7.8-GHz Graphene-Based 2-μm Monolithic Waveguide Laser

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***Abstract*—We report a pulsed waveguide laser working at 1944 nm, mode-locked with a saturable absorber consisting of a graphene film deposited on an output coupler mirror. The waveguide is created into a ceramic Thulium-doped Yttrium Aluminium Garnet by ultrafast laser inscription. Q-switched mode-locking is achieved, with 6.5 mW average output power and ~7.8 GHz pulse rate. This is a convenient, compact, high repetition rate laser for various applications, such as medical diagnostics and spectroscopy.**

***Index Terms*—Graphene Saturable Absorbers, Laser made-locking, Laser applications, Optical waveguides, Solid lasers.**

I. INTRODUCTION

GRAPHENE and carbon nanotubes (CNTs) have emerged as promising saturable absorbers (SAs) for ultrafast laser development. In CNTs, broadband operation is achieved by using a diameter distribution [1], while it is an intrinsic property of graphene [2]. This, along with the ultrafast recovery time [3], [4], and low saturation fluence [5], [6], makes graphene an excellent broadband SA [5]–[12]. Passively Q-switched and mode-locked lasers using CNT and graphene SAs have been demonstrated for a wide spectral range [1], [5]–[10], [12]–[21]. A regime of Q-switched mode-locking (QML) was also demonstrated using graphene based SAs [9]. In QML, the laser output consists of passively mode-locked pulses underneath a Q-switched envelope [22]. In spite of the Q-switching tendency, the high energy of the mode-locked pulses has potential applications in nonlinear frequency conversion [23] and surgery [24].

Tm^{3+} doped solid-state lasers operating in the 2 μm spectral range are of great interest for applications such as medicine [24], material processing [25], and environmental monitoring [26]. The operating wavelength is important because water, the main constituent of human body [27], absorbs more at 2 μm ($\sim$100 cm^{-1}) than at other conventional wavelengths, i.e., $\sim$1.5 μm ($\sim$10 cm^{-1}) and $\sim$1 μm ($\sim$1 cm^{-1}) [28]. Furthermore, gas molecules, such as CO_2, show characteristic absorption lines [26], making 2 μm lasers promising for industrial process monitoring [28] or environmental control [26]. The possibility of ultrafast operation with multi-GHz repetition rates at this wavelength is opening new application avenues, such as pumps for mid-infrared frequency combs [29]. The pre-requisite of a short cavity length for high repetition rate operation can be achieved by using a waveguide cavity configuration. In a waveguide, the pump and laser modes are tightly confined within the waveguide core, facilitating a lower lasing threshold and improved slope efficiency [7], [30], [31]. It inherently guarantees good beam quality [31] and a stable cavity construction. Also, waveguide cavities allow easy incorporation of SAs within the integrated cavity to facilitate efficient pulsed operation [7], [30].

A simple and flexible waveguide fabrication technique is ultrafast laser inscription (ULI) [32]. ULI employs fs pulses focused beneath a substrate to induce material modifications by virtue of nonlinear absorption processes at the focus. Translation of the substrate along any arbitrary path extends this modification to create a waveguide [33]. Mode-locked ULI waveguide lasers were demonstrated at 1.5 μm using CNT-SAs [15], [30] and at 1 μm with graphene-SA (GSA) [7].

A variety of techniques have been implemented in order to integrate GSAs into lasers [34]. GSAs have been used to mode-lock lasers over a wide-spectral range [34]. E.g., at 2 μm, 1–2 layers of graphene chemical vapor deposited (CVD) [35]–[37] or grown by carbon segregation on SiC [38] were used for mode-locking [35], [36] or Q-switching [38] of solid-state lasers. For mode-locking of Thulium-doped fiber lasers, graphene polymer-composites prepared by liquid phase exfoliation (LPE) of graphite [39] were used [8]. Graphene oxide (GO) films were also used for mode-locking of solid-state lasers [40]. However, GO is fundamentally different from graphene: it is an insulating material with many defects and gap states [41], and may not offer the wideband tunability of graphene [2]. CVD and carbon segregation from SiC require high substrate temperatures [35]–[37], [41], followed by transfer [35]–[37], [41]. LPE has the advantage of scalability, room temperature processing and high yield, and does not require any substrate [41]. Dispersions produced by LPE can easily be embedded into polymers and

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integrated into various systems [2], [41]. LPE graphene can also be used as a film [7]. This reduces non-saturable losses, allowing high average-power.

