided stable self-started CW mode-locking with a build-up time around 500 μs. The input-output characteristics of the laser, pulse spectra and autocorrelation trace are plotted in the Fig. 2.

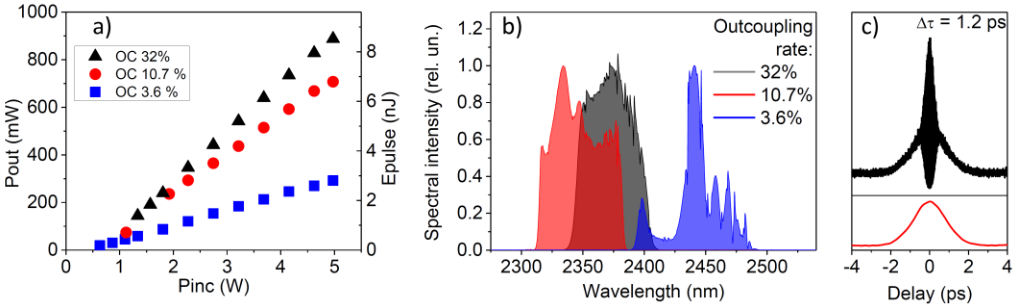

Fig. 2. (a) Input-output characteristics of graphene mode-locked Cr:ZnS chirped pulse oscillator for different output coupling rates. (b) Laser pulse spectra for different output coupling rates at the highest output power. (c) Typical interferometric (above) and intensity (below) autocorrelation trace of the laser pulse

Three output couplers were tested with transmittance of 1.8%, 5.5% and 17.5%, thus giving us total output coupling rate of 3.6%, 10.7% and 32%. The mode-locking could be achieved in the very broad range of output power and pulse energy with all the available output couplers. For output coupler transmittance of 3.6% the mode-locked output power could be varied from 15 to 290 mW (about 20 times) without any readjustment, and is still limited only by the pump power available. The highest achieved output power of 890 mW (32% output coupling rate) equals to 8.6 nJ of pulse energy. The broadest spectral bandwidth reaches almost 100 nm, while the spectra have the typical flat-top shape for chirped pulse oscillators. Pulse durations in the range of 1-3 ps were detected. The pulses are characterized by the natural chirp thus allowing extracavity compression down to sub-100-femtosecond range.

3.2. Pulse energy scaling

Chirped pulse oscillator concept allows quite simple pulse energy scaling. The cavity was extended to the total length of about 6.5 m corresponding to the repetition frequency of 46 MHz. Implementation of the HR mirrors with high radii and operation in the second stability zone allowed to form mode diameters of 220 μm and 45 μm on the passive shutter and active element, respectively. The expanded cavity is shown on Fig. 3.

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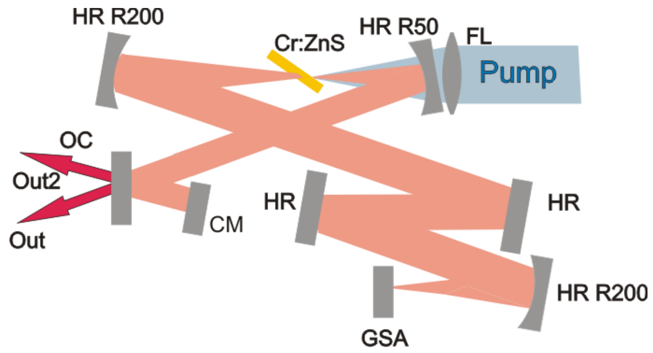

Fig. 3. Experimental setup of the graphene mode-locked Cr:ZnS CPO with expanded cavity. FL is the pump focusing lens (f′ = 40 mm), HR are the highly-reflective flat and concave mirrors, CM is the flat chirped mirror, GSA is the graphene-based saturable absorber mirror, OC is the output coupler.

