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OSA CONTINUUM

Graphene mode-locked Tm,Ho-codoped crystalline garnet laser producing 70-fs pulses near 2.1 µm

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Abstract: Bilayer graphene synthesized by chemical vapor deposition is successfully applied as a saturable absorber (SA) for the passive mode-locking of a Tm,Ho:CLNGG laser at 2093nm. Near transform-limited pulses as short as 70 fs, i.e., 10 optical cycles are produced at a 89 MHz repetition rate with 69 mW average output power. To the best of our knowledge, these are the shortest pulses ever reported from graphene-SA mode-locked Tm, or Ho-lasers in the 2 µm spectral region, including bulk and fiber lasers.

1. Introduction

Mode-locked ultrafast lasers in the 2 µm spectral region are actively investigated due to their promising applications in various fields such as high-field physics [1], transparent materials processing [2], medicine and military [3], etc. Moreover, such laser sources can be used for producing optical frequency combs with f_{CEO} stabilization [4], pumping optical parametric oscillators for generation of ultrabroadband frequency combs in the mid-IR spectral region [5], or seeding of near-degenerate chirped-pulse optical parametric amplifiers [6]. Present laser amplifier systems for generation of high pulse energies tend to use Ho-doped gain media, mainly Ho:YLF and Ho:YAG [7–9]. However, these amplifier systems suffer from rather complex seed sources. The latter contain typically a number of nonlinear conversion stages to generate the required femtosecond pulses with broad seed spectrum, i.e., at 2050 nm for Ho:YLF or at 2090 nm for Ho:YAG. One approach for a more straightforward realization is to apply femtosecond Ho-doped fiber oscillators as seed [10,11]. Compared to femtosecond fiber oscillators, mode-locked bulk lasers possess a much lower excess noise arising from the unwanted intracavity nonlinearities

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and amplified spontaneous emission [12]. An alternative femtosecond seed source solution are Tm,Ho-codoped disordered crystals. To date, numerous Tm^{3+} and/or Ho^{3+} doped laser materials have been investigated. Those of them exhibiting multi-site structure are one of the most interesting gain media for femtosecond pulse generation due to the inhomogeneous broadening of their emission spectra [13–16]. Tm^{3+}-doped [15] or Tm^{3+}, Ho^{3+}-codoped [16] disordered multicomponent garnets $Ca_3(LiNbGa)_5O_{12}$ (shortly CLNGG) with Li^+ cations serving for charge compensation are prominent examples for generation of sub-100 fs pulses at ~2 µm.

The other optical element which is crucial for initiating and stabilizing the mode-locked operation in femtosecond solid-state lasers is the saturable absorber (SA). Graphene features low saturation fluence, high optical damage resistivity, easy fabrication and low cost [17–19]. The gapless band structure and linear energy dispersion of the charge carriers give rise to a nearly constant absorption of ~ 2.3% per layer over a broad wavelength range extending from 0.7 to 25 µm [20]. The electron-hole interband recombination process in graphene occurs on a time scale of 1-2 ps, while the intraband carrier-carrier scattering exhibits a faster dynamics with a relaxation time around 100 fs [21]. The ultrafast recovery time of graphene renders it suitable for generation of optical pulses in the femtosecond regime. With a graphene-SA (GSA), sub-20-fs pulse generation was reported very recently from a mode-locked transition-metal Cr^{3+}:LiSAF laser near 850 nm [17], and 30 fs pulses at a central wavelength of 1068 nm were produced from a rare-earth Yb^{3+}:$CaYAlO_4$ laser [18]. In the longer wavelength range above 2 µm, 41-fs pulse at ~ 2.4 µm have been generated from another transition-metal Cr^{2+}:ZnS laser pumped by a high brightness Er-fiber laser at 1.61 µm [19]. In comparison, GSA mode-locked lasers employing Tm^{3+} or Tm^{3+}, Ho^{3+} rare-earth doped materials that can be directly pumped by commercial diode lasers at ~ 800 nm, generated much longer pulse durations. Figure 1 shows recent progress of femtosecond Tm- and Ho- lasers mode-locked by GSAs [10,11,13,22–26]. With the exception of the 86-fs pulses achieved using a GSA mode-locked Tm:MgW ($MgWO_4$) laser [13], the reported durations are around or above 200 fs [10,11,22–26].

