High-power, passively mode-locked Nd:GdVO₄ laser using single-walled carbon nanotubes as saturable absorber

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We use a new (to our knowledge) fabrication method of a single-walled carbon nanotube (SWCNT) absorber without polymer to sustain high-power illumination. Using a series of saturable absorbers (SAs) incorporating different amounts of SWCNTs, we demonstrate mode-locking for a Nd:GdVO₄ laser in the 1-μm spectral range. Continuous-wave mode-locking (CWML) pulses with a maximum output power of 3.6 W at 1063 nm and high noise extinction of 61 dB has been achieved to give the highest pulse peak power of 3.6 kW and pulse energy of 30 nJ under 15 W pumping. To our knowledge, this is the highest CWML output power with SWCNT-SAs reported. The measured nonlinear absorption of the SWCNT-SAs shows a modulation depth of ~3% with subpicosecond recovery time. © 2011 Optical Society of America

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High-power and short-pulse lasers operating at a wavelength of 1 μm are of continuous interest in laser physics because they have various practical applications such as wavelength conversion and supercontinuum generation [1], terahertz generation and detection, and nonlinear optical measurements. Although semiconductor saturable absorber mirrors have been commonly used in laser resonators to start up and stabilize the mode locking, they require expensive and complicated fabrication processes such as metal organic chemical vapor deposition [2], and the limitation of spectral operation ranges with available materials. Hence a new material with a broader wavelength operation range and simpler procedure of fabrication is demanded.

Single-walled carbon nanotubes (SWCNTs) have wide application as electro-optic devices due to their excellent electrical and optical properties. Among their prominent features, the fast recovery time, broad spectral range in the near-IR, (NIR) huge third-order optical nonlinearities [3,4], and excellent chemical stability make SWCNTs a promising saturable absorber (SA) for mode locking a laser.

Since the first report of a passively mode-locked fiber laser with SWCNTs [4], SWCNT-SAs have been used for mode locking fiber lasers operated near 1.0 and 1.5 μm [4,5]. Until recently, the SWCNT-SAs have been used to passively mode lock diode-pumped solid-state lasers such as Er/Yb:glass laser at 1.56 μm [6], Cr:forsterite near 1.25 μm [7], Nd:GdVO₄ at 1.34 μm [8], and Yb:KLuW [9] in the 1 μm range. However, the output power of these lasers is still very low (less than 500 mW) due to the low heat resistance of the SWCNT/polymer composite films used as SAs. The reason is that the glass transition temperature ($T_g$) of the polymers, which is a measure of the heat resistance, is too low to operate in high-power lasers [10]. In this Letter, we use SWCNT-SA film, which is fabricated without the polymer material by using a vertical evaporation technique [11], to passively mode lock a Nd:GdVO₄ laser at 1 μm. We obtain the highest output power to our knowledge with the newly prepared SWCNT-SA films.

The SWCNTs used in this experiment were grown by the electric-arc discharge technique. The mean diameter of the SWCNTs is about 1.5 nm. The detailed preparation of SWCNT-SA films can be found in Ref. [11]. We briefly describe it as follows. First, 0.15, 0.3, or 0.6 mg SWCNT powder was dispersed in 10 mL 0.1% sodium dodecyl sulfate aqueous solution in a polystyrene cell. Second, the sedimentation of larger SWCNT bundles was removed. Finally, we inserted a hydrophilic quartz or glass substrate into the cell vertically and kept it steady for complete evaporation into the atmosphere over two weeks. Then the substrate coated with SWCNTs is ready for use as an SA.

The Nd:GdVO₄ laser has a cavity structure similar to the one shown in Ref. [12], but with an output coupler (OC) reflectivity of 89% at 1063 nm. A commercial 3 mm × 3 mm × 8 mm Nd:GdVO₄ crystal with 0.5 at. % Nd³⁺ concentration was used. The radius of the cavity mode at the gain medium was estimated to be ~200 μm using the ABCD law and considering the thermal lensing effect. The SWCNT-SA was put close to the OC. The beam radius at the SWCNT-SA was ~35 μm. Various combinations of OCs and SWCNT-SAs with concentrations of 0.15, 0.3, and 0.6 mg for tuning the initial transmittance and modulation depth were used to optimize the generation of high-power continuous-wave mode-locking (CWML) pulses.

The nonlinear transmittance and saturation fluence were measured by the pump–probe setup described in...
Ref. [11] using the ultrafast regeneratively amplified optical parameter amplifier laser system (TOPAS-C) to provide 100 fs laser pulses at a 1 kHz repetition rate.

The transmission spectra of different concentrations of SWCNT-SAs are shown in Fig. 1(a), which were measured by a UV-visible-NIR spectrophotometer. This measurement is mainly attributed to the distribution of SWCNTs with different diameters. The transmission dip around 1 μm increases as the concentration of SWCNTs is increased. The measured transmissions of samples A, B, and C normalized to those of the substrates are 74%, 82%, and 88% at 1063 nm, respectively.

The measured nonlinear transmission of sample A at 1063 nm is shown in Fig. 1(b). Here we have only shown the result for sample A because the nonlinear transmission signals associated with samples B and C are too weak to be measured. We fitted the pump–probe trace by a biexponential function and obtained a nearly instantaneous (<200 fs) response followed by two decay time constants, 220 fs and 630 fs, respectively. The faster one (220 fs) is attributed to the fast carrier cooling by phonon scattering, and the slower one (630 fs) is the absorption recovery time.

