

# 7.8 GHz Graphene-based 2 $\mu\text{m}$ Monolithic Waveguide Laser

Yingying Ren, Graeme Brown, Rose Mary, Giorgos Demetriou, Daniel Popa, Felice Torrisi, Andrea C. Ferrari, Feng Chen and Ajoy K. Kar, *Member, IEEE*

**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  $\sim 7.8$  GHz pulse rate. This is a convenient, compact, high repetition rate laser for various applications, such as medical diagnostics and spectroscopy.

**Index Terms**— Solid lasers, Optical waveguides, Laser mode locking, Laser applications.

## 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 [1, 5-10, 12-21] SAs have been demonstrated for a wide spectral range. 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 observed underneath a Q-switched envelope [22]. In spite of the Q-switching tendency, the high pulse energy of the mode-locked pulses has potential applications in nonlinear frequency conversion [23] and surgery [24].

Tm<sup>3+</sup> doped solid-state lasers operating in the 2  $\mu\text{m}$  spectral range are of great interest for applications such as medicine [24], material processing [25], and environment monitoring

[26]. The operating wavelength is important because water, the main constituent of human body [27], absorbs more at 2  $\mu\text{m}$  ( $\sim 100 \text{ cm}^{-1}$ ) than at other conventional wavelengths, i.e.  $\sim 1.5 \mu\text{m}$  ( $\sim 10 \text{ cm}^{-1}$ ) and  $\sim 1 \mu\text{m}$  ( $\sim 1 \text{ cm}^{-1}$ ) [28]. Furthermore, gas molecules, such as CO<sub>2</sub>, show characteristic absorption lines [26], making 2  $\mu\text{m}$  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 prerequisite 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  $\mu\text{m}$  using CNT-SAs [15, 30] and at 1  $\mu\text{m}$  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  $\mu\text{m}$ , 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 integrated into various systems [2, 41]. LPE graphene can also be used as a film [7].

Y. Ren is with the College of Physics and Electronics, Shandong Normal University, Jinan 250014, China. (e-mail: [ryywy@sdnu.edu.cn](mailto:ryywy@sdnu.edu.cn))

G. Brown is with Optoscribe Ltd, 0/14 Alba Innovation Centre, Alba Campus, Livingston EH54 7GA, UK (e-mail: [g.brown@optoscribe.com](mailto:g.brown@optoscribe.com)).

R. Mary, G. Demetriou and A. K. Kar are with the Institute of Photonics and Quantum Sciences, School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh EH14 4AS, UK (e-mail: [rm330@hw.ac.uk](mailto:rm330@hw.ac.uk); [gd123@hw.ac.uk](mailto:gd123@hw.ac.uk); [A.K.Kar@hw.ac.uk](mailto:A.K.Kar@hw.ac.uk)).

F. Chen is with the School of Physics, State Key Laboratory of Particle Physics and Particle Irradiation (Ministry of Education), Shandong University, Jinan 250100, China (e-mail: [drfchen@sdu.edu.cn](mailto:drfchen@sdu.edu.cn)).

D. Popa, F. Torrisi and A. C. Ferrari are with Cambridge Graphene Centre, University of Cambridge, Cambridge CB3 0FA, UK (e-mail: [dp387@cam.ac.uk](mailto:dp387@cam.ac.uk); [ft242@cam.ac.uk](mailto:ft242@cam.ac.uk); [acf26@cam.ac.uk](mailto:acf26@cam.ac.uk)).

This reduces non-saturable losses, allowing high average-power.