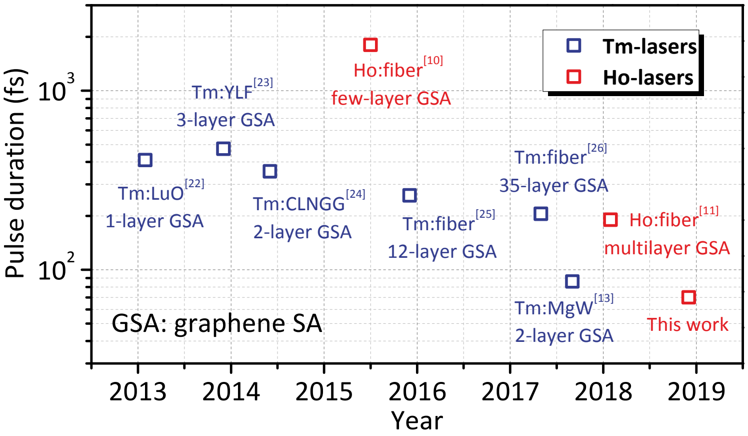

Fig. 1. Progress of GSA mode-locked femtosecond Tm- and Ho-lasers: LuO: Lu_2O_3, YLF: $LiYF_4$, MgW: $MgWO_4$.

In this letter, mode-locking of disordered Tm^{3+}, Ho^{3+} co-doped CLNGG laser is demonstrated by employing a bilayer GSA, the first GSA mode-locked bulk Ho-laser. Pulses as short as 70 fs at 2093 nm, i.e., 10 optical cycles, are generated, which to the best of our knowledge is the shortest pulse duration ever reported from GSA mode-locked Tm- or Ho-lasers at 2 µm, including bulk and fiber oscillators.

2. Experimental details

Figure 2 shows the scheme of the GSA mode-locked Tm,Ho:CLNGG laser. The 796 nm pump beam emitted from a continuous-wave (CW) Ti:sapphire laser was focused by a lens ($f = 70$ mm) into the crystal with a pump spot radius of 30 µm. The Tm^{3+} (2.34 at.%), Ho^{3+} (0.54 at.%) co-doped CLNGG crystal had a size of $3 \times 3 \times 6$ mm^3 with both end faces (3×3 mm^2) antireflection (AR)-coated. To mitigate the thermal load, the sample, tightly mounted in a copper holder, was water cooled to 14.0°C. M_1 and M_2 are plano-concave mirrors both with radius of curvature (RoC) of –100 mm. CM_1 - CM_4 are chirped mirrors (Layertec GmbH, Germany) providing –125 fs^2 group delay dispersion (GDD) per bounce at ~ 2 µm. The bilayer GSA was placed at Brewster's angle in the position of the second cavity waist formed by CM_1 (RoC = –100 mm) and CM_2 (RoC = –50 mm). The beam radii on the GSA were around 110 µm in the sagittal and 160 µm in the tangential plane. The output couplers (OCs) are plane-wedged mirrors with transmission of 0.5%, 1.5%, and 3%. A 3-mm thick ZnS plate with ~ +150 fs^2/mm group velocity dispersion (GVD) at 2.1 µm was employed for external compression.

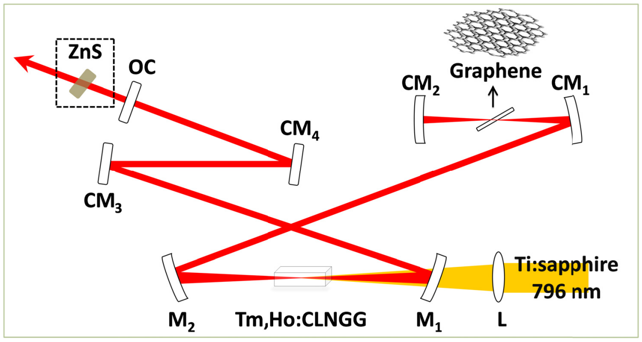

Fig. 2. Scheme of the GSA mode-locked Tm,Ho:CLNGG laser: L, lens; M: dichroic mirror; CM, chirped mirror; OC, output coupler.

The bilayer GSA was fabricated by stacking layer-by-layer, where the monolayer graphene was initially synthesized on a Cu foil by chemical vapor deposition employing a mixture of methane and hydrogen gases [27]. Then the Cu foil was etched by aqueous $FeCl_3$ solution after spin-coating of polymethyl-methacrylate (PMMA) on the graphene side, subsequently, transferring the graphene layer onto the surface of a 1-inch uncoated CaF_2 substrate (2-mm thickness) and finally removing the PMMA layer using acetone. The single-pass transmission was measured to be ~ 4.2% at 2.1 µm. Given the GVD in the crystal and the CaF_2 plate, the total intracavity round trip GDD was calculated (see Fig. 3(a)) and it amounts to ~ –1500 fs^2 around 2.1 µm.

