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Graphene mode-locked femtosecond Yb:KLuW laser

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Large-area monolayer graphene, synthesized by chemical vapor deposition, was transferred to a 1-in. quartz substrate. The high-quality monolayer graphene has been subject to characterization of the nonlinear properties near 1 μm and was successfully applied as saturable absorber for passive mode-locking of a femtosecond Yb:KLuW laser. The diode-pumped mode-locked Yb:KLuW laser was tunable around 1.04 μm and delivered pulses as short as 160 fs. The maximum output power of 160 mW was demonstrated for 203 fs pulse duration. The mode-locked laser results are comparable to those demonstrated with the same laser gain medium using single-walled carbon nanotubes as saturable absorbers. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4760265]

Passive mode locking realized by incorporating an appropriate saturable absorber into the laser cavity is a well-established method for femtosecond pulse generation. Semiconductor based saturable absorbers, like SESAMs, are commonly used in femtosecond laser technology, providing a robust intracavity mode-locking mechanism that allowed for several novel real-world applications for ultrafast lasers.[1] However, SESAMs only provide useful optical nonlinearity in a small bandwidth and often require sophisticated manufacturing technology.[2] Consequently, low-dimensional carbon nanostructures such as carbon nanotubes (CNTs) and graphene have been introduced as one alternative because of their unique physical and optical properties.[3–5] Both exhibit a very fast recovery time and a high damage threshold and can be fabricated by relatively simple processes. Compared to CNTs where the possible operating wavelength depends on the nanotube diameter and chirality defining the bandgap, graphene has the great advantage of ultrafast saturable absorption over an ultrabroad spectral region without bandgap engineering due to its point-bandgap structure and linear energy dispersion of Dirac electrons.

By utilizing the nonlinear absorption of semiconducting single-walled CNTs, passive mode-locking of different fiber and bulk solid-state lasers has been successfully demonstrated in the wavelength range between 0.8 and 2.1 μm.[6–10] Graphene enables the extension of ultrafast photonic applications in the mid-IR spectral range beyond 2.1 μm, where ultrafast components are not easily attainable.

Graphene is a monolayer of carbon atoms arranged in a two-dimensional hexagonal lattice and the ultrafast nonlinear properties of graphene have been intensively studied.[11] The relaxation properties of graphene are characterized by a two-fold carrier dynamics composed of a fast relaxation on the time scale of about 100 fs associated with intraband carrier-carrier scattering and a slower response of about 1–2 ps attributed to optical phonon cooling and interband carrier recombination.[11–13] Additionally, graphene stands out due to its high thermal conductivity,[14] which is beneficial for high-power laser operation as thermally induced damage can be minimized.

The initial investigations on laser mode-locking employing graphene-based saturable absorbers (graphene SAs) were focused on Er-doped fiber lasers,[3] with the demonstration in 2009, yielding about 800 fs at 1.5 μm.[4] The graphene applied in these first attempts as SAs were characterized by an undefined number of layers. Graphene/polymer composites, and graphene flakes produced by exfoliation or chemical vapor deposition (CVD) methods were mostly applied as mode-lockers after depositing them onto fiber ends.[4,15–17] As a result, the linear and nonsaturable losses were quite high and the output power was limited to the milliwatt range, and therefore such SAs are not applicable for femtosecond bulk laser mode-locking. Furthermore, stable mode-locking of bulk lasers requires graphene films large enough to cover the whole laser beam size in the cavity. One possible way to realize the latter requirement is mixing and overlapping small graphene sheets. However, the uniform deposition of such graphene flakes on a substrate is a quite difficult issue. In the last years, many efforts have been devoted to deposit few layers of high-quality graphene in order to fabricate low-loss SAs capable to mode-lock femtosecond bulk solid-state lasers. Successful graphene SA mode-locking of bulk lasers was achieved at 1 μm using Nd-doped crystals, Nd:YAG[18] and Nd:$GdVO_4$[19] delivering quite long pulses of 4 and 16 ps duration, respectively. Recently, applying Yb^{3+}:$KGd(WO_4)_2$

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(Yb:KGdW) sub-ps pulses with a duration of 428 fs was demonstrated.[20] The listed bulk lasers operating at 1 μm are based on graphene oxide (GO)[19,20] or on reduced GO,[18] whereas the intracavity loss through the absorber has to be further improved. More about the GO fabrication technology can be found in Ref. 3. The manufacturing of a SA based on high-quality large-area graphene grown by the CVD method[15] allowed for femtosecond bulk laser mode-locking at other wavelengths. Using Er,Yb:glass as active medium, pulse durations of 260 fs were reported at 1.5 μm.[21] Recently, a Cr:forsterite laser was mode-locked using a high-quality monolayer graphene SA fabricated by CVD and transfer technique on a quartz substrate, delivering sub-100 fs pulses at 1.25 μm.[22] Applying a graphene SA manufactured by the same fabrication technology, we report, in the present work, sub-200 fs mode-locked laser operation around 1 μm using Yb^{3+}:$KLu(WO_4)_2$ (Yb:KLuW) as active medium.