Here we report QML at 1.94  $\mu\text{m}$  by using a GSA based on a graphene film vacuum filtrated 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

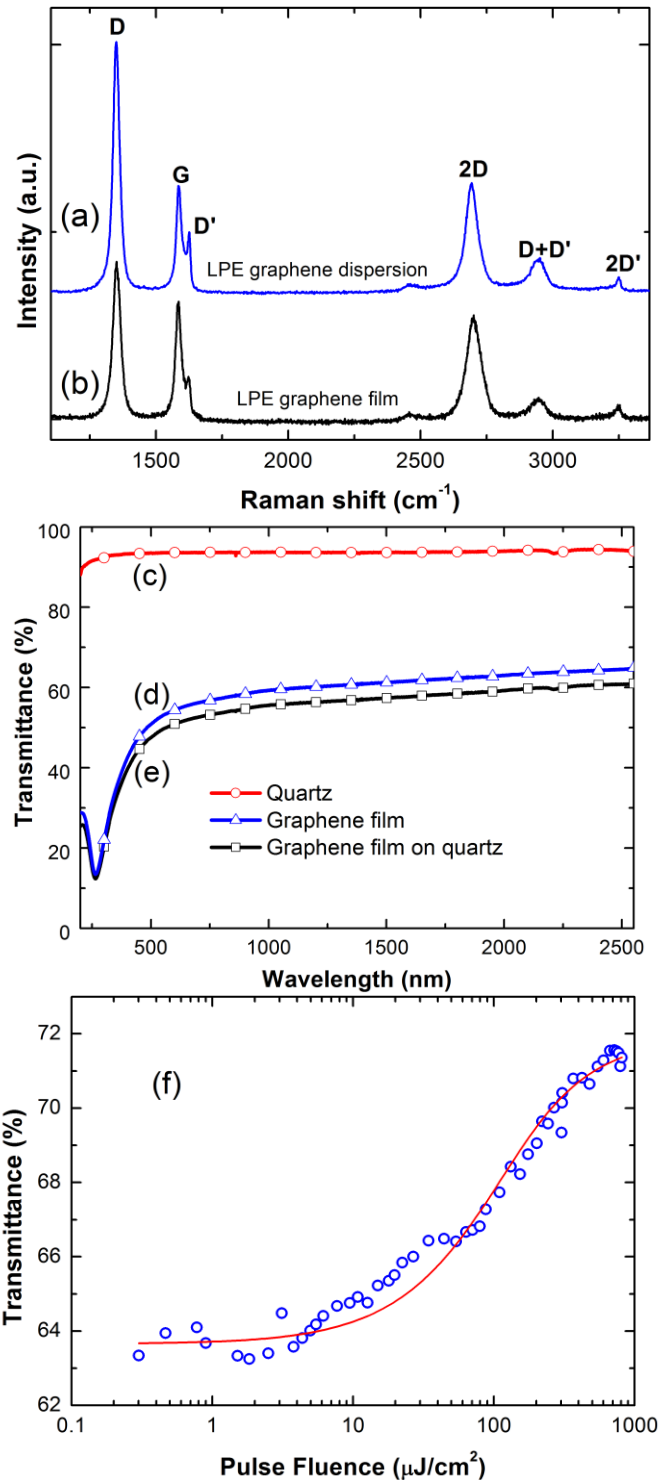
The cladding waveguide is fabricated by ULI with an ultrafast Yb-doped fiber master-oscillator power amplifier laser (IMRA FCPA  $\mu$ -Jewel D400), delivering 460 fs pulses at 1047 nm and 500 kHz repetition rate. ULI is done by focusing 220 nJ pulses through a 0.4 numerical aperture (NA) lens, below the polished surface of a Tm:YAG ceramic (1 at.% Tm-doped). A 36- $\mu\text{m}$ -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  $\mu\text{m}$  in horizontal and vertical direction respectively, leading to a  $9.6 \times 10^{-6} \text{ cm}^2$  mode area [42]. The propagation loss ( $\alpha_p$ ) of the waveguide can be estimated from the waveguide laser slope efficiency,  $\eta$  [43]:

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

where  $R = 80\%$  is the reflectance of the output coupler,  $\lambda_{s,p}$  [m] are the signal and pump wavelengths,  $\alpha_{abs}$  [ $\text{m}^{-1}$ ] is the absorption coefficient for the pump beam,  $l = 10.5 \text{ mm}$  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  $\alpha_{abs} = 2.146 \text{ cm}^{-1}$  [42], Eq. (1) gives  $\alpha_p = 0.77 \text{ dB/cm}$  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  $\sim 26\%$  single-,  $\sim 22\%$  bi- and  $\sim 18\%$  tri-layers [10, 11], with  $\sim 1 \mu\text{m}$  average size. The dispersion is then vacuum filtrated on a 5% OC mirror, resulting in a  $\sim 45 \text{ nm}$  film, as determined by profilometry [7], with  $\sim 0.72 \text{ gcm}^{-3}$  density [7],  $\sim 1/3$  of that of graphite.

Raman spectra are acquired at 457, 514, and 633 nm [7]. Figure 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 GF Raman spectrum at 514 nm.



**Figure 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 transmittance vs pulse fluence for the graphene-film on quartz.

Similar to the individual flakes discussed above,  $\text{Disp}(G)$  is  $0.02 \text{ cm}^{-1} \text{ nm}^{-1}$  [47]. The 2D peak is still single Lorentzian, but  $\sim 24 \text{ cm}^{-1}$  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  $\sim 2$ , thus we have a doping  $\sim 1.3 \times 10^{13} \text{ cm}^{-2}$  [49] i.e. a

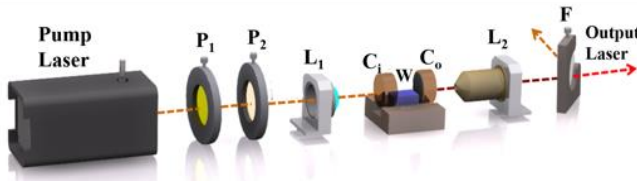
Fermi level shift  $\sim 4\text{-}500$  meV [49, 50].

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

The number of graphene layers in film is estimated using a recurrent matrix method [7], and estimated to be  $\sim 40$  layers. The calculation procedure is detailed in Ref. [7]. A 40 layer thick graphene film with a density  $\sim 1/3$  of graphite corresponds to a film thickness of 40 nm, which is in good agreement with the profilometry value. The nonlinear transmittance is measured with an optical parametric amplifier generating  $\sim 100$  fs pulses at a repetition rate of 1 kHz, centered at 2  $\mu\text{m}$ . The sample is placed at the path of the incident beam and the nonlinear transmittance is calculated as a ratio of the output power to the incident laser power. Figure 1(f) plots the nonlinear transmittance as a function of incident pulse fluence. The sample has a saturation fluence  $\sim 59 \mu\text{J cm}^{-2}$ , and a modulation depth  $\sim 8.4\%$ .

### III. EXPERIMENTAL SETUP

A CW Ti: Sapphire laser at 800 nm is used as a pump source, as shown in Figure 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  $\mu\text{m}$ , 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  $\sim 800$  nm and  $\sim 98\%$  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.



