Graphene Mode-Locked Fiber Laser at 2.8 μm

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***Abstract*— Mid-infrared erbium (Er^{3+})-doped ZrF_4–BaF_2–LaF_3–AlF_3–NaF fiber laser mode-locked by multilayer graphene saturable absorber was demonstrated. Mode-locked pulses at 2.8 μm with an average output power of 18 mW at a repetition rate of 25.4 MHz, corresponding to a pulse energy of 0.7 nJ, were obtained. The pulsewidth was measured to be ∼42 ps by a home-made autocorrelator. Our experiment has validated the mode-locking capability of graphene in the 3-μm wavelength region.**

***Index Terms*— Mode-locked laser, erbium-doped ZBLAN fiber laser, mid-infrared laser.**

I. INTRODUCTION

LASERS in the mid-infrared (mid-IR) region have been attracting great attention because of their extensive applications including medical surgery, spectroscopy, frequency metrology, remote sensing, materials processing, and pump sources for nonlinear wavelength convertors [1]–[4]. Among various mid-IR laser platforms, fiber lasers offer advantages such as inherent simplicity and flexibility, high efficiency, outstanding heat-dissipating capability, and excellent beam quality, and have been extensively studied during the last few decades [5], [6]. In particular, mid-IR fiber lasers at 3 μm have been successfully demonstrated with erbium (Er^{3+}), holmium (Ho^{3+}), and dysprosium (Dy^{3+}) ions doped into ZrF_4–BaF_2–LaF_3–AlF_3–NaF (ZBLAN) fibers [5], [6] and recent advances in output power scaling have resulted in continuous-wave (CW) operation at 20-W or even 30-W level [7]–[10]. Nevertheless, pulsed lasers with peak powers some orders of magnitude higher than in the CW regime are highly demanded for laser surgery, remote sensing, and nonlinear wavelength convertors [2], [3]. Laser pulses are generally produced by the processes of Q-switching or mode-locking. Compared to Q-switched lasers, mode-locked lasers offer much higher peak powers and much shorter pulse durations. Therefore, mode-locked operation of a mid-infrared fiber laser has become an attractive research topic in recent years.

Mode-locked operation of an Er^{3+}-doped ZBLAN fiber laser in the 3 μm wavelength region was first demonstrated by Frerichs and Unrau using the flying mirror technique and an InAs saturable absorber [11]. However, the mode-locked operation was Q-switched and non-continuous. A continuous-wave mode-locked Er^{3+}-ZBLAN fiber laser was recently demonstrated by C. Wei et al using a Fe^{2+}:ZnSe saturable absorber [12]. Stable and continuous mode-locked pulses at 2.78 μm with a pulse duration of 19 ps and an average power of 51 mW were obtained [12]. Meanwhile, J. Li et al reported a partially mode-locked $Ho^{3+}Pr^{3+}$-codoped ZBLAN fiber laser at 2.87 μm by using a GaAs-based semiconductor saturable absorber mirror (SESAM) [13]. Very recently, they demonstrated a mode-locked $Ho^{3+}Pr^{3+}$-codoped ZBLAN fiber laser with improved stability by employing a ring cavity fiber laser configuration with a transmissive semiconductor saturable absorber [14]. A SESAM-mode-locked Er^{3+}-doped ZBLAN fiber laser at 2.8 μm was also recently demonstrated by A. Haboucha et al. [15]. Though SESAMs are currently the most prevalent saturable absorbers and have shown their capability for mode-locking mid-IR fiber lasers in the 3 μm region, they usually have a narrow operating wavelength range and require complex fabrication and packaging. Kerr nonlinearity is also widely used for passive mode-locking and most recently, S. Duval et al demonstrated the first femtosecond fluoride fiber laser operating near 3 μm based on nonlinear polarization evolution (NPE) [16]. However, mode-locked lasers based on NPE usually require careful alignment and frequent adjustment. In recent years, graphene-based devices have emerged as innovative and remarkable saturable absorbers for mode-locking lasers because of their ultra-broad absorption band, low saturation intensity, and ultrafast recovery time [17], [18]. A graphene mode-locked laser was first demonstrated in 2009 in the 1.5 μm wavelength region due to the readily available fiber components in this telecommunication window [19], [20]. The ultra-broad operating wavelength range of graphene-based saturable absorbers has been validated by their use in mode-locked solid-state lasers and fiber lasers in the 1 μm, 2 μm and even 2.5 μm wavelength region [21]–[23]. Most recently we have demonstrated a Q-switched Er^{3+}-doped ZBLAN fiber laser at 2.8 μm and