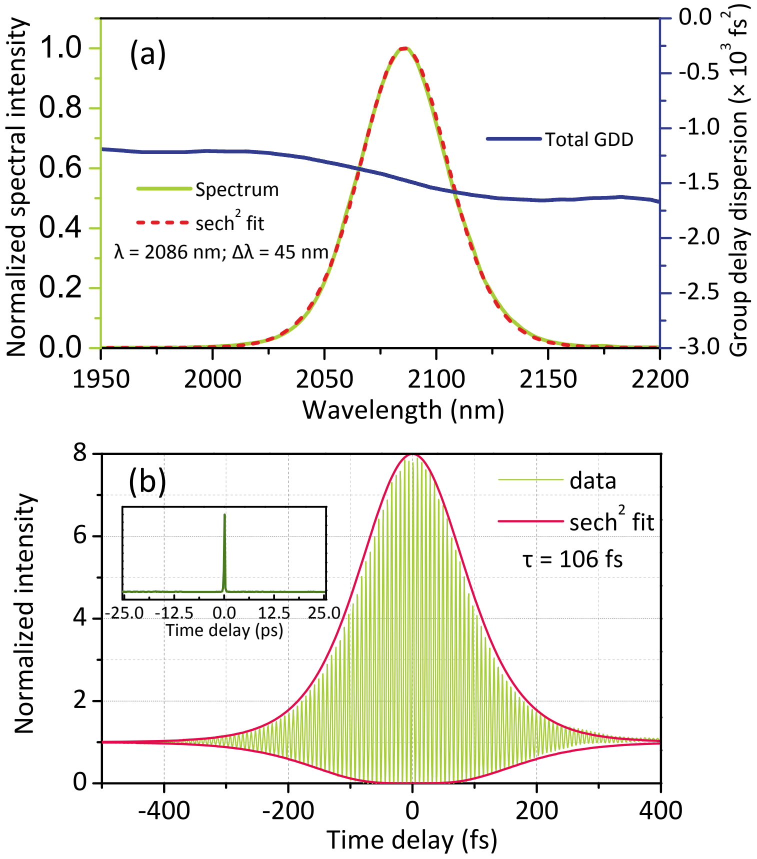

Fig. 3. Optical spectrum and calculated total round trip GDD (a) and interferometric autocorrelation trace (b) of the GSA mode-locked Tm,Ho:CLNGG laser (OC: 1.5%), Inset: corresponding noncollinear autocorrelation trace recorded in a wide-range of 50 ps.

3. Results and discussions

With a total cavity length of ~1.7 m, mode-locking performance was firstly studied by employing a 3% OC. By comparing the slope efficiencies with that of the empty cavity [16], the insertion loss of the GSA was estimated to be 9%. The lasing and mode-locking thresholds were 0.79 W and ~1.4 W, respectively. At the maximum absorbed pump power of 2.1 W, 94 mW average output power was obtained at a pulse repetition rate of ~ 89 MHz, yielding a pulse energy of ~ 1 nJ and a spatially averaged intracavity fluence of ~ 70 µJ/cm^2 on the GSA. The FWHM of the optical spectrum (centered at 2078 nm) amounted to 12.7 nm and the corresponding pulse duration was 370 fs measured by noncollinear second-harmonic generation (SHG) with a commercial autocorrelator (APE Berlin). Thereafter an OC with lower transmission (1.5%) was employed with the aim of generating shorter pulses. Figure 3(a) shows the recorded optical spectrum and its sech^2 fit. The spectral FWHM is 45 nm with a central wavelength located at 2086 nm. The corresponding interferometric autocorrelation trace is shown in Fig. 3(b). The accurate 8:1 peak-to-background ratio and the nearly perfect sech^2-fit of the envelope profiles indicate chirp-free pulse generation [28]. The pulse duration amounted to 106 fs, corresponding to a time-bandwidth product (TBP) of 0.329, which means the pulse duration is very close to the Fourier-transform limit (FTL). In this case the average energy fluence on the GSA was calculated to be ~ 120 μJ/cm^2. Single pulse operation without satellites was confirmed by recording the intensity autocorrelation trace on a long time scale of ± 25 ps, see the inset of Fig. 3(b).