Here we report QML at 1.94 μm by using a GSA based on a graphene film vacuum filtered on an output coupler (OC) mirror in a highly compact ceramic Thulium-doped Yttrium Aluminium Garnet (Tm:YAG) waveguide laser. Mode-locked pulses with 7.8 GHz repetition rate and Q-switched envelopes with 6.5 mW average output power are achieved.

II. CLADDING WAVEGUIDE AND GRAPHENE SATURABLE ABSORBER

The cladding waveguide is fabricated by ULI with an ultrafast Yb-doped fiber master-oscillator power amplifier laser (IMRA FCPA μ-Jewel D400), delivering 460 fs pulses at 1047 nm and 500 kHz repetition rate. ULI is done by focusing 220 nJ pulses through a 0.4 numerical aperture lens, below the polished surface of a Tm:YAG ceramic (1 at.% Tm-doped). A 36-μm-diameter waveguide is inscribed by translating the substrate at 3 mm/s. After inscription, a continuous wave (CW) waveguide laser is realized by using a 20% output coupler. The waveguide mode field diameter (MFD) is measured to be 32.7 and 36.9 μm in the horizontal and vertical directions, respectively, leading to a 9.6×10^{-6} cm^2 mode area [42]. The propagation loss (α_p) of the waveguide can be estimated from the waveguide laser slope efficiency, η [43]:

$$\eta = \frac{\ln\left(\frac{1}{R}\right)}{\ln\left(\frac{1}{R}\right) + 2\alpha_p l} \cdot \frac{\lambda_s}{\lambda_p}[1 - \exp(-\alpha_{abs} l)]\frac{dS}{dF} \qquad (1)$$

where $R = 80\%$ is the reflectance of the output coupler, $\lambda_{s,p}$ [m] are the signal and pump wavelengths, $\alpha_{\alpha bs}$ [m^{-1}] is the absorption coefficient for the pump beam, $l = 10.5$ mm is the waveguide length, and $\frac{dS}{dF} \approx 1$ is the mode-overlap factor (i.e., conversion efficiency of the pump light [44]). For $\eta = 11.5\%$ and $\alpha_{\alpha bs} = 2.146$ cm^{-1} [42], Eq. (1) gives $\alpha_p = 0.77$ dB/cm at the signal wavelength.

Our GSA is prepared following the process reported in Ref. [7]. For this, LPE graphene is dispersed in deionised water with sodium deoxycholate [5], [7], [8], [10]. The dispersion is then characterized by High Resolution Transmission Electron Microscopy (HRTEM), optical and Raman Spectroscopy. HRTEM reveals ~26% single-, ~22% bi- and ~18% tri-layers [10], [11], with ~1 μm average size. The dispersion is then vacuum filtered on a 5% OC mirror, resulting in a ~45 nm film, as determined by profilometry [7], with ~0.72 gcm^{-3} density [7], ~1/3 of that of graphite.

Raman spectra are acquired at 457, 514, and 633 nm [7]. Fig. 1(a) plots a typical spectrum of the LPE dispersion. We assign the D and D' peaks to the sub-micrometer edges of our flakes [45], [46], rather than to a large amount of disorder within the flakes. Fig. 1(b) plots the Raman spectrum of the graphene film at 514 nm.