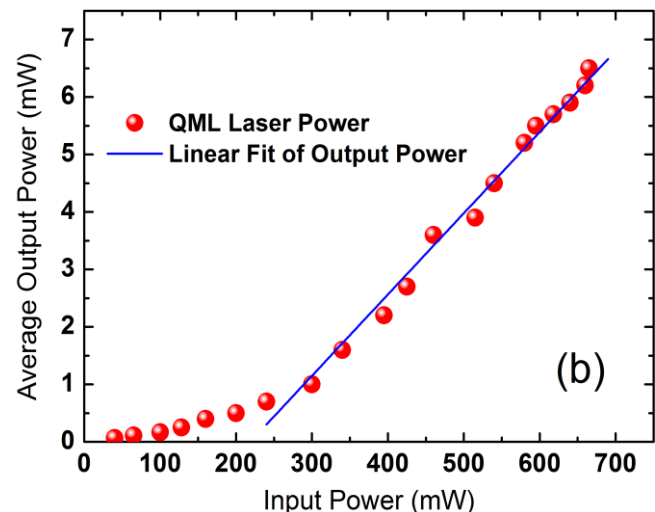
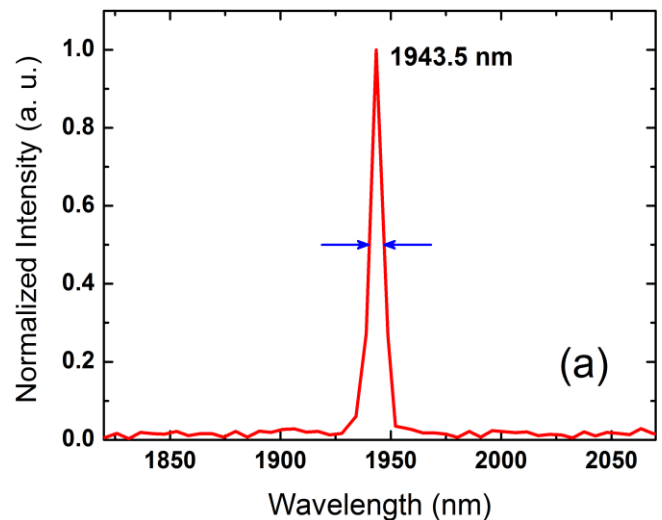
**Figure 2.** Laser setup.  $P_1$ : half-wave plate;  $P_2$ : Polarizer;  $L_1$ : Coupling convex lens;  $L_2$ : Coupling Lens;  $C_i$ : Pump mirror;  $C_o$ : GSAM; W: Tm:YAG cladding waveguide; F: Si filter

### IV. RESULTS AND DISCUSSION

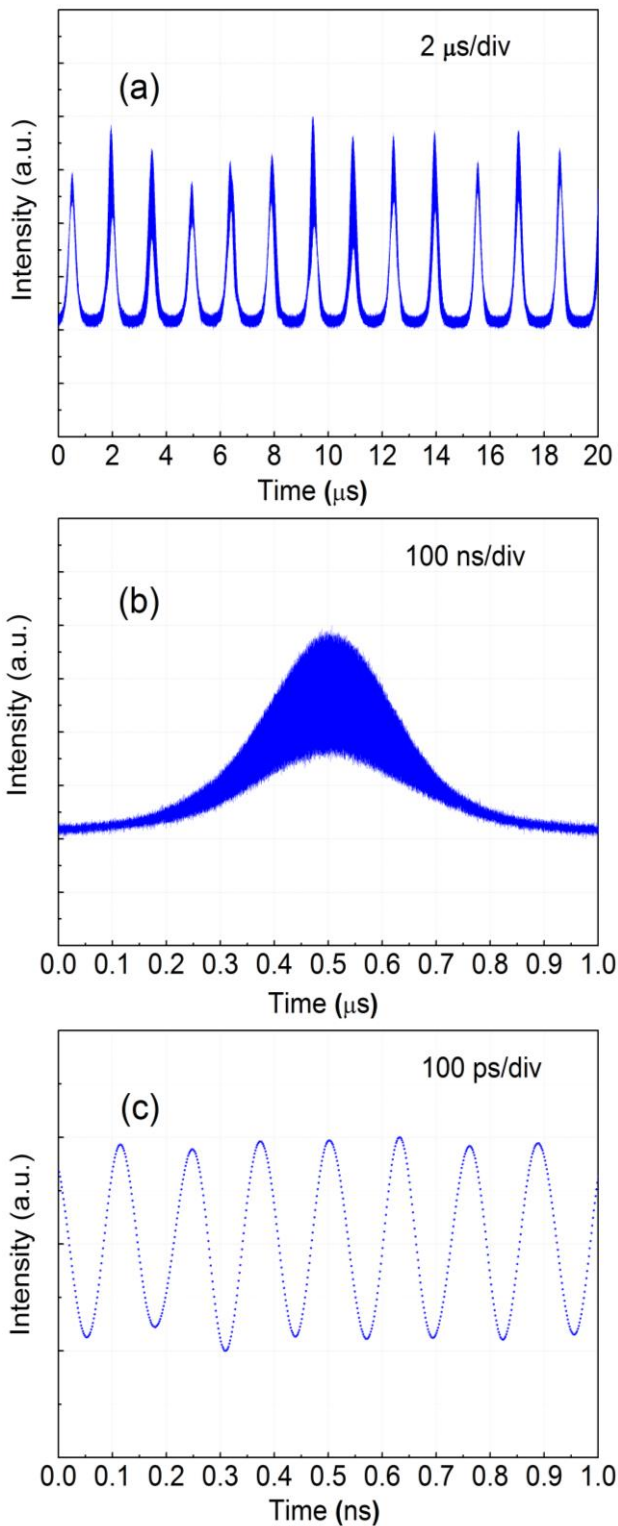
By adjusting the laser cavity elements and the GSA position, pulsed operation is realized. The spectrum, centered at 1943.5 nm, is shown in Figure 3(a) with a full width at half maximum (FWHM) bandwidth of 6.7 nm. Figure 3(b) plots the average output power as a function of the input power. At the highest 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  $\sim 2\%$ , as given by the linear fit (blue solid line) of experimental results (red balls).