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Manuscript received June 25, 2015; revised August 25, 2015; accepted September 11, 2015. Date of publication September 15, 2015; date of current version November 23, 2015. This work was supported in part by the Engineering Research Center for Integrated Access Networks through the National Science Foundation under Grant EEC-0812072 and in part by the Technology Research Initiative Fund Photonics Initiative through the University of Arizona. The work of F. Wang and X. Guo was supported by the National Natural Science Foundation of China under Grant 21173041 and Grant 61378025.

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Digital Object Identifier 10.1109/LPT.2015.2478836

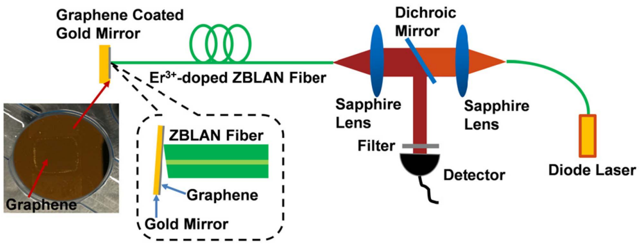

Fig. 1. Schematic of the experimental setup of a multi-layer graphene mode-locked Er^{3+}-doped ZBLAN fiber laser at 2.78 μm.

Ho^{3+}-doped ZBLAN fiber laser at 2.93 μm using graphene deposited fiber mirrors as the saturable absorbers [24], [25]. These developments suggested the pursuit of mode-locked fiber lasers in the 3 μm region by using a graphene device with proper absorption and modulation depth as the mode locker. In this letter we report the mode-locked operation of an Er^{3+}-doped ZBLAN fiber laser at 2.78 μm induced by a multiple layer graphene coated gold mirror as the saturable absorber.

II. EXPERIMENTAL SETUP

The schematic of the experimental setup is shown in Fig. 1. A fiber-coupled diode laser at 975 nm was used as the pump source. Two identical plano-convex sapphire lenses (focal length 25.4 mm) were used to collimate and focus the pump laser to the inner cladding of the 4-m Er^{3+}-doped double-clad ZBLAN fiber (FiberLabs Inc.). The Er^{3+}-doped ZBLAN fiber has a core with dopant concentration of 8 mol.%, NA of 0.1 and diameter of 15 μm, an inner cladding with a diameter of 125 μm, and NA of 0.4. The front end of the Er^{3+}-doped ZBLAN fiber was flat cleaved to work as the output coupler of the laser cavity and provide ~4% feedback, while the rear end is angle cleaved (~8°) to eliminate the influence of Fresnel reflection. A multilayer graphene coated gold mirror was placed close to the angle-cleaved Er^{3+}-doped ZBLAN fiber end as shown in the inset of Fig. 1 and used to serve as the saturable absorber and cavity mirror. It is noted that the graphene coated gold mirror was adjusted at an angle (~ 4°) to the vertical direction and thus the laser beam can be coupled back into the gain fiber core with a very low loss ($<$ 0.1 dB) because the air gap between the gold mirror and the fiber core (~ 15 μm) is much smaller than the Rayleigh range (~ 144 μm). Between the two collimating and focusing sapphire lenses a dichroic mirror with high transmissivity at 975 nm (T = 91%) and high reflectivity at 2.78 μm (R = 98%) was placed at a 45° angle of incidence to outcouple the signal laser. A long-pass filter was used to block the light below 1.7 μm. An InSb detector with a rise time of 7 ns was used to measure the time domain performance of the mode-locked laser. The pulse train was recorded by an oscilloscope (Tektronix TDS 1012) and the average output power was measured by a thermal detector (Thorlabs, S310C).

