Energy scaling of a carbon nanotube saturable absorber mode-locked femtosecond bulk laser

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We report successful energy scaling of a room-temperature femtosecond Cr⁴⁺: forsterite laser by using a single-walled carbon nanotube saturable absorber (SWCNT-SA). By incorporating a q-preserving multipass cavity, a repetition rate of 4.51 MHz was realized, and the oscillator produced 121 fs, 10 nJ pulses at 1247 nm, with an average output power of 46 mW. To the best of our knowledge, the peak power of 84 kW is the highest generated to date from a SWCNT-SA mode-locked oscillator. Furthermore, energy scaling of a femtosecond multipass-cavity laser, mode-locked using a SWCNT-SA, is demonstrated for the first time. © 2012 Optical Society of America

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Recently, single-walled carbon nanotube saturable absorbers (SWCNT-SAs) have emerged as viable alternatives to semiconductor saturable absorber mirrors (SESAMs) [1] for initiating mode-locked operation of lasers. SWCNT-SAs can be prepared at low cost without requiring complex growth techniques and possess exceptional optical characteristics. In particular, the peak of the saturable absorption band of SWCNT-SAs can be varied by controlling the diameter distribution and the chirality of the carbon nanotubes. To date, SWCNT-SAs have been utilized to obtain mode-locked operation in lasers operating over a very broad spectral range from 0.8 to 2.1 μm [2–4].

In previous studies, the peak output powers of lasers mode-locked with SWCNT-SAs were limited to a few kilowatts [2,3,5]. Therefore, it is of great interest to explore the possibility of energy scaling in lasers mode-locked with SWCNT-SAs and also the performance of SWCNT-SAs at higher pulse energies.

In this work, we report, for the first time to our knowledge, successful energy scaling in a femtosecond solid-state laser mode-locked with a SWCNT-SA. We chose a room-temperature Cr⁴⁺: forsterite laser operating at 1247 nm to test the SWCNT-SAs because coherent light sources in this wavelength range are ideally suited for numerous biomedical applications such as multiphoton microscopy and optical coherence tomography [6,7]. The experiments were performed in a q-preserving multipass cavity (MPC) that was added to the short Cr⁴⁺: forsterite resonator to obtain higher pulse energies at low average output powers [8]. Negative dispersion was further incorporated into the cavity to balance the nonlinearities. The composite cavity containing the MPC, gain medium, dispersion controlling optics, and the SWCNT-SA produced 121 fs, 10 nJ pulses with a peak power of 84 kW and spectral width of 16 nm. The corresponding time–bandwidth product was 0.37. Limits to energy scaling were further explored in the experiments.

Figure 1 shows the experimental setup of the composite femtosecond Cr⁴⁺: forsterite laser mode-locked using a SWCNT-SA. The short cavity was initially constructed with: two curved dispersion compensation mirrors (DCM) (M1 and M2) each with 10 cm radius of curvature (RC); three flat DCMs (M3, M4, and M5); and a 4.7% transmitting output coupler (M10, shown with dashed lines in Fig. 1). M3 and M4 were used to adjust the inclination angle of the beam entering the multipass cavity. The output coupler and high reflector arm lengths were 50.5 cm and 43.5 cm, respectively. A 20 mm long, Brewster-cut Cr⁴⁺: forsterite crystal with 70% absorption at the pump wavelength was placed between M1 and M2. A continuous-wave (CW) Yb: fiber laser operating at 1064 nm served as the pump source for end-pumping the crystal. The pump beam was focused with a 20 cm lens (L1) to an estimated 30 μm waist inside the crystal. The Cr⁴⁺: forsterite crystal was mounted in a copper holder and maintained at a constant temperature of 20 °C by water cooling.