A 0.5% OC was subsequently used for mode-locking with expectation for further pulse shortening. The average output power dropped to 69 mW at the maximum pump level (2.1 W of absorbed power), corresponding to a spatially averaged intracavity fluence of ~ 280 μJ/cm^2 on the GSA. The recorded optical spectrum is shown in Fig. 4(a), the central wavelength is located at 2093 nm. The sideband observed at longer wavelengths is considered to be an artefact due to the increasing transmission of the OC (i.e. 30 times higher at 2205 nm). The fit with the sech^2 function resulted in a spectral FWHM of 69 nm. The corresponding pulse duration is 81 fs by assuming a sech^2-intensity profile, giving a TBP of 0.382. The larger value compared to the FTL indicates the presence of residual chirp. Therefore, a 3-mm-thick ZnS plate, as shown in Fig. 2, was used for external compression. In this way the linear chirp provided by the ZnS plate is much weaker compared to the contribution of the intracavity elements which are multiply passed by the pulse depending on the photon lifetime in the cavity. Since the measured pulse spectrum remained unaffected by the plate, nonlinear effects in the ZnS plate could be ruled out as could be expected without tight focusing of the output laser beam.

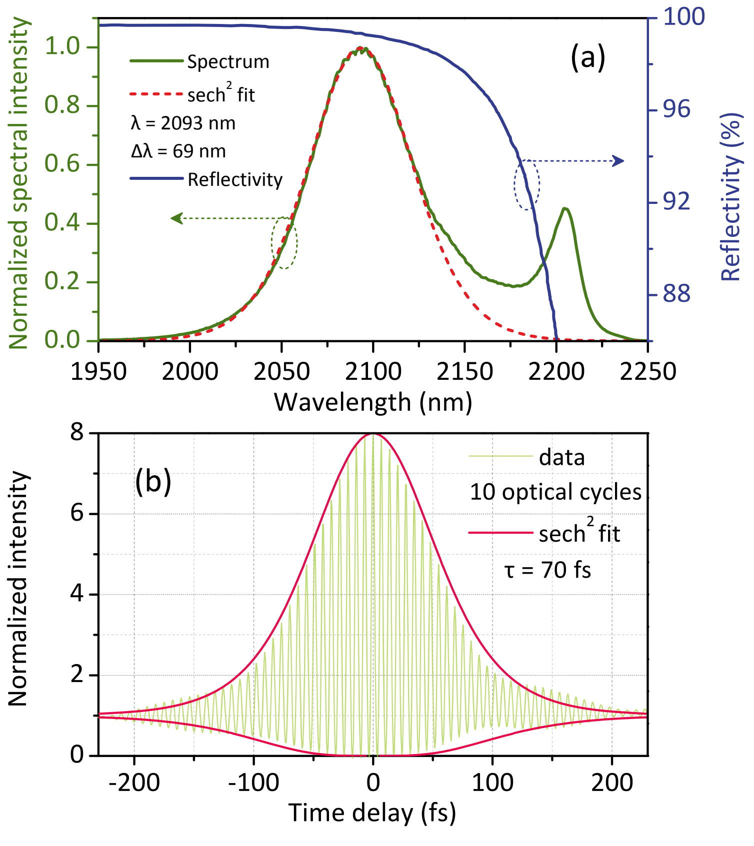

Fig. 4. Optical spectrum (a) and interferometric autocorrelation trace (b) of the GSA mode-locked Tm,Ho:CLNGG laser with external compression (OC: 0.5%).

Figure 4(b) shows the recorded interferometric autocorrelation trace after external compression. Deriving from the sech^2-fit, the pulse duration amounts to 70 fs, corresponding to 10 optical cycles judged from the fringe-resolved interferometric autocorrelation trace. To the best of our knowledge, this is the shortest pulse ever reported from GSA mode-locked Tm or Ho- lasers at 2 µm, including bulk and fiber lasers. The corresponding TBP is 0.331, thus the pulse duration is close to the FTL. The TBP value reduction is solely attributed to the pulse shortening in ZnS, for comparison, calculations show that an unchirped sech^2 pulse of 70 fs duration broadens to 77 fs after passing an element with GDD of ± 460 fs^2. The spectral feature in Fig. 4(a), although designated as an artefact since it can be easily suppressed by an optimized mirror design, is in fact an integral part of the mode-locked laser spectrum. The spectrum of the circulating intracavity pulse is suppressed from the long wave side at each reflection from the OC. Thus, the tiny deviations of the temporal characteristics from the perfect sech^2-fit and the TBP from the 0.315 value is attributed to this spectral feature.