Similar to the individual flakes discussed above, Disp(G) is 0.02 cm^{-1} nm^{-1} [47]. The 2D peak is still single Lorentzian, but ~24 cm^{-1} larger than in individual flakes [2]. Thus, even if the flakes are multi-layers, they are electronically decoupled and, to a first approximation, behave as a collection of single layers [48]. The ratio of the 2D and G integrated areas, A(2D)/A(G), is at most ~2, thus we have a doping ~1.3×10^{13} cm^{-2} [49] i.e., a Fermi level shift ~4–500 meV [49], [50].

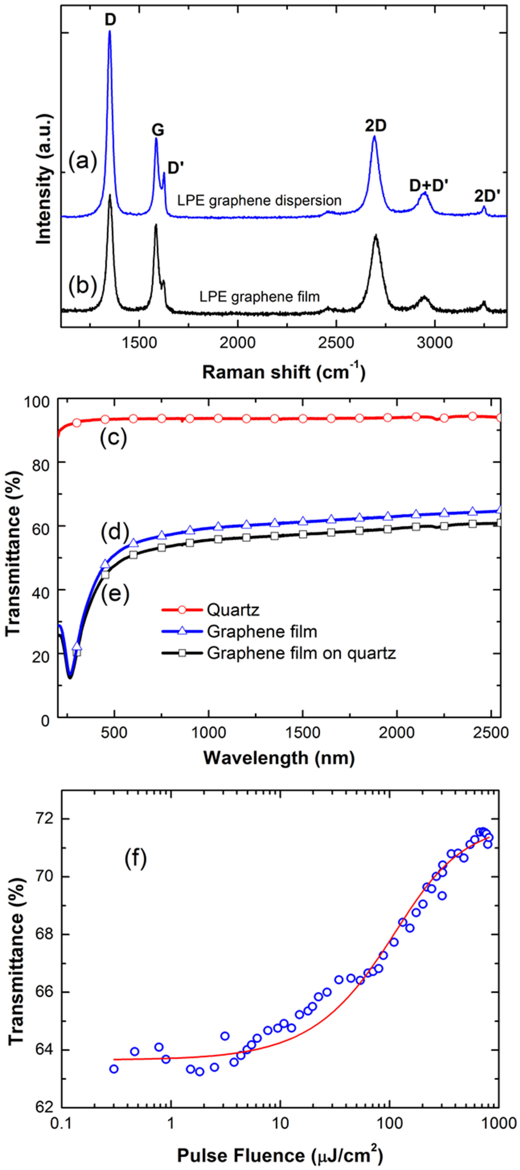

Fig. 1. Raman spectra at 514 nm of (a) graphene dispersion in deionized water and (b) graphene film. Transmittance of (c) quartz, (d) graphene-film and (e) graphene-film on quartz (f) Nonlinear transmission versus pulse fluence (blue dots) for the graphene-film on quartz, fitted to a model function (red line).

Fig. 1(c), (d), (e) plot the transmittance of quartz, the GSA and the GSA on quartz. The transmittance and reflectance at 1944 nm (our laser wavelength) are ~63% and ~11% respectively. The peak at ~266 nm is a signature of the van Hove singularity in the graphene density of states [51].

The number of graphene layers in the film is estimated to be ~40, using a recurrent matrix method, as discussed in Ref. [7]. A 40 layer graphene film with a density ~1/3 of graphite corresponds to a film thickness of 40 nm, in good agreement with the profilometry value. The nonlinear transmittance is measured

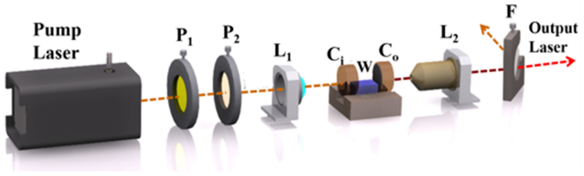

Fig. 2. Laser setup. P_1: Half-wave plate; P_2: Polarizer; L_1: Coupling convex lens; L_2: Coupling Lens; C_i: Pump mirror; C_o: GSAM; W: Tm:YAG cladding waveguide; F: Si filter.

