Graphene mode-locked Cr:ZnS laser with 41 fs pulse duration

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Abstract: We report the ultrashort-pulse Cr:ZnS laser mode-locked by graphene-based saturable absorber mirror. Using the combination of bulk material and a chirped mirror, we demonstrate the shortest reported so far mid-IR pulses of only 5.1 optical cycles (41 fs) centered at 2.4 µm with 190 nm spectral bandwidth. The pulse spectrum almost completely fills the water-free atmospheric window. The output parameters reach 2.3 nJ pulse energy and 250 mW average output power at 108 MHz repetition rate.

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

In the recent years, graphene has become a most sought-after saturable absorber for passive mode-locking of lasers. Compared to well-established saturable absorber techniques such as semiconductor quantum wells [1] graphene bears a promise of virtually infinite bandwidth, having a spectrally uniform saturable absorption, allowing reaching ultimately short pulse durations, and technologically simple and inexpensive manufacturing. With saturation fluences of tens of µJ/cm^2 and double decay time of 100-200 fs and ~1.5 ps for intraband electron scattering and optical phonon cooling, respectively [2], graphene becomes a convenient and extremely fast saturable absorber, with absorption equal to $\pi\alpha$ = 2.3% per pass per layer. In practice the absorption may differ from the theoretical value when graphene is doped, or forms bi-, tri- etc. layers. To the contrary, absorption of a stacked (randomly oriented) multilayer graphene should be a multiple of 2.3% per pass. Doping, causing *p*- or *n*-type conductivity would result in reduced absorption at the infrared edge [3]. Nevertheless, successful graphene-based femtosecond lasers have been demonstrated for virtually every configuration and in various spectral regions (a recent review can be found in [4], and new results keep arriving almost on a monthly basis). Practical implementations of graphene saturable absorbers (GSA) can take different forms, but for solid-state lasers with sizeable mode diameters, a CVD-based process [5] proved to be quite convenient. Current state-of-the art ranges from demonstration of the mode-locked pulses as short as 63 fs in a 480-mW Ti:Sapphire laser at 800 nm [2] to 176-fs pulses in a 185-mW Cr:ZnSe laser at 2500 nm [6, 7]. In this work we demonstrate a reflection-type GSA, with single- to triple-layer graphene deposited directly onto the high reflector end mirror of the cavity and using chirped-mirror dispersion compensation, making especially simple and compact cavity. A Cr:ZnS laser with this kind of GSA produces up to 250 mW of the shortest to-date generated pulses of 41 fs at 2.4 μm central wavelength.

2. Experimental setup

The experimental setup is shown in Fig. 1. The pump beam of the Er-fiber laser at 1.61 μm was focused by the 40-mm AR-coated lens to a 60x130-μm spot on the crystal. The X-folded near-symmetrical astigmatically compensated four-mirror cavity with round-trip length of about 2.8 m was implemented. Active element (2.5-mm-thick Cr:ZnS single crystal) was positioned at Brewster angle between the concave mirrors having 50 and 75 mm ROC. Graphene saturable absorber was deposited on the surface of the flat high-reflector mirror thus forming graphene-based saturable absorber mirror. The cavity mode was additionally focused to graphene saturable absorber by ROC = 200 mm concave mirror resulting in cavity mode diameter of about 150 μm on graphene. Active cooling was applied neither to active crystal, nor to saturable absorber. The set of output couplers with transmittance ranging from 1.8 to 18% were used in the experiments. The compensation of the group-delay dispersion (GDD) was achieved by the single chirped HR mirror.

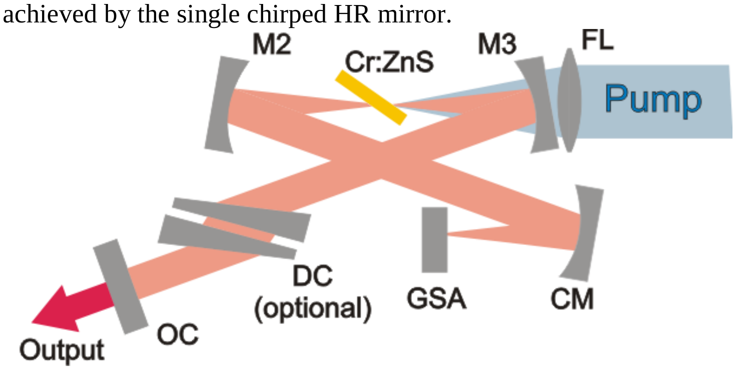

Fig. 1. Experimental setup of the graphene mode-locked Cr:ZnS laser. FL is the pump focusing lens (f′ = 40 mm), M2 and M3 are the highly-reflective concave mirrors, CM is the concave chirped mirror, GSA is the graphene-based saturable absorber mirror, DC is the YAG wedge pair, OC is the output coupler.