Mode-locking in this case was not self-starting, hence an extra perturbation was required. Once mode-locking was initiated, it remained stable for hours. However, pure Kerr-lens mode-locking was not achieved without the usage of GSA or substituting it with a 1-mm-thick quartz plate. Thus, we attribute the generation of such short pulses partially to Kerr lensing effect under tight mode focusing conditions with increased intracavity pulse energy [29], meanwhile, the broadband GSA was a critical factor for initiating and stabilizing the mode-locking operation.

To characterize the stability of the GSA mode-locked Tm,Ho:CLNGG laser in the regime with the shortest pulses, we measured the radio frequency (RF) spectra. The fundamental beat note recorded on a 300 kHz span with 100 Hz resolution bandwidth (RBW) is shown in Fig. 5(a). The

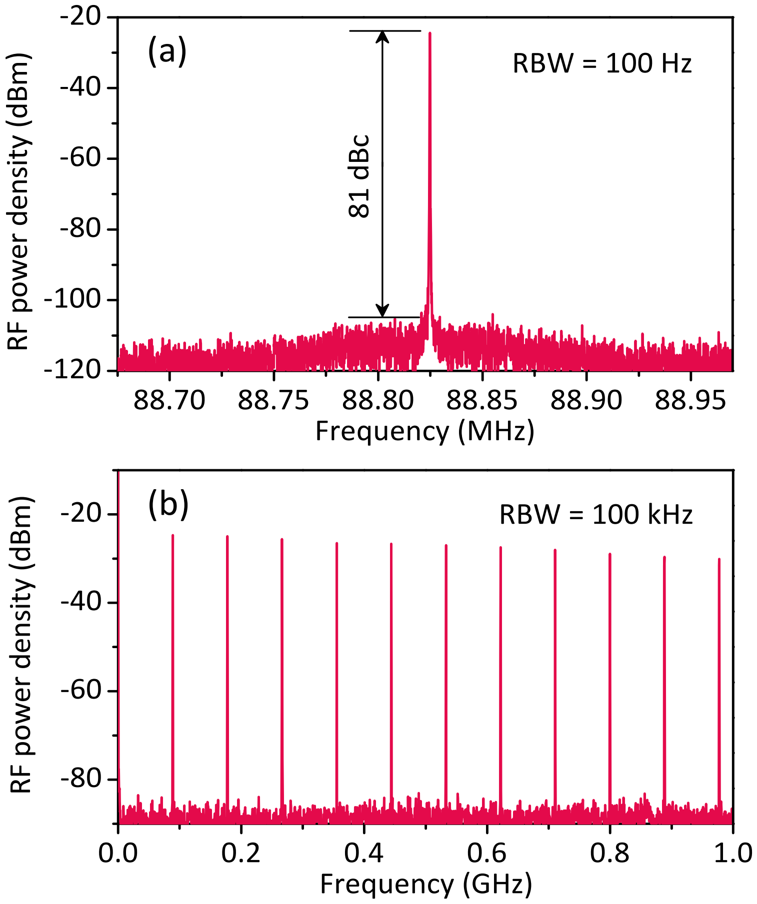

Fig. 5. RF spectra of the GSA mode-locked Tm,Ho:CLNGG laser with external compression: (a) fundamental beat note and (b) 1 GHz span. RBW: resolution bandwidth.

high extinction ratio of 81 dBc against the noise indicates an excellent stability of the steady-state mode-locking. Moreover, as can be seen in Fig. 5(b), the uniform harmonic beat notes recorded on a 1 GHz span are a further indication of stable and clean steady-state mode-locking without any multi-pulse behavior [30].

4. Conclusion

In conclusion, we have experimentally demonstrated stable mode-locking of a Tm,Ho:CLNGG laser by using a bilayer GSA. 94 mW average output power was achieved with a 3% OC, corresponding to a single pulse energy of ~ 1 nJ at ~ 89 MHz repetition rate. With a 1.5% OC, almost chirp-free pulses with 106 fs duration were generated. The pulses were further shortened to 70 fs by applying a 0.5% OC and external single-pass compression through a 3-mm-thick ZnS plate. The optical spectrum of the shortest pulse was centered at 2093 nm with a FWHM of 69 nm, corresponding to 10 optical cycles. The broad spectrum was facilitated by the broadband modulation provided by the GSA, the smooth and broad gain bandwidth above 2 µm of Tm,Ho:CLNGG, a proper intracavity GDD management and partially the Kerr lensing effect. Optimizing the number of the graphene layers and compensation of the residual 3rd order GDD will constitute the future work towards further pulse shortening. On the other hand, pure Kerr-lens mode-locking would be expected by enhancing the self-phase modulation (SPM) and the optimization of the overall GDD.

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