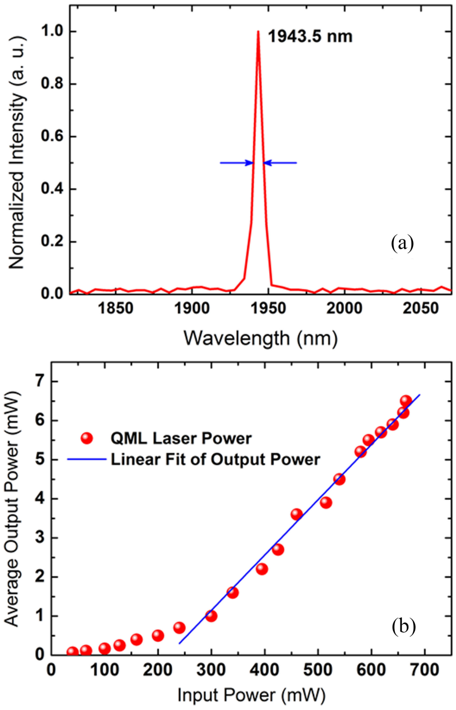

Fig. 3. (a) Optical spectrum of waveguide laser. (b) Output laser power versus incident pump power.

with an optical parametric amplifier generating ~100 fs pulses at a repetition rate of 1 kHz, centered at 2 μm. The sample is placed at the focus of the incident beam and the nonlinear transmittance is calculated as a ratio of the output power to the incident laser power. Fig. 1(f) plots the nonlinear transmittance as a function of incident pulse fluence. The sample has a saturation fluence ~59 μJ cm^{-2}, and a modulation depth ~8.4%.

III. Experimental Setup

A CW Ti: Sapphire laser at 800 nm is used as a pump source, as shown in Fig. 2. A half-wave plate (P_1) and a polarizer (P_2) adjust the input power and polarization. The pump beam is focused into the waveguide through a convex lens (L_1) with a focal length of 30 mm, resulting in a calculated diffraction limited spot size of 32 μm, which has a good match with the waveguide MFD, ensuring high coupling efficiency. The Fabry-Perot laser cavity is formed by adhering the pump mirror (C_i) and GSA mirror (GSAM) (C_o) to the facets of the sample with index matching gel ($n \approx 1.45$). The pump mirror has a high transmittance at ~800 nm and ~98% reflectivity at the laser wavelength. The cavity length is 10.5 mm. A Si filter (F) is utilized to separate the output and residual pump. The experimentally obtained pulse trains are recorded using a fast photodiode and a 50 GHz wide-bandwidth Agilent Infiniium DCA 86100A oscilloscope.