The commercially ordered graphene saturable absorber has been deposited on the surface of broadband dielectric high-reflector mirror by the copper-based CVD process in three stages, creating areas with 1, 2, and 3 layers of CVD graphene. Figure 2(a) shows a microscopic picture of a transferred graphene. A number of dark spots could be seen, especially on the 3-layer area, thus indicating contamination and slight non-uniformity of graphene. By comparing the relative intensities of the G and G’ lines in the Raman spectra

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(Fig. 2(b)) we can confirm the number and structure of the deposited layers [5]. As the shape and width of the G' peak does not change significantly with increasing number of layers, we may assume that the coatings are a superposition of monolayers and do not form bi- or trilayers [8], and that the optical properties of all coated areas are the same except initial absorption. The initial absorption was measured at the single wavelength of 2.38 μm corresponding to the emission wavelength of continuous-wave Cr:ZnS laser. The average absorbance of 9.8%, 18.2%, and 23.2% per bounce was measured for the absorbers consisting of 1, 2, and 3 layers of CVD graphene, respectively.

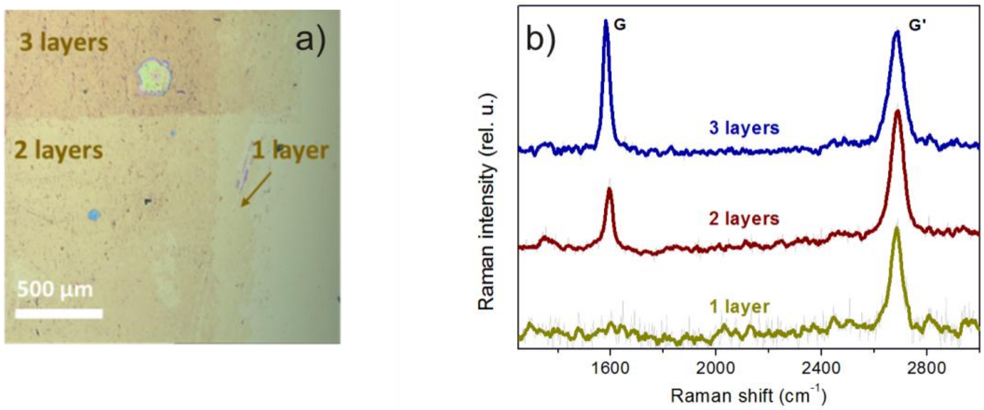

Fig. 2. (a) Photograph of the graphene saturable absorber mirror with 1, 2, and 3 graphene layers on a mirror surface. (b) Raman spectra, recorded in the three positions with different number of layers.

From the technical point of view, a high-reflector mirror with a graphene saturable absorber is in many respects analogous to a very thin semiconductor single quantum well saturable absorber, positioned at the top of a Bragg reflector structure [9]. Special care should be taken to the positioning of the absorber with respect to the nodes and antinodes of the light wave electric field (positioning of the graphene layer too close to the field node, like e.g. on a bare metal mirror, may result in very low absorption and too high saturation fluence). Figure 3 shows the calculated standing electrical field pattern in the vicinity of the HR mirror top layers for the central wavelength of the laser pulse. This calculation presumes single bounce absorption value of 5.5%, or 56% of the measured low signal absorption at 2380 nm. We attribute the rest of the absorption to the losses, introduced by the graphene transfer process.