A saturable absorber with optimized absorption and modulation is very critical for mode-locking a fiber laser. We used a thermal chemical vapor deposition (CVD) method [26] to fabricate the graphene thin films, because with this method it is easier to achieve the desired thickness of graphene thin films than with the optically driven deposition method [24]. In addition, the CVD method can make large-area uniform graphene thin films that can be diced into smaller pieces and used for a variety of laser development and optical characterization efforts. The multilayer graphene thin films were grown on copper foil (100 μm thick, 99.9% purity) by use of the CVD system (base pressure ~5Pa) at a growth temperature of ~1050°C in a mixture of 300 sccm argon gas, 20 sccm hydrogen and 0.8 sccm methane for ~8 minutes. A standard PMMA assisted transfer process was used to deposit the graphene thin films onto gold mirrors [27]. We first used $FeCl_3$ to etch away the copper substrate and then rinsed the graphene (with PMMA) multiple times in de-ionized (DI) water to completely get rid of the residual copper etchant. 2 ml of 2% HCl solution in 15 ml of DI water was used to wash away residual iron particles. The graphene sample was then picked up by the gold mirror from DI water. Acetone was used to remove the PMMA to result in a graphene coated gold mirror. The Raman spectrum of the multilayer graphene was measured using a Horiba Jobin Yvon Raman system with a pump laser at 514 nm and is shown in the inset of Fig. 2, from which we estimate the saturable absorber to be about 4-6 layers thick. The linear and nonlinear absorption characteristics of the graphene saturable absorber sample were measured by using the mode-locked Er^{3+}-doped ZBLAN fiber laser reported here. The power-dependent transmittance of the graphene sample is shown in Fig. 2. The linear absorption of the multilayer graphene was measured to be about 12% and its modulation depth and saturation power intensity were estimated to be ~10% and 2 MW/cm^2, respectively.

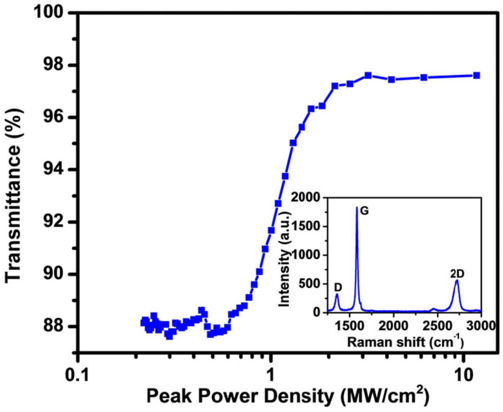

Fig. 2. Power-dependent transmittance of the graphene saturable absorber sample. Inset shows the Raman spectrum of the graphene sample used in the experiment.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

The threshold power of the Er^{3+}-doped ZBLAN fiber laser was measured to be 350 mW. However, the mode-locked operation was not stable until the pump power was increased to 420 mW. Stable mode-locked operation of the Er^{3+}-doped ZBLAN fiber laser was maintained at a pump power of 420-470 mW. Pulse trains of the graphene mode-locked fiber laser at a pump power of 470 mW over 500 ns and 5 μs

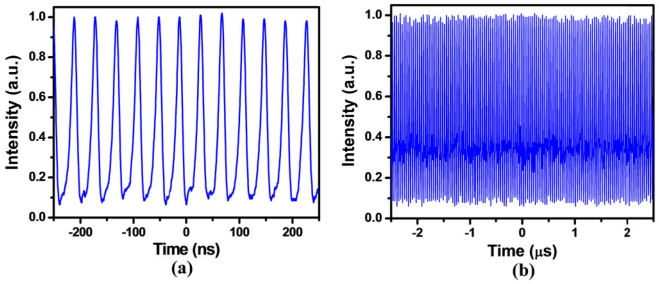

Fig. 3. The pulse train of the graphene mode-locked Er^{3+}-doped ZBLAN fiber laser over (a) a 500 ns and (b) a 5 μs duration at a pump power of 470 mW.