With output coupling rate of 32% we reach 700 mW of average output power corresponding to about 15.5 nJ pulse energy. The pulses have 0.87 ps duration and spectral bandwidth of 42 nm (Fig. 4). The output power and pulse energy were limited due to switching to the chaotic mode-locking regime [16]. Further energy scaling up to 30 nJ is possible subject to implementation of the fine dispersion tuning [3] with the same pump source. Much higher pulse energies are feasible if the pump laser could be upgraded.

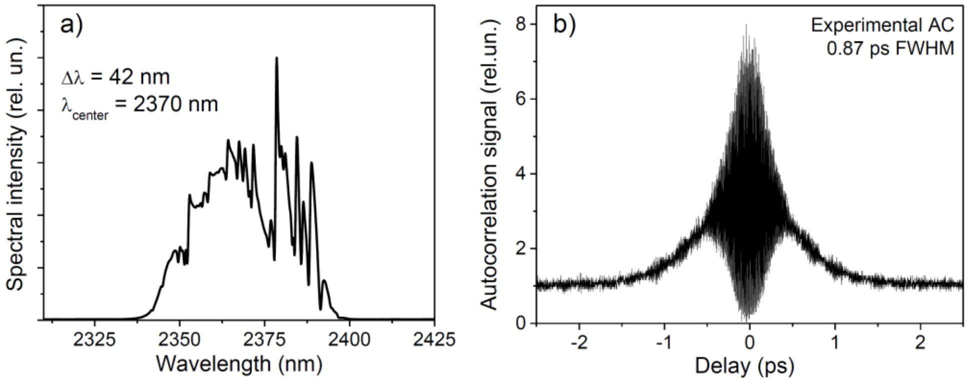

Fig. 4. (a) Laser spectrum of graphene mode-locked Cr:ZnS chirped pulse oscillator with extended cavity. The high-frequency modulation is due to the intracavity water vapor absorption in the atmosphere [17]. (b) Interferometric autocorrelation of laser pulse from such oscillator.

3.3. Pulse compression

An important property of a chirped-pulse oscillator is the possibility to compress the output pulses to much shorter duration by providing sufficient negative GDD after the output coupler. This compression should be possible by simple means and allow adjustment to match the actual chirp amount of the pulse. In our experiment, the compression was easily achieved by a pair of low OH content infrasil Brewster prisms with apex distance in the range of 10-90 cm (Fig. 5(a)). The shortest compression was achieved at an apex distance ~63 cm, resulting in 189 fs pulse duration (Fig. 5(b)). It can be seen from the Fig. 5., that extrapolating the red dots to zero apex distance gives about 650-700 fs duration, which is shorter than the actual output pulse duration of ~810 fs in that experiment. This is due to quite significant material dispersion of fused silica at this wavelength (–200 fs^2/mm), so that a round-trip material path

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of the order of 1-2 cm alone produces notable compression. To make the system even more compact, it is advisable to use the tandem prism pairs. This would provide four times shorter apex distance ~15 cm allowing fitting the compression unit into the laser housing.

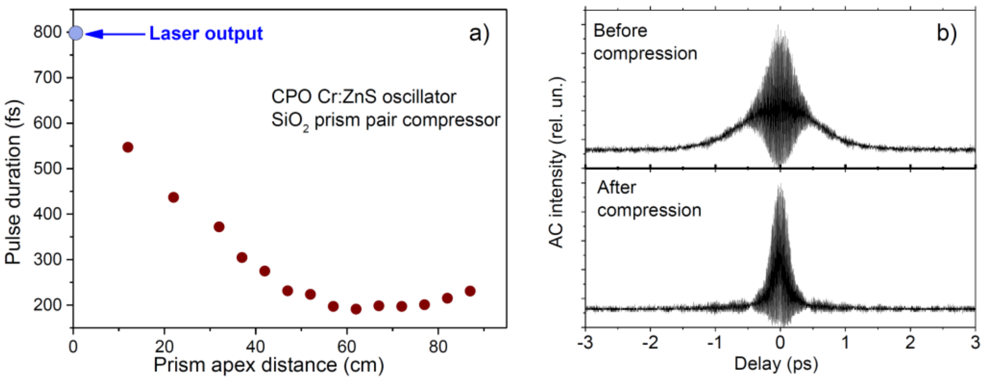

Fig. 5. (a) Extracavity compression of the CPO oscillator output. The pulse duration is estimated from the autocorrelation width assuming a sech^2 pulse form. (b) Interferometric autocorrelation traces of uncompressed and compressed pulses.