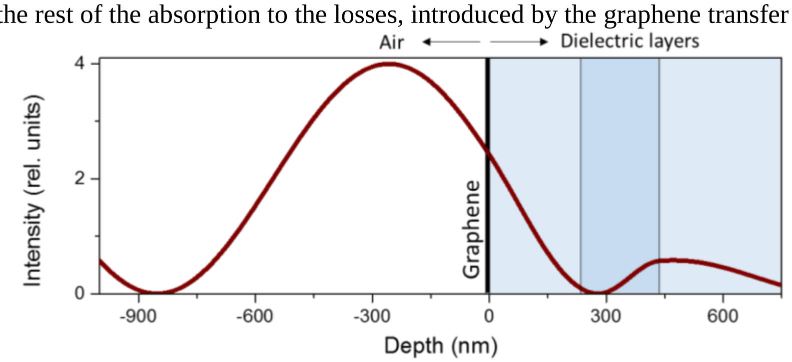

Fig. 3. Effective light field distribution for λ = 2380 nm near the HR mirror surface and in the surface layers. The black thick line denotes position of graphene absorber at the top of the upper layer, giving filed intensity of 2.4 normalized to the intensity of the incident wave.

All laser experiments 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

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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 best results were demonstrated with the SA consisting of 2 layers of graphene. With 18% output coupling the laser produced up to 250 mW of average output power on the cavity fundamental frequency of 108 MHz, providing the maximum pulse energy of 2.3 nJ. The pulse energy was limited by the pulse breakup due to the high third-order optical nonlinearity of the Cr:ZnS crystal. Up to 450 mW of output power could be obtained in multi-pulse regime (see Fig. 4(a)), but the pulse separation was uncontrollable in that case. While increasing the pump power, the laser spectral bandwidth increased, and the pulse duration shortened (see Fig. 4(b)). The pulse duration of around 50 fs and spectral bandwidth over 120 nm FWHM were reached near the pulse breakup limit. The pulse spectrum was centered at 2.37 μm where the calculated intracavity group-delay dispersion is about –400 fs^2. The laser was self-started in the output power range between 150 and 250 mW with the approximate mode-locking build-up time of about 500 μs. The laser input-output characteristics, laser spectrum, pulse autocorrelation trace, mode profile and mode-locking build-up oscilloscope trace are shown on Fig. 4.

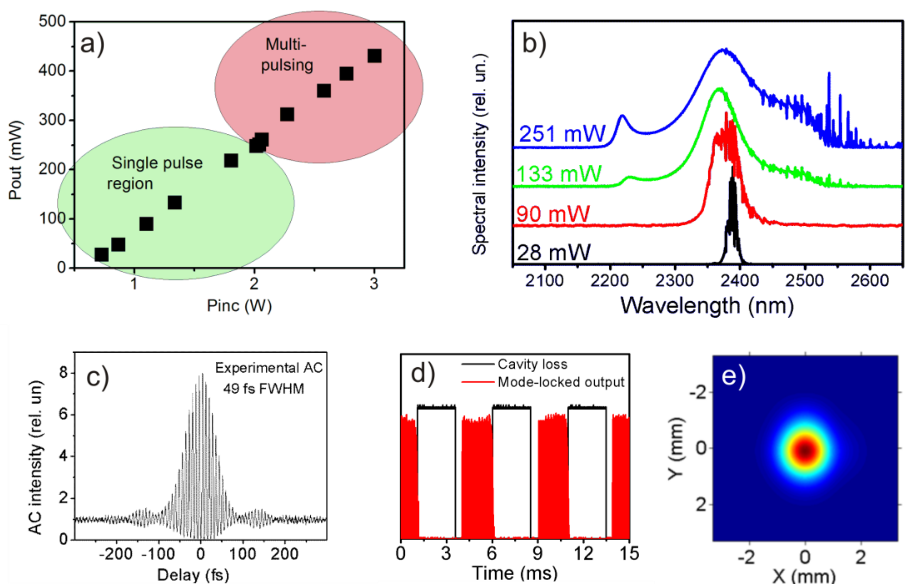

Fig. 4. Main characteristics of the graphene mode-locked Cr:ZnS laser with 18% output coupling. (a) input-output characteristics with single- and multi-pulsing regions shown, (b) evolution of the pulse spectrum while increasing the pump power; (c) interferometric autocorrelation trace of laser pulse at maximum output power in single-pulse regime; (d) self-starting behavior of the laser with optical chopper inside the cavity; (e) beam profile of the mode-locked laser output at maximum output power in single-pulse regime.