By fitting the absolute transmission of the SWCNT-SAs versus the input pulse fluence in the inset of Fig. 1(d), we obtained the saturation fluence \( F_{\text{sat},A} \approx 68 \mu J/cm^2 \) or the saturation intensity \( I_{\text{sat}} \approx 108 \text{ MW/cm}^2 \) for sample A if 630 fs recovery time is used and the modulation depth (\( \Delta \sigma \)) is about 2.9%. The determined \( I_{\text{sat}} \approx 108 \text{ MW/cm}^2 \) is less than 220 MW/cm\(^2\) of the E\(_{22s}\) transition in Ref. [13], and \( F_{\text{sat},A} \approx 68 \mu J/cm^2 \) is larger than 50 \( \mu J/cm^2 \) of Ref. [14].

The saturable loss and modulation depth are attributed to the absorption of the van Hove E\(_{22s}\) transition of semiconductor SWCNTs and the nonsaturable background absorption (dashed line in Fig. 1), which is due to \( \pi \) plasmas and impurities such as amorphous carbon and metal catalysts [15] and light scattering. Therefore, \( \Delta \sigma \) and nonsaturable loss should increase as the sample thickness increases [16]. The thickness of our sample A is about 20 μm. The transmission spectrum of sample A in Fig. 1 indicates a static loss of 26% with background absorption of 20% and saturable absorption about 6%. The nonsaturable loss is slightly smaller than the sample in Ref. [8] having similar thickness, and it is reasonable to obtain \( \Delta \sigma \sim 2.9% \) out of 6% saturable absorption.

Figure 2 shows the output powers versus pump powers for the CW state (without SWCNT-SA, open black squares) and the CWML states in use of sample A with \( T_0 = 74% \) (blue triangles), sample B with 82% (red circles), and sample C with 88% (green stars). The threshold of CW lasing is 0.9 W with a slope efficiency of 37%. As expected, upon inserting the SWCNT-SAs into the laser cavity, the laser not only increases in CW lasing threshold but also eventually achieves the CWML state (solid symbols) through the intermediate irregular Q-switched mode locking (QML) state (open symbols) and regular QML state (half-filled symbols) for all the samples. To obtain stable CWML, it is required for the intracavity pulse energy \( E_p \) to go beyond a threshold against the Q-switching instability [9]:

\[
E_p > (F_{\text{sat},A}^2 A_I A_A \Delta \sigma)^{1/2}.
\]  

Here \( F_{\text{sat},A} \) and \( A_I \) are the saturation fluence and the laser mode area in the gain medium, \( F_{\text{sat},A} \) and \( A_A \) are those in the SWCNT-SA, and \( \Delta \sigma \) is the modulation depth of the SWCNT-SA. In a standing-wave cavity, the saturation fluences of gain medium and SWCNT-SA can be expressed as \( F_{\text{sat},A} = h\nu/(2\sigma L_{(A)}), \) where \( h\nu \) is the photon energy and \( \sigma L_{(A)} \) is the emission (absorption) cross section. By using Eq. (1), we calculate the CWML threshold for sample A to be about 13 W, which is very close to the measured value of 12.5 W in Fig. 2.
Because sample A has the highest concentrations of SWCNTs, it should present the largest modulation depth and the highest CWML threshold. In order to obtain a lower CWML threshold with higher output power, we have used sample B and obtained the lasing threshold of ~3.1 W with slope efficiency of 29%, and the CWML pulses were obtained as the pump power \( (P_p) \) was raised above 6 W. A remarkable output power of 3.63 W was obtained in CWML regime under \( P_p = 15 \) W. An even lower CWML threshold of 2.5 W can be obtained by using sample C. However, the major limitation of the output power with sample C is the lower damage threshold and the tendency toward multipulsing at high \( P_p \).

The typical CWML pulse train shows the pulse spacing of ~8.2 ns, corresponding to a repetition rate of 121.8 MHz (not shown here), which agrees with the RF spectrum in the inset of Fig. 3. The pulse train observed with the long time scale reveals that the CWML state is free of Q-switching modulation. As shown in Fig. 3 for fundamental beating at 121.8 MHz, a very high extinction ratio of 61 dB against the noise with an absence of any spurious modulations has demonstrated clean CWML operation of the Nd:GdVO\(_4\) laser.

We also measured the mode-locked pulses by an intensity autocorrelator and an optical spectral analyzer (not shown here). By fitting the autocorrelation trace to a Gaussian function, we found that the pulse has an FWHM of 8.4 ps. The optical spectrum is centered at 1063.1 nm with an FWHM of 0.49 nm. It corresponds to the time-bandwidth product of ~1.1, which is larger than the transform-limited value of 0.44 for Gaussian pulses, indicating that the mode-locked pulses are frequency chirped and their duration could be further narrowed. Using the measured average output power of 3.63 W, repetition rate of 121.8 MHz, and pulse width of 8.4 ps, we estimated the pulse energy and peak power to be 30 nJ and 3.55 kW, respectively, at 15 W pumping. The SWCNT-SAs have a better heat sink and thermal damage threshold than SWCNT-SAs with polymer. The mode-locked laser can be operated for about 15 min before degradation by oxidation. Nevertheless, the mode locking lasts for more than an hour if we apply a N\(_2\) purge to the SWCNT-SA.

In conclusion, we have demonstrated a new (to our knowledge) fabrication method for preparing polymer-free SWCNT films to sustain high-power illumination, which is suitable for passive mode locking of solid-state lasers in the 1 \( \mu \)m spectral range. In the use of a SWCNT-SA with a fast recovery time of 630 fs under 15 W pumping, an Nd:GdVO\(_4\) mode-locked laser operating at 1063 nm with a repetition rate of 121.8 MHz and maximum output power of 3.63 W was obtained. It gives the high pulse energy of 30 nJ and peak power of 3.57 kW, free of Q-switching modulation.

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