durations were recorded with a digital oscilloscope and are shown in Figs. 3(a) and 3(b), respectively. The variation of the pulse peaks is about 6%, indicating the fairly stable operation of this mode-locked fiber laser, which is degraded mainly by the Fresnel reflection of the uncoated plano-convex sapphire lens and self-pulsing in the highly Er^{3+}-doped ZBLAN fiber [28], [29]. The stability of this graphene mode-locked fiber laser can be significantly improved by employing an FBG with high reflection as the output coupler [15], utilizing a ring cavity to eliminate spatial hole burning [14], or using anti-reflective (AR) coated lenses to reduce the influence of Fresnel reflections. The average output power of this mode-locked fiber laser at a pump power of 470 mW was measured to be about 18 mW. The repetition rate is about 25 MHz, which corresponds to a cavity length of 4 meters. As the pump power exceeded 470 mW, the mode-locked operation of the Er^{3+}-doped ZBLAN fiber laser became unstable and degraded with the increased pump power. However, the unstable mode-locked operation was maintained even at a pump power of 2 W with more than 100 mW average output power. The unstable mode-locked operation at high pump power may be primarily attributed to self-pulsing in the Er^{3+}-doped ZBLAN fiber, which is usually caused by saturable absorption of the ion clusters in heavily ion doped fibers [28], [29]. When the pump power was more than 2 W, the multilayer graphene was usually damaged and the output of the fiber laser decreased.

The spectrum of the multilayer graphene mode-locked Er^{3+}-doped ZBLAN fiber laser at a pump power of 470 mW was measured with a Yokogawa AQ6375 optical spectrum analyzer at a resolution of 0.05 nm and is shown in Fig. 4. The central wavelength is about 2784.5 nm with a full width at half-maximum (FWHM) of 0.21 nm, corresponding to a pulse width of 39 ps for sech^2-shaped and transform limited pulses. The radio frequency (RF) spectrum of the multilayer graphene mode-locked Er^{3+}-doped ZBLAN fiber laser at a pump power of 470 mW was measured with a spectrum analyzer (Advantest R3267) and is shown in Fig. 5. The signal-to-noise ratio (SNR) of the RF spectrum was measured to be about 43.5 dB at a resolution bandwidth of 1 kHz. The RF spectrum over 0-100 MHz was measured and is shown in the inset of Fig. 5.

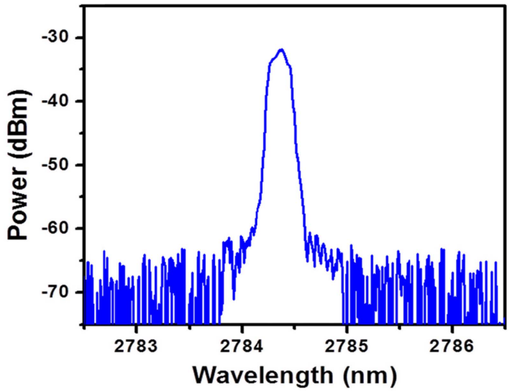

Fig. 4. The optical spectrum of the multilayer graphene mode-locked Er^{3+}-doped ZBLAN fiber laser at a pump power of 470 mW.

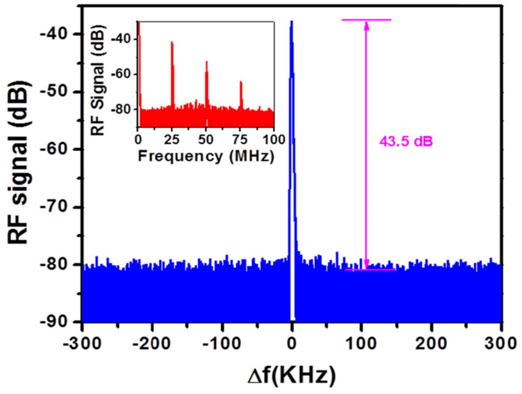

Fig. 5. The RF spectrum of the mode-locked pulses centered at 25.4 MHz with a SNR of 43.5 dB. The pump power is 470 mW. (Inset, the RF spectrum over a 100 MHz range).