Three output couplers were tested in the experiments with the output coupling rates of 1.8%, 5.5% and 18%. For all three output couplers we obtained similar pulse durations and spectral bandwidths near the pulse breakup limit. The maximum output pulse energies of 0.23, 0.74 and 2.3 nJ, respectively, were demonstrated for three tested output couplers, all corresponding to the intracavity pulse energy around 13 nJ (see Table 1). Considering the calculated mode diameter of 150 μm on graphene-based mirror, the reached intracavity pulse

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energy corresponds to the energy density of about 150 μJ/cm^2 in graphene layer. We did not observe any degradation of the saturable absorber while operating in these energy limits.

The value of the graphene saturation fluence could be estimated from the threshold of the mode-locking regime. Analysis of the threshold conditions in different cavity configurations described in this paper resulted in a rather broad range of fluence values. This is due to some uncertainty of the exact mode size in the cavity with additional focusing, as well as non-uniformity of the graphene. Nevertheless it is certain that in some cavity configurations the mode-locking could be supported by the fluence around 15 μJ/cm^2.

Table 1. Main Laser Characteristics of Graphene Mode-locked Cr:ZnS Laser

OC transmission, %	Output power, mW	Repetition rate, MHz	Output pulse energy, nJ	Intracavity pulse energy, nJ	Spectral bandwidth, nm	Pulse duration, fs
18	251	108	2.32	13.3	125	49
5.5	80	108	0.74	13.5	122	54
1.8	25	108	0.23	12.9	115	55
1.8	75	240	0.31	17	190	42.8

3.2. Pulse duration

Reaching the shortest possible pulse duration required fine control on the intracavity dispersion. This was realized by a YAG thin wedge pair introduced at Brewster's incidence with variable insertion. Since YAG itself provides quite significant negative (anomalous) dispersion at this wavelength, we had to further reduce the number of reflections from the chirped mirror to a single bounce. The concave CM was substituted by the usual HR mirror with the same radius of curvature, and the OC was turned to folding position with flat CM installed as the cavity end-mirror. The total cavity length was decreased down to 1.25 m. to simplify purging and to decrease the intracavity water absorption. The cavity was encapsulated with a possibility to purge it with nitrogen.

With OC transmittance of 1.8% the laser output power in the modified cavity reached 75 mW resulting in 0.31 nJ of extracavity pulse energy at the pulse breakup limit. In terms of intracavity pulse energy, the limit was slightly increased, due to a bit wider calculated cavity mode inside the active element. After the experimental optimization of the intracavity GDD by adjusting the YAG wedge insertion, the laser pulses with a record duration of 42.8 fs and spectral bandwidth of 190 nm were obtained ($\Delta\nu\Delta\tau \sim 0.38$). A chirp-free pulse calculated from the measured output spectrum has the duration of 35.8 fs (FWHM). We assume that the extra 7 fs in the measured autocorrelation trace arises from the dispersion accumulated in the OC substrate (2 mm of YAG) and beamsplitter (1 mm ZnSe). The spectrum and autocorrelation trace of the shortest pulses, as well as the cavity round-trip GDD and atmospheric absorption, are plotted in Fig. 5.

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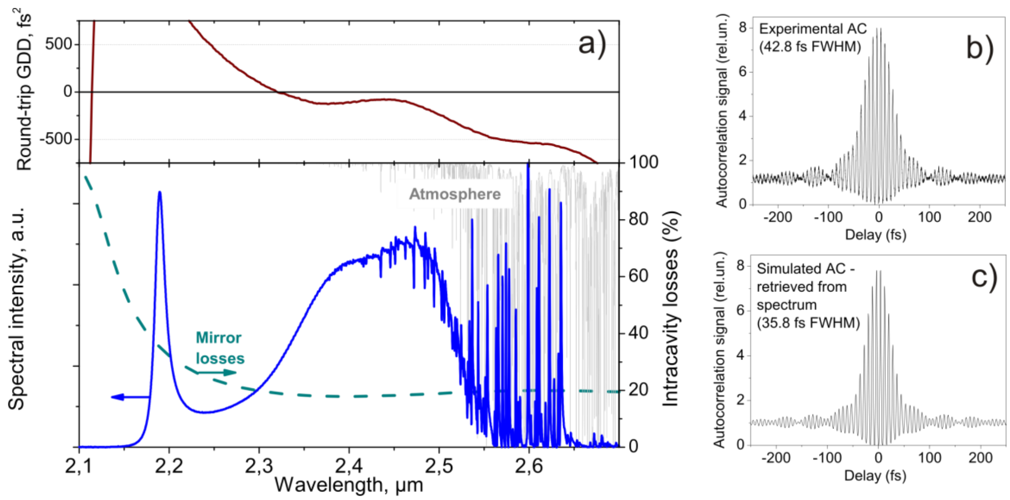