In order to extend the cavity length, the high reflector flat mirror (M5) was removed and the q-preserving MPC was added to the short laser resonator. The MPC

Fig. 1. (Color online) Schematic of the composite Cr⁴⁺: forsterite laser containing the multipass cavity, dispersion controlling optics, and the carbon nanotube saturable absorber (SWCNT-SA).

contained a curved mirror (M6) with a 4 m RC and a flat mirror (M7). Both M6 and M7 had notches to allow the injection and extraction of the beam. In order to maintain the q-preserving characteristics of the MPC, the distance between M6 and M7 was set to 165 cm. At this separation, the angular advance of the circular spot pattern was 80° per round trip. Before reflecting back to the original cavity, the exiting beam was retro-reflected by a curved end mirror (M9, RC = 2 m) located at a distance of 165 cm from M7 [8]. The MPC provided an additional effective optical path length of 59.4 m. Together with the dispersion controlling optics, the repetition rate of the laser cavity was reduced to 4.51 MHz from 144 MHz. To avoid mode-locking instabilities due to air currents, the extended cavity was further kept inside a box.

To manage the intracavity dispersion and to create a beam waist for effectively saturating the SWCNT-SA, a cavity extension was constructed with dispersion-compensating mirrors on the output coupler side of the short resonator. Here, four flat Gires-Tournois Interferometer (GTI) mirrors (M11, M12, M17, and M18), each with a group delay dispersion (GDD) of -250 fs^2 per bounce and three DCMs (M13, M14, M19), each with a GDD of -150 fs^2 per bounce, were used. To maximize the total negative dispersion, the cavity was aligned such that the beam reflected four times from each GTI mirror in each round trip, as can be seen in Fig. 1. In total, four GTI mirrors with an effective dispersion of -4000 fs^2 and eight DCMs with a total negative dispersion of around -2400 fs^2 were used. After accounting for the dispersion due to the crystal ($+800 \text{ fs}^2$), MPC ($+360 \text{ fs}^2$), SWCNT on quartz substrate ($+7 \text{ fs}^2$), and air ($+794 \text{ fs}^2$), the net GDD of the composite cavity was estimated to be around -4440 fs^2 . The composite resonator was operated by using the 4.7% transmitting mirror as a folded output coupler (OC in Fig. 1), giving an effective output coupling of 9.4%.

The transmission-type SWCNT-SA sample was positioned near the waist of two curved mirrors (M15, M16), each with a RC of 500 mm, yielding an estimated waist of $97 \text{ } \mu\text{m}$. For fabrication of the SWCNT-SA, SWCNTs synthesized by the high-pressure CO conversion (HiPCO) technique were used. The details of the procedure can be found in [9]. To fabricate the transmitting SWCNT-SA, a SWCNTs/PMMA mixture was deposited by spin-coating on a quartz substrate, resulting in a layer thickness of about 300 nm. These SWCNT-SAs exhibit ultrabroadband nonlinear absorption between 1.0 and $1.8 \text{ } \mu\text{m}$, corresponding to the E_{11} transition. The resonant response and the nonlinear transmission of a similar SWCNT-SA at $1.3 \text{ } \mu\text{m}$ were characterized, resulting in a fast relaxation time of $<2 \text{ ps}$, a saturation fluence of about $10 \text{ } \mu\text{J}/\text{cm}^2$, and a modulation depth of 0.5% [9].

Figure 2 shows the efficiency curves for the different resonator configurations of the Cr^{4+} : forsterite laser in the CW regime. The threshold pump power and the slope efficiency for the short x-cavity were measured to be 2.13 W and 13.7%, respectively. With the addition of the MPC, the threshold power increased to 2.63 W, and the slope efficiency decreased to 12.4%. Moreover, by adding the dispersion control optics and the SWCNT-SA, the threshold power increased to 5.13 W, and the slope efficiency decreased to 10.2%. The passive

Fig. 2. Power efficiency curves during CW operation of the Cr^{4+} : forsterite laser with different resonator architectures (DCO, dispersion control optics).

loss of the Cr^{4+} : forsterite crystal was around 3.14%. By using the threshold power analysis, the additional losses due to the MPC, the dispersion control optics, and the SWCNT-SA were estimated as 1.72%, 0.8%, and 3.76%, respectively. Note that the 3.76% loss of the SWCNT-SA estimated in the CW regime is the sum of the saturable and nonsaturable loss and reduced by the saturable loss in the mode-locked regime. In the final configuration including the SWCNT-SA and the 9.4% effective output coupling, as much as 280 mW of CW output power was obtained at 20 °C temperature with 8.0 W of input power.