3.4. Graphene damage

Laser operation with high intracavity power requires special care to avoid damage of the saturable absorber. This is especially true for the chirped-pulse operation mode, when the pulse duration is significantly longer than the short decay time (100-200 fs) and comparable to the longer decay time of the graphene (1.5 ps) [1], meaning that a large part of the absorption cannot be saturated and that the absorber is subject to much higher thermal load as compared to the short-pulse system [3]. The cavity mode size has to be strictly controlled and scaled with increasing of the pulse energy. Particularly, operation of the extended cavity described above required adjusting the cavity to the second stability region in order to keep the mode diameter on the graphene mirror sufficiently large. If for some reasons the cavity was operated in the first stability zone, where the mode diameter is several times smaller, the energy density on the graphene reached several millijoules per cm^2, followed by the immediate damage of the graphene mirror.

Furthermore, during extended operation with energy density on graphene approaching 1 mJ/cm^2 (basic CPO cavity described in Section 3.1) we have observed gradual 5–10% increase of the laser output power. At the same time, the mode-locking would not self-start anymore, unless the graphene sample is shifted to a new position, indicating a certain damage to the absorber. The damaged spot could still support the mode-locked operation indefinitely long, when initiated by tapping the mirror. The damaged spot is visible as a low-density spot with diameter corresponding to the beam size (Fig. 6(a)). Raman spectra of the spot area (Fig. 6(b)) suggest that about 10-20% of graphene is still present in the damaged area.

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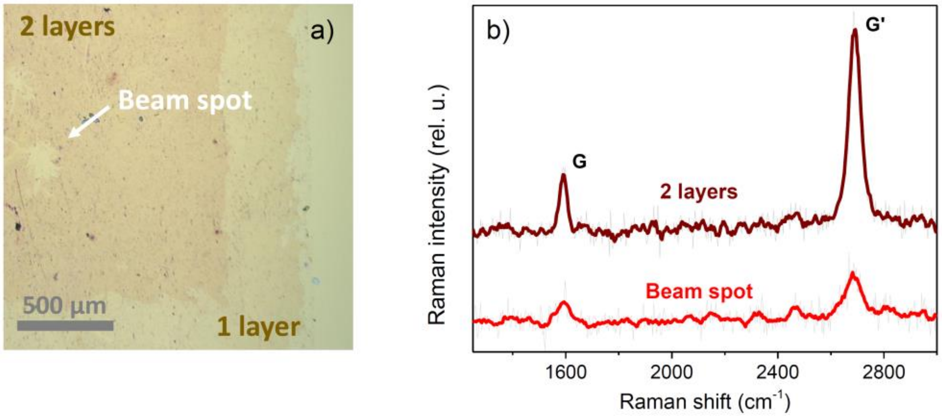

Fig. 6. (a) Photograph of the damaged beam spot (~120 µm diameter) in the double-layer region. (b) Raman spectra of the beam spot area and the double layer right near the spot.

4. Conclusion

Summarizing, we report the first graphene mode-locked Cr^{2+}:ZnS chirped-pulse oscillator and, to the best of our knowledge, the first solid-state dissipative soliton laser ever mode-locked by graphene. The adopted CPO technique allows scaling the laser pulse energy from 2.3 to 15.5 nJ in comparison with the traditional negative-dispersion design. The chirped pulse can be compressed from 810 to 189 fs by a simple and compact prism setup.

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