Fig. 5. (a) Laser spectrum of graphene mode-locked Cr:ZnS laser with advanced dispersion compensation, cavity round-trip GDD and atmospheric absorption; (b) Interferometric autocorrelation of the shortest laser pulses; (c) Simulated autocorrelation retrieved from the widest spectrum .

3.3. Nitrogen purging

All the spectra of the graphene mode-locked Cr:ZnS laser, especially one corresponding to the shortest pulse duration, contain some dispersion-like distortions on the long-wavelength wing. These are the traces of intracavity molecular absorption, particularly water vapor [10]. These features look rather intensive, though they contain a fairly small share of the pulse energy. It is nevertheless important, especially on the way to further pulse shortening, to define their influence on the pulse duration. Intracavity absorption could be eliminated by complete evacuation of the laser [11] or by purging the laser cavity with appropriate gas, e.g. nitrogen.

For our experiments we have put the folded setup into a 30x40x20 cm box. No special care was taken to isolate all the connections and pump and output beam ports, but after about one hour of nitrogen purging the relative humidity inside the cavity box could be reduced from 30% down to around 10%. The construction of the box did not allow any readjustments of the cavity while purging.

The experiment showed that though the intensity of the intracavity absorption lines of water vapor slightly reduced depending on the humidity, much more important was the change in the intracavity GDD. At the humidity lower than about 24% we repeatedly observed the significant change in the shape of the pulse spectrum, position of the dispersive wave and, in some cases, which resulted in the pulse breakup. The evolution of the spectrum on the relative humidity is shown in Fig. 6(a). At slightly reduced humidity level of about 20-25% the pulse duration decreased down to 41 fs, thus being two femtosecond (5%) shorter than without purging.

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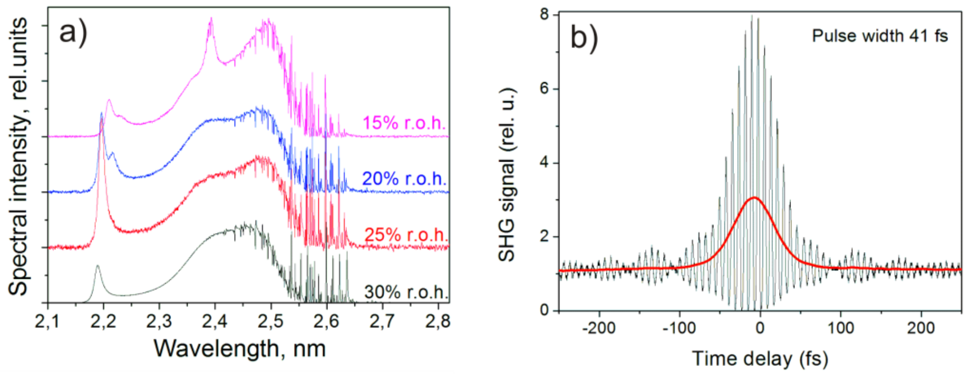

Fig. 6. (a) Spectra of the graphene mode-locked Cr:ZnS laser with nitrogen purging; (b) Interferometric autocorrelation of the shortest laser pulses at 25% r.h. At lower humidity the laser operated at too much intracavity energy, resulting in periodic switching to multi-pulse operation. The spectrum becomes a sum of a broad single-pulse spectrum and much narrower multiple pulse spectrum peak at 2.4 µm.