Figures 4(a,b) present the Q-switched envelopes on microsecond (2  $\mu\text{m}/\text{div}$ ) and nanosecond (100 ns/div) time scales, respectively. The repetition rate is  $\sim 684$  kHz. A 9.5 nJ pulse energy corresponds to each Q-switched envelope. Figure 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  $\sim 7.8$  GHz. The fundamental repetition frequency  $f_{rep}$  of mode-locking in a linear Fabry-Perot cavity determined by the free spectral range of the laser cavity is [52]  $f_{rep} = \frac{c}{2nl}$ , where  $c$  [ $\text{ms}^{-1}$ ] 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, showing good agreement with the observed mode-locking behavior.



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



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

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  $E_{P,c}$  is defined as [22]:  $E_{P,c} = (E_{sat,L} E_{sat,A} \Delta R)^{1/2}$ , where  $E_{sat,L}$  is the saturation energy of the gain medium,  $E_{sat,A}$  represents the absorber saturation energy,

and  $\Delta R$  is the modulation depth of the SA. The values of the  $E_{sat,A}$  and  $\Delta R$  of the GSA, derived from the GSA saturation measurements at 2  $\mu\text{m}$  yield a  $E_{P,c}$  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. CONCLUSIONS

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

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**Yingying Ren** was born in Fenyang, China, in 1985. She was awarded her BSc degree from Shandong University, China in 2004. She received her PhD, also from Shandong University, in 2013. From 2011-12, she worked at the Nonlinear Optics Lab at Heriot-Watt University, Edinburgh, UK as a joint student.

Currently, she is a lecturer at Shandong Normal University, China. Her research activity is focused on the interaction of energetic ion beams or ultrafast lasers with materials for waveguide device applications.

**Graeme Brown** was awarded his MPhys (Hons) degree in Physics with Laser Science from Heriot-Watt University, UK in 1997. He received his PhD in Physics in 2001, also from Heriot-Watt University.

From 2000-08 he worked in the optoelectronics industry in the development of new telecommunication components and advanced optical test systems. In 2009 he returned to Heriot-Watt and led the technical development on a Scottish Enterprise Proof-of-Concept on the commercialization of Ultrafast Laser Inscription (ULI) for sensing applications. His work in this field also includes the development of integrated, compact laser sources fabricated by ULI, in particular the development of integrated mode-locking elements based on saturable absorbers such as graphene and carbon nanotubes.

He is Co-Founder of a spin-out company, Optoscribe, which is commercializing components fabricated by ULI.



**Rose Mary** received her MSc in Photonics from Cochin University of Science and Technology, India, in 2010. Currently, she is working towards the PhD degree in the Nonlinear Optics Group at Heriot-Watt University, Edinburgh, UK. Her research work and interests include the design and development of compact waveguide lasers by the technique of ultrafast laser inscription.