A methodology similar to the one reported in Ref. 23 was applied for growing high-quality, large-area graphene layers. The monolayer graphene was synthesized by CVD on Cu foils employing a mixture of methane and hydrogen gases. After spin-coating polymethyl-methacrylate (PMMA) on the well-grown monolayer graphene, the underlying Cu foil was etched by aqueous $FeCl_3$ solution. Subsequently, the graphene layer supported by PMMA was transferred onto the whole surface of a 1-in. quartz substrate (thickness: 1 mm) without any other additional treatments and dried on a hot plate. Finally, acetone was used to remove the PMMA layer. The quality and uniformity of the grown monolayer graphene film are verified by the Raman spectroscopy. The linear transmission of the monolayer graphene near 1 μm approaches almost the theoretical value of 97.7% indicating its high quality.

The nonlinear characteristics of the monolayer graphene SA, such as nonlinear transmission and nonlinear response, were investigated near 1.04 μm using a synchronously pumped near-IR tunable femtosecond optical parametric oscillator delivering <200 fs pulses and a maximum pulse fluence on the sample of about 0.4 mJ/cm^2. The nonlinear transmission measurements enable to determine the saturation fluence (F_{sat}), the modulation depth (ΔT), and the non-saturable loss (l_{ns}) of the monolayer graphene SA. The measured transmission change in dependence on the fluence is shown in Fig. 1. From the fit to the data, we extracted $\Delta T \approx 0.75\%$, $F_{sat} \approx 50\ \mu J/cm^2$, and an l_{ns} of 1.59%. These values are well-suited to achieve stable mode-locking of typical bulk lasers. The measured modulation depth was much larger as compared to the values of the single exfoliated graphene layer reported in Ref. 11. This is mainly attributed to the high-quality of our CVD-grown monolayer graphene. Most recently, large nonlinear transmission changes in CVD-grown monlayer graphene were reported.[24] It should be noted that the nonlinear optical characteristics including modulation depth and nonlinear response strongly depend on the quality of graphene layers. Additionally, the measured pump-probe trace (Fig. 1, inset) shows a biexponential decay of saturable absorption with a fast relaxation rate within 200 fs, limited by the pump-probe resolution and a slow 1/e recovery time of ~1.5 ps.

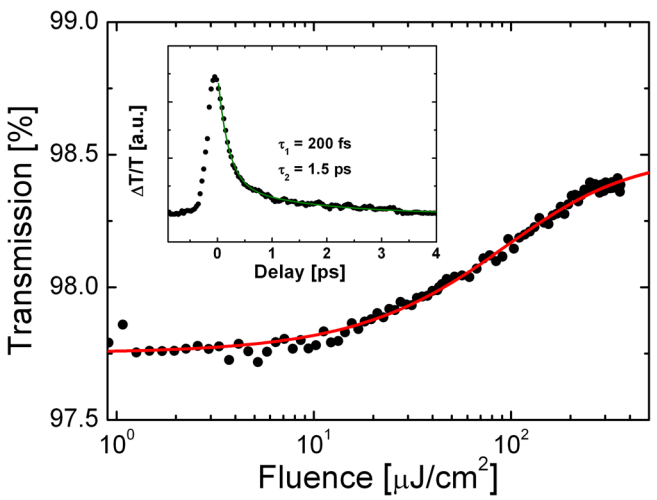

FIG. 1. Nonlinear transmission and pump-probe trace (inset) measurements near 1.04 μm in the monolayer graphene SA.