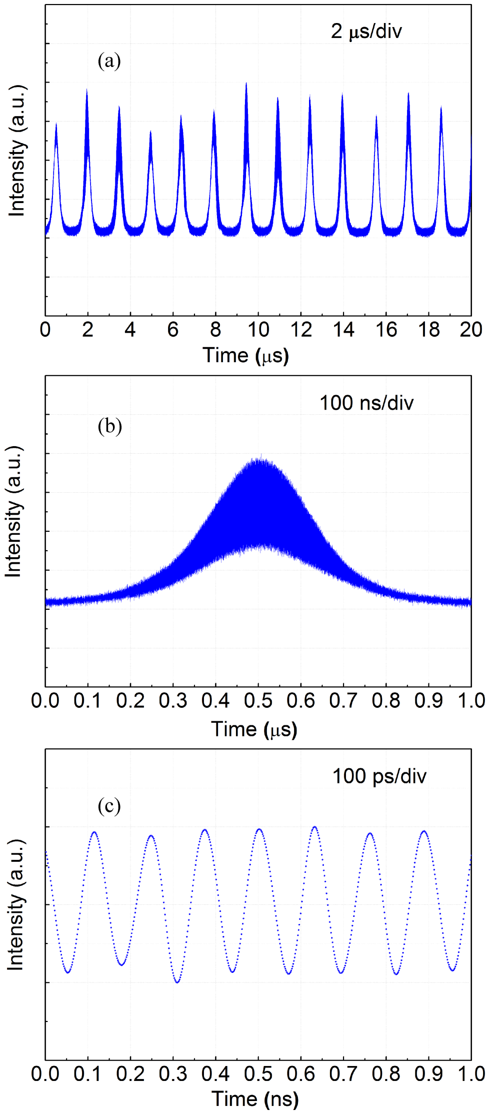

Fig. 4. (a), (b) Q-switched pulse envelopes. (c) Mode-locked pulse train.

IV. Results And Discussion

By adjusting the laser cavity elements and the GSA position, pulsed operation is realized. The spectrum, centered at 1943.5 nm, is shown in Fig. 3(a) with a full width at half maximum bandwidth of 6.7 nm. Fig. 3(b) plots the average output power as a function of the input power. At the highest available incident pump power of 665 mW, an average output power of 6.5 mW is achieved, giving an optical-to-optical conversion efficiency (i.e., rate of output to pump power [43]) of 1%. The waveguide laser slope efficiency (i.e., rate of output to pump power in excess of the lasing threshold [43]) is ∼2%, as given by the linear fit (blue solid line) of experimental results (red balls).

Fig. 4(a), (b) present the Q-switched envelopes on microsecond (2 μm/div) and nanosecond (100 ns/div) time scales, respectively. The repetition rate is ∼684 kHz. A 9.5 nJ pulse energy corresponds to each Q-switched envelope. Fig. 4(b) shows a single Q-switching envelope, containing the mode-locking pulses. The mode-locked pulse trains measured with a timescale of 100 ps/div are shown in Fig. 4(c), from which the mode-locking repetition rate is ∼7.8 GHz. The fundamental repetition frequency $f_{\rm rep}$ of mode-locking in a linear Fabry-Perot cavity determined by the free spectral range of the laser cavity is [52] $f_{\rm rep} = \frac{c}{2nl}$, where c [ms^{-1}] is the speed of light and n is the waveguide refractive index. A cavity length $l = 10.5$ mm yields a repetition frequency of 7.81 GHz, in good agreement with the observed mode-locking behavior.

The waveguide laser performance regime is also verified by applying the stability criterion which describes the stability limit between CW mode-locking and QML [22]. The critical intracavity pulse energy $E_{\rm P,c}$ is defined as [22]: $E_{\rm P,c} = (E_{\rm sat,L} E_{\rm sat,A} \Delta {\rm R})^{1/2}$, where $E_{\rm sat,L}$ is the saturation energy of the gain medium, $E_{\rm sat,A}$ represents the absorber saturation energy, and ΔR is the modulation depth of the SA. The values of the $E_{\rm sat,A}$ and ΔR of the GSA, derived from the GSA saturation measurements at 2 μm yield a $E_{\rm P,c}$ value two orders of magnitude lower than required for stable CW mode-locking, in agreement with the experiments. Stable CW mode-locking could be achieved with further optimization of the waveguide laser system, resulting in even lower waveguide propagation losses, highly doped gain media or reduced absorber modulation depth.

V. CONCLUSION

We reported a passively Q-switched mode-locked monolithic waveguide laser at 2 μm. A graphene film was integrated into the laser cavity employing a cladding waveguide fabricated in Tm:YAG by fs laser inscription. The laser features QML with 7.8 GHz mode-locked pulses, suitable for practical, compact mid-infrared pulsed laser sources.

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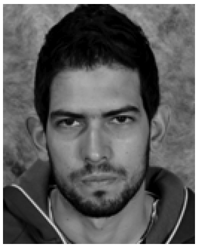

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