**Giorgos Demetriou** was born in Larnaka, Cyprus in 1986. He received the Diploma in Electrical Engineering and Computer Science from the National Technical University of Athens in 2012. He is currently pursuing the PhD degree in Physics at Heriot Watt University, Edinburgh. His research interests include ultrafast laser inscription and investigation of nonlinearities in Gallium Lanthanum Sulfides for Mid-IR applications.



**Daniel Popa** received his MSc, in Engineering, from University of Rome "Tor Vergata", Italy in 2008. He received his PhD, in Engineering, from University of Cambridge, U.K., in 2013. He is a Research Fellow in Emmanuel College and Cambridge Graphene Centre, U.K., working on the development of graphene and related materials based photonics.



**Felice Torrisi** is a Research Associate in the Cambridge Graphene Centre of the University of Cambridge and Schlumberger Research Fellow at Darwin College. He graduated at the University of Catania, Italy after a research period at Institute of Microelectronics and Microsystems of the Italian National Research Council as Research Assistant. He joined the Department of Engineering at the University of Cambridge in October 2008 where he obtained his PhD in 2013. His research interests cover the development of nanomaterials based dispersions,

inks and coatings and their incorporation into polymer composites for printed flexible/stretchable and transparent electronics and optoelectronics. He is regular reviewer of numerous international journals in nanomaterials, nanoscience and flexible electronics. He holds two patents.



**Andrea C. Ferrari** earned a PhD in electrical engineering from Cambridge University, after a Laurea in nuclear engineering from Politecnico di Milano, Italy. He is Professor of Nanotechnology and head of the Nanomaterials and Spectroscopy group at the Department of Engineering and Nanoscience Centre of Cambridge University. He is the founding Director of the Cambridge Graphene Centre and of the EPSRC Centre for Doctoral Training in Graphene Technology. He is the Chair of the Executive Board of the European Graphene Flagship. He is Fellow of

Pembroke College, Cambridge, Fellow of the American Physical Society, and Royal Society Wolfson Research Merit Award Holder. His research interests include nanomaterials growth, modelling, characterization, and devices. In particular, he focuses on graphene, nanotubes, diamond-like carbon, and nanowires for applications in electronics and photonics.



**Feng Chen** was born in Binzhou, China in 1975. He is currently a Professor at the School of Physics and the Director of Ion Beam Laboratory, Shandong University, China. He received a Bachelors of Science degree in 1997 from Shandong Normal University, followed by PhD in 2002 from Shandong University.

After finishing his PhD, he joined Shandong University as a Lecturer. He was at the Clausthal University of Technology, Germany from 2003-05 as an Alexander von Humboldt Research Fellow.

He became an Associate Professor and Professor at Shandong University in 2004 and 2006, respectively.

His research interests include ion-beam modifications of materials, optical waveguides produced by energetic ion beams or ultrafast lasers, photonic crystals, lasers, solitons etc. He is a Fellow of the Institute of Physics, UK, a Senior Member of the Optical Society of America and Chinese Optical Society, and a Member of the Director Board of the Chinese Society of Nuclear Physics.



**Ajoy K. Kar** received his MSc degree from the Indian Institute of Technology, Delhi, India. He received his PhD degree from the University of Essex, during which he developed his interest in lasers and nonlinear optics.

He joined Heriot-Watt University, Edinburgh, UK in 1979, where he is currently a Professor at the Institute of Photonics and Quantum Sciences. He has over 30 years of experience in the investigation of nonlinear optical properties of materials and their applications. He has also pioneered the Photonics Education in the UK and Europe. Some of his

current projects include ultrafast laser inscription (ULI) of photonic devices for a broad spectral range from visible to Mid-IR and the development of microfluidic devices for biophotonics applications.

Prof Kar is a founding member of Optoscribe, a spin-out company specialized in ULI based photonic device fabrication. He is a member of Institute of Electrical and Electronics Engineers, and the Optical Society of America.