The pulse duration of the multilayer graphene mode-locked Er^{3+}-doped ZBLAN fiber laser at 2.78 μm was measured with a home-made autocorrelator. The schematic of the autocorrelator is shown in Fig. 6(a). A 45:55 pellicle beam splitter is used to split the incident beam into two beams and direct them onto retroflectors 1 and 2, respectively. The two beams reflected from the two retroreflectors intersect at the beam splitter and combined into an interfered beam. The combined beam was focused by a CaF_2 lens onto an InGaAs detector, which can generate and detect the autocorrelation signal through the two-photon absorption process of InGaAs with linear absorption at 700-1800 nm. Retroreflector 1 was fixed while Retroreflector 2 was mounted on a motorized translation stage to change the temporal overlap of the pulses reflected from the two retroreflectors. The incident beam was modulated by a chopper and a lock-in amplifier (SR830 DSP) was used to detect the two-photon absorption signal. A long-pass filter was placed in front of the InGaAs detector to eliminate background noise. Since the 18 mW average output power of the mode-locked fiber laser oscillator was too low to be detected with this home-made autocorrelator, we amplified the signal average power to about 300 mW using a 3-m Er^{3+}-doped ZBLAN fiber amplifier [30]. Since the pulse width of this mode-locked laser is estimated to be 39 ps and the peak power is about 17 W, the accumulated nonlinear phase shift (B-integral) of the 3-m Er^{3+}-doped ZBLAN fiber amplifier is less than 0.2 and we can assume that the pulse width

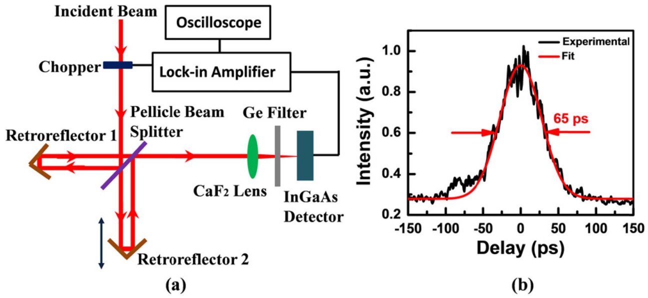

Fig. 6. (a) Schematic of the interferometric autocorrelator based on two-photon absorption in an InGaAs detector; (b) the autocorrelation trace recorded by the oscilloscope. The black line represents the experimental results and the red line represents the fitting results.

changes very little after the amplification. The autocorrelation trace of the amplified pulses was recorded by an oscilloscope that was connected to the lock-in amplifier and is shown in Fig. 6(b). Since we used a lock-in amplifier to detect the two-photon absorption signal, the interferometric signal inside the autocorrelation trace was averaged out. The full width at half maximum (FWHM) of the autocorrelation trace was calculated to be 65 ps by fitting the autocorrelation trace with a sech^2 function, yielding an FWHM pulse width of 42 ps. The time-bandwidth product of this mode-locked laser is 0.342. Therefore, the pulses of this multilayer graphene mode-locked Er^{3+}-doped ZBLAN fiber laser are nearly transform-limited.

IV. CONCLUSION

In conclusion, mode-locked operation of an Er^{3+}-doped ZBLAN fiber laser induced by multilayer graphene was demonstrated for the first time, to the best of our knowledge. Mode-locked pulses at 2.78 μm with an average output power of 18 mW and a pulse width of 42 ps at a repetition rate of 25.4 MHz were obtained. Our experiment has validated that graphene is a remarkable saturable absorber for mode-locking lasers in the 3 μm region.

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