The laser setup is similar to that applied for the SWCNT-SA mode-locked Yb:KLuW laser[7] and illustrated in Fig. 2. An uncoated, 2.2-mm-thick 10 at. % Yb-doped KLuW crystal with an aperture of 3×8 mm^2 served as active medium. Its plane-parallel surfaces are oriented normal to the N_p-principal optical axis, and the sample is oriented for polarization parallel to N_m and propagation approximately along the N_p-optical axis. The diode used for pumping was a DBR tapered laser diode operated at about 1.5 W of pump power with excellent beam quality at 978 nm.[25] Even though the pump wavelength was about 3 nm away from the absorption peak of Yb:KLuW, approximately 95% of the polarized incident pump radiation was absorbed by the crystal under lasing conditions, independent of the transmission of the output coupler (OC) used. The diode pump beam was focused by an f = 6.28 cm lens through the folding mirror M1. We employed a Z-shaped astigmatically compensated resonator with two folding mirrors (radius-of-curvature: ROC = 10 cm) in the middle to form a 25-μm cavity waist radius at the position of the Yb:KLuW crystal, placed at Brewster angle. One arm contained an additional folding (M3 and M4 both with ROC = 10 cm) where the transmission-type graphene SA was placed in the vicinity of the second waist with a radius

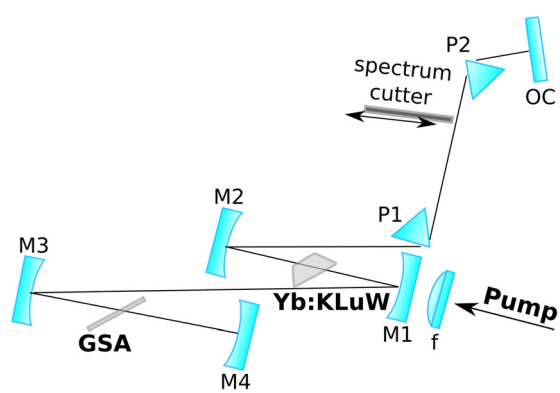

FIG. 2. Setup of the mode-locked Yb:KLuW laser: f: focusing lens; M1, M2, M3, M4: curved mirrors; OC: plane output coupler; GSA: graphene saturable absorber; pump: DBR tapered diode laser; P1, P2: SF10-prisms.

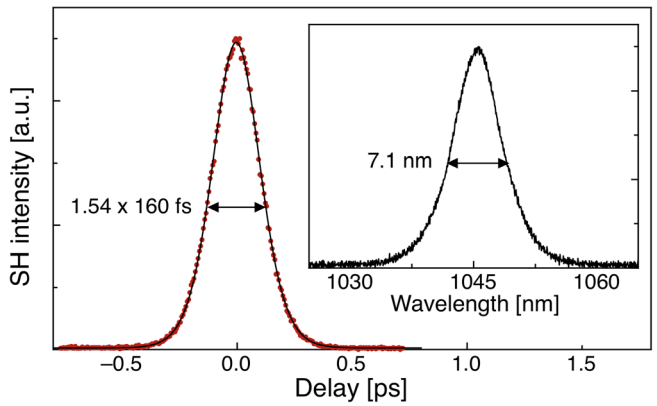

FIG. 3. Graphene SA mode-locked Yb:KLuW laser: Autocorrelation trace (symbols) with a fit (curve) assuming sech^2-shaped pulses and the corresponding optical spectrum (inset).

size of about 32 μm. The other arm contained the plane OC and a prism pair which could be inserted for dispersion compensation.

In the first part of the experiments, the continuous-wave (cw) laser performance was evaluated. Using an OC with 5% transmission and without having the graphene SA and the prism pair incorporated in the cavity, the laser generated a maximum output power of about 350 mW with a slope efficiency of up to 41% at 1030 nm. The laser threshold was reached at 555 mW of absorbed power. A lower threshold of about 413 mW was obtained with 1% OC yielding a maximum output power of 215 mW and a corresponding slope efficiency of 22%.

For femtosecond operation, the graphene SA and two SF10 prisms with a tip-to-tip separation of 39 cm were inserted into the resonator (Fig. 2), resulting in a pulse repetition frequency of 92.5 MHz. The prisms compensate for the positive group-velocity dispersion inside the cavity and balance the self-phase modulation introduced by the Kerr nonlinearity of the laser crystal and the quartz substrate of the absorber. The mode-locking threshold was achieved at an absorbed pump power of 660 mW for the 1% OC yielding an output power of 16 mW. The measured autocorrelation trace could be fitted well assuming sech^2-pulse shapes and is shown in Fig. 3 together with the emission spectrum at maximum output power. For the shortest pulses, the deconvolved FWHM was 160 fs at an average output power of 47 mW. The optical spectrum, centered at 1047 nm, was 7.1 nm wide (FWHM) and the corresponding time-bandwidth product amounts to 0.31, which nearly equals to the Fourier limit. The incident pulse fluence on the graphene SA in the mode-locked regime varied between 0.3 and 1.5 mJ/cm^2. According to the measured saturation fluence (Fig. 1), the absorption is strongly bleached. Fine tuning of the fluence was achieved by slightly displacing the graphene SA around the minimum beam waist in the second folding.