3.4. Cavity without focusing on saturable absorber

The important advantage of the graphene saturable absorber is its low saturation fluence in comparison with many quantum-well based SA. In order to demonstrate the potential of that characteristic we aimed to realize the mode-locking without any additional focusing of the cavity mode on saturable absorber. In this initial experiment the cavity was extended to over 8 m length to ensure the sufficient intracavity pulse energy (Fig. 7(a)) and designed to form the desired mode configuration on cavity elements. The cavity was operated in the second stability region, where comparatively dense cavity mode spot on graphene-based mirror could be obtained (270 µm in diameter) while keeping the cavity mode inside the active element rather large (60 µm). We also used a thinner Cr:ZnS active element with higher Cr^{2+} concentration to optimize the pump absorption efficiency.

At the cavity fundamental frequency of 36.8 MHz the average output power of 77 mW was obtained resulting in a pulse energy of 2.1 nJ outside the cavity and 11.6 nJ inside the cavity. The pulse energy was limited by the Cr:ZnS nonlinearity, just as in the previous experiments. The pulse duration of 96 fs and a spectral bandwidth of 45 nm were demonstrated (Fig. 7). The energy density on graphene-based saturable absorber mirror was estimated to be 40 $\mu J/cm^2$ near the pulse breakup limit and around 15 $\mu J/cm^2$ near the mode-locking threshold.

The described result is just the initial demonstration of the passively mode-locked Cr:ZnS laser without additional focusing on the passive shutter, and the laser has a great potential for improvements. The ratio of the cavity length to cavity mode diameter in the active element is far from optimal resulting in the pulse break-up limit appearing at power level <10% of the total available output power. Subject to proper cavity configuration, the simple and reliable watt-level femtosecond Cr:ZnS laser with graphene saturable absorber just deposited on the HR cavity mirror is feasible.

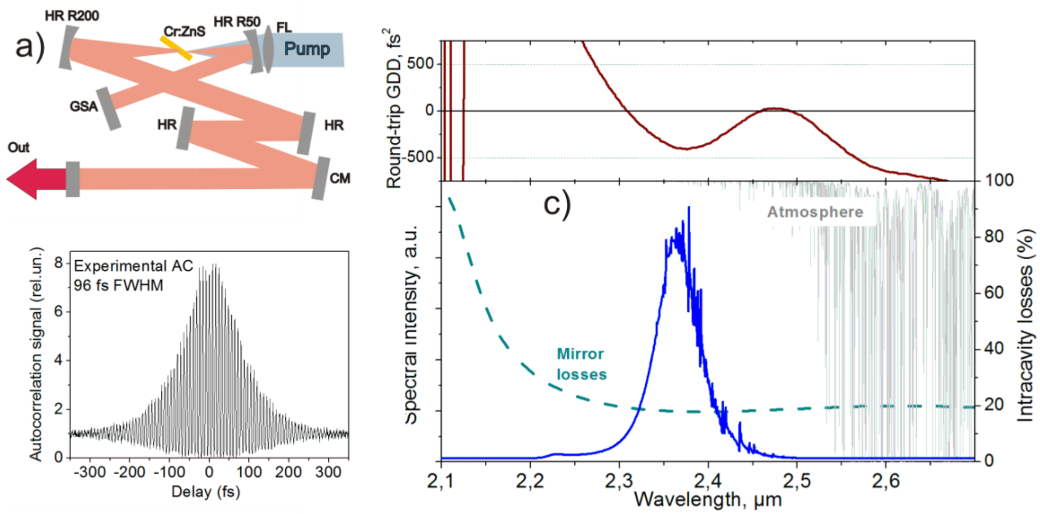

Fig. 7. (a) The cavity setup of the graphene mode-locked Cr:ZnS laser without additional focusing on the graphene-based mirror; (b) Interferometric autocorrelation of the laser pulses; (c) Laser spectrum and cavity round-trip GDD.

4. Conclusion

Summarizing, we report the first graphene mode-locked Cr^{2+}:ZnS laser. The laser generated pulses of 41 fs duration at the central wavelength near 2.4 µm, which corresponds to just over five optical cycles. To the best of our knowledge, these are the shortest pulses generated in the mid-IR, as well as the shortest pulses obtained with the graphene saturable absorber to-date at any wavelength. Under these conditions, the laser reaches the limits set by the water vapor absorption in the atmosphere and nearly completely fills the water-free window.

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