In the mode-locked regime, stable single-pulse operation was obtained at 1247 nm with the SWCNT-SA up to a total output power of 46 mW in two output beams. At this power level and for the repetition rate of 4.51 MHz, the incident fluence was determined to be $368 \text{ } \mu\text{J}/\text{cm}^2$, which produced sufficient saturation to start mode locking. The spectrum and the autocorrelation trace at the output power of 46 mW are shown in Fig. 3. The FWHM spectral bandwidth and the pulsewidth were measured to be 16 nm and 121 fs, respectively. The pulse width was determined by assuming a sech^2 intensity profile. The resulting time bandwidth product of 0.37 suggests nearly transform limited pulses. The CW peak around 1290 nm is believed to be due to the GDD oscillations of the GTI mirrors. The average output power of 46 mW at the repetition rate of 4.51 MHz corresponds to total pulse energy of around 10 nJ and, for 121 fs pulses, to a peak

Fig. 3. Spectrum and collinear autocorrelation (inset) of the Cr^{4+} : forsterite laser mode-locked with SWCNT-SA.

power of 84 kW. These results were obtained at the input pump power of 6 W.

The nonlinear refractive index of the Cr⁴⁺: forsterite medium can be estimated by using the experimental mode-locking data and the well-known soliton-area theorem which relates the pulse energy to the pulsewidth through the relation [10]

$$W\tau = \frac{4|D|}{\gamma}. \quad (1)$$

In Eq. (1), W is the intracavity energy, D is the net GDD of the cavity, and τ is the pulse width (FWHM) divided by 1.76. γ is the net nonlinear coefficient due to the Cr⁴⁺: forsterite crystal and the air, and can be calculated from

$$\gamma = \gamma_g + \gamma_{\text{air}} = \frac{2\pi}{\lambda} \left(n_2 \frac{2l_g}{A_{\text{eff}}^g} + n_2^{\text{air}} \frac{2l_{\text{air}}}{A_{\text{eff}}^{\text{air}}} \right), \quad (2)$$

where, λ is the center wavelength; n_2 and n_2^{air} are the nonlinear refractive indices of the Cr⁴⁺: forsterite crystal and air, respectively; l_g and l_{air} are the crystal length and air path length, respectively; and A_{eff}^g and $A_{\text{eff}}^{\text{air}}$ are the respective effective areas of the beam inside the crystal and in the air region of the composite cavity. Even though the MPC introduces a long optical path length of 59.4 m in air, due to the small value of n_2^{air} at 1247 nm [11], the nonlinear coefficient of air was negligible. Using the soliton-area theorem and attributing all the cavity nonlinearities to the crystal, the n_2 value of Cr⁴⁺: forsterite was determined to be $9.36 \times 10^{-20} \text{ m}^2/\text{W}$, which is in reasonable agreement with the value of $6 \pm 4 \times 10^{-20} \text{ m}^2/\text{W}$, previously reported by Chassagne *et al.* [12].

In the mode-locking experiments, any further increase in the output power above 46 mW resulted in multipulse operation. At this power level with 9.4% effective output coupling, the maximum intracavity pulse energy was 108.5 nJ, corresponding to a peak power of 0.9 MW. By using the n_2 value for Cr⁴⁺: forsterite determined here, the critical power for self focusing was estimated to be 1.5 MW [13], which is in reasonable agreement with the experimentally observed limiting peak power of 0.9 MW. To further confirm the value of the limiting intracavity power, the laser was operated by replacing the locations of M19 and OC to give an output coupling of 4.7%. In this case, the CW output power increased from 210 to 295 mW at the pump power of 7.1 W because 4.7% output coupling was closer to the optimum output coupling of the resonator (6%). However, during mode-locked operation, the maximum single-pulse output energy was limited to 5 nJ, confirming the maximum intracavity peak power of 0.9 MW. In principle, the output energy during single-pulse operation can be further increased by increasing the output coupling of the resonator. However, in the experiments, the Cr⁴⁺: forsterite oscillator could not be operated with output coupling