Figure 4 depicts the radio frequency spectrum of the fundamental beat note at 92.52 MHz recorded with a resolution bandwidth (RBW) of 1 kHz and a 1 GHz wide-span measurement (inset) for the shortest pulse operation. The high extinction down to 50 dBc and the absence of any spurious modulation prove clean cw mode-locked operation of the graphene SA mode-locked Yb:KLuW laser. Although the laser was not reliably self-starting, only a slight perturbation was sufficient to initiate mode-locked operation.

Much higher output power of 160 mW was obtained using the 5% OC with only slightly longer pulse duration of 203 fs at the same central wavelength, also displaying bandwidth-limited pulse quality. In this case, the monolayer graphene SA mode-locked Yb:KLuW laser showed improved stability against perturbations and fine spectral tuning was easily realized. The latter was achieved by placing a knife edge between the two prisms (P1, P2) in the cavity. The results are shown in Fig. 5, displaying a tuning range of 14 nm, from 1036 to 1050 nm. When tuning the laser, the pulse duration increased slightly but remained below 300 fs while the output power dropped by not more than ~30%.

Some tendency of double pulsing was observed, in particular, when using the 1% OC. This tendency seems to be related to an additional narrow-band (below the spectral resolution) peak that sometimes appeared in the optical spectrum. Pulses of duration down to ~90 fs were recorded in these conditions but the steady state regime could not be stabilized. We attribute double pulsing to the low modulation depth of the monolayer graphene SA. A more stable operation is expected by choosing a bilayer graphene SA due to the nearly doubled modulation depth.

In conclusion, high-quality monolayer graphene SAs were fabricated by the CVD technique and successfully

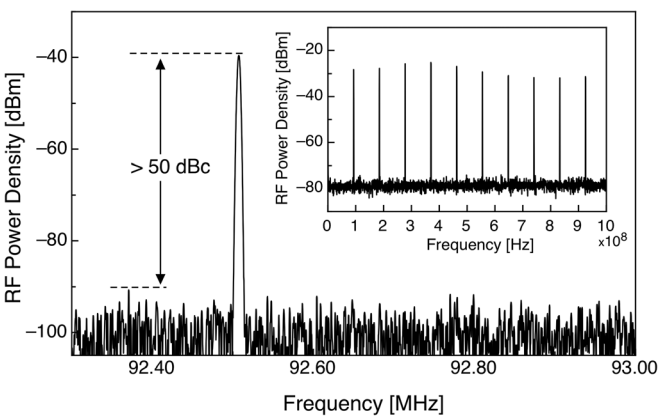

FIG. 4. Radio frequency-spectrum (fundamental beat note) of the graphene SA mode-locked Yb:KLuW laser (RBW: 3 kHz). Inset: 1 GHz wide-span (RBW: 100 kHz).

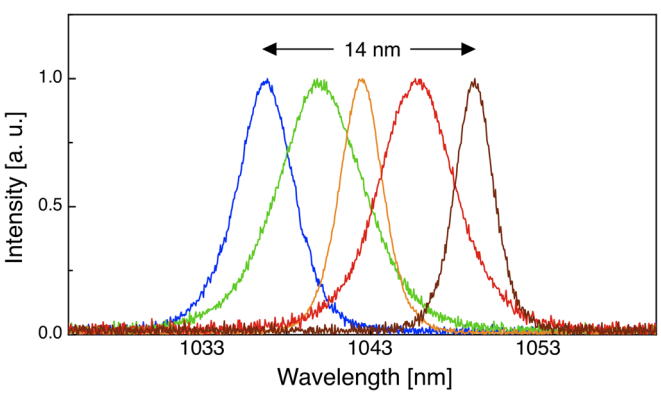

FIG. 5. Spectral tunability of the graphene SA mode-locked Yb:KLuW laser.

applied for femtosecond mode-locking of an Yb-doped KLuW laser. The shortest pulse width of 160 fs measured represents the shortest pulse duration for bulk 1 μm lasers mode-locked by graphene-based SAs. Although a detailed comparison of the present graphene SA mode-locked and the previously reported SWCNT-SA mode-locked Yb:KLuW laser[7] would obviously be very informative, such a comparison could not be performed with sufficient precision as the Yb-doping level and the pump diodes were not identical. Despite these differences, similar performances were achieved. When absorbing approximately the same amount of pump power, the SWCNT-SA mode-locked Yb:KLuW laser delivered 170 fs pulses at 1048 nm with slightly lower output power.[7]

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