beyond 11% at room temperature, suggesting that any further increase in output coupling will not improve energy scaling. Alternatively, the intracavity GDD can be further increased by using more advanced GTI designs. However, we expect that this approach will increase both the pulse energy and the pulse width, keeping the peak intracavity power below the experimentally observed 0.9 MW limit.

In conclusion, we have successfully demonstrated energy scaling of a room-temperature MPC Cr⁴⁺: forsterite oscillator mode-locked using a SWCNT-SA and generated 10 nJ, 121 fs pulses at 1247 nm with an average output power of 46 mW and a peak power of 84 kW. To our knowledge, this is the highest peak power directly generated from a mode-locked laser by using SWCNT-SAs. Our results further indicate that the peak power limitation in the experiments arose only due to self-focusing-induced multipulse instabilities in the Cr⁴⁺: forsterite gain medium, and further energy scaling should be possible in other types of lasers mode-locked with SWCNT-SAs.

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References

1. U. Keller, K. J. Weingarten, F. X. Kartner, D. Kopf, B. Braun, I. D. Jung, R. Fluck, C. Hönninger, N. Matuschek, and J. A. d. Au, *IEEE J. Sel. Top. Quantum Electron.* **2**, 435 (1996).
2. I. H. Baek, S. Y. Choi, H. W. Lee, W. B. Cho, V. Petrov, A. Agnesi, V. Pasiskevicius, D. I. Yeom, K. Kim, and F. Rotermund, *Opt. Express* **19**, 7833 (2011).
3. W. B. Cho, J. H. Yim, S. Y. Choi, S. Lee, U. Griebner, V. Petrov, and F. Rotermund, *Opt. Lett.* **33**, 2449 (2008).
4. A. Schmidt, P. Koopmann, G. Huber, P. Fuhrberg, S. Y. Choi, D. I. Yeom, F. Rotermund, V. Petrov, and U. Griebner, *Opt. Express* **20**, 5313 (2012).
5. Y. Liu, Y. Wang, J. Liu, and C. Liu, *Appl. Phys. B* **104**, 835 (2011).
6. T.-M. Liu, S.-W. Chiu, C.-K. Sun, B.-L. Lin, P. C. Cheng, and I. Johnson, *Scanning* **23**, 249 (2001).
7. G. J. Tearney, M. E. Brezinski, B. E. Bouma, S. A. Boppart, C. Pitris, J. F. Southern, and J. G. Fujimoto, *Science* **276**, 2037 (1997).
8. A. Sennaroglu, A. M. Kowalevich, E. P. Ippen, and J. G. Fujimoto, *IEEE J. Quantum Electron.* **40**, 519 (2004).
9. W. B. Cho, J. H. Yim, S. Y. Choi, S. Lee, A. Schmidt, G. Steinmeyer, U. Griebner, V. Petrov, D. I. Yeom, K. Kim, and F. Rotermund, *Adv. Funct. Mater.* **20**, 1937 (2010).
10. H. A. Haus, *IEEE J. Sel. Top. Quantum Electron.* **6**, 1173 (2000).
11. V. Y. Fedorov and V. P. Kandidov, *Opt. Spectrosc.* **105**, 280 (2008).
12. B. Chassagne, A. Ivanov, J. Oberle, G. Jonusauskas, and C. Rulliere, *Opt. Commun.* **141**, 69 (1997).
13. D. Huang, M. Ulman, L. H. Acioli, H. A. Haus, and J. G. Fujimoto, *Opt. Lett.* **17**, 511 (1992).