

# Recent progress in carbon nanotube saturable absorbers for ultrafast bulk solid-state lasers

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## Abstract

Carbon nanotubes are one of the most investigated nanomaterials due to their unique electric and optical properties. In recent years, the nonlinear optical characteristics were intensively investigated not only for electronic, but also for photonic applications. In this work, recent progress in novel saturable absorbers based on single-walled carbon nanotubes (SWCNTs) applicable for passive mode-locking of ultrafast bulk solid-state lasers is reviewed. Important linear and nonlinear optical characteristics of SWCNT saturable absorbers and their application as ultrafast mode-locking devices for different bulk lasers in the spectral range between 800 nm and 2  $\mu\text{m}$  will be shown.

## 1. Introduction

Saturable absorbers based on semiconductor heterostructures, i. e. semiconductor saturable absorber mirrors (SESAMs), have revolutionized ultrafast laser technology [1]. They provide a robust nonlinear optical switching mechanism, which enabled applications for ultrafast lasers. In recent years, SWCNT-based saturable absorbers (SWCNT-SAs) were successfully developed and employed for mode-locking of different fiber and bulk lasers. These novel absorbers exhibit broad absorption with large third-order nonlinearities and require relatively simple fabrication processes, whereas SESAMs provide a spectrally narrowband nonlinearity and require sophisticated manufacturing processes with additional treatments to decrease the response time. To overcome the limiting characteristics of SESAMs, SWCNT-SAs are suggested as one alternative. The absorption band of SWCNT-SAs is controllable by varying the nanotube diameter and chirality. Depending on the electronic transitions of semiconducting nanotubes, SWCNT-SAs are readily applicable within a broad spectral range throughout the near-infrared from 800 nm up to 2  $\mu\text{m}$ . SWCNT-SA was successfully utilized as mode-locking device in Er-doped fiber lasers operating near 1.5  $\mu\text{m}$  [2]. Up to now, passive mode-locking with SWCNT-SAs is still mostly restricted to fiber lasers, although passive mode-locking of bulk solid-state lasers was also reported near 1.3 and 1.5  $\mu\text{m}$  [3-4]. Since the single-pass gain of fiber lasers is much higher, they can easily tolerate large non-saturable losses. For application in bulk solid-state lasers, however, it is mandatory to reduce these losses to the lowest level possible. Here we present linear and nonlinear optical characteristics of SWCNT-SAs that exhibit low linear losses, small modulation depths, and fast recovery times comparable to SESAMs. Additionally, applications of different SWCNT-SAs as mode-locker in different bulk solid-state lasers operating in the spectral range between 800 nm and 2  $\mu\text{m}$  will be shown.

## 2. Fabrication of SWCNT-SAs

For fabrication of SWCNT-SAs, SWCNTs of different kinds, grown by Arc-discharge and high-pressure CO conversion (HiPCO) methods, were used as starting materials. The thermo-gravimetric analysis showed purity as high as 90% in weight. The SWCNTs were dried in vacuum and these with different concentrations between 0.1 - 0.3 mg/ml were dispersed in dichlorobenzene (DCB) with ultrasonication process. The SWCNT dispersion was subsequently mixed with a separately prepared poly(methyl methacrylate) (PMMA) solution. Before stirring the prepared SWCNT/PMMA mixture, the mixture was again treated by the ultrasonic process for several minutes. The SWCNT absorber layer was deposited on substrates by spin-coating process. Uncoated quartz substrates were used for deposition. Alternatively, SWCNT layer could be directly deposited on commercial highly reflecting dielectric mirrors for manufacturing reflective devices. The coated samples were finally baked at 150°C for minutes. The resulting typical thickness of the coated SWCNT/PMMA layer was measured to be about 200-500 nm. For controlling the absorption level of the samples, we could slightly change SWCNT concentrations in the solution or layer thicknesses. The deposited absorber layer was uniform across the whole substrate. Figure 1 shows typical transmission spectra of transmitting SWCNT-SAs applicable for bulk laser mode-locking near 1- 2  $\mu\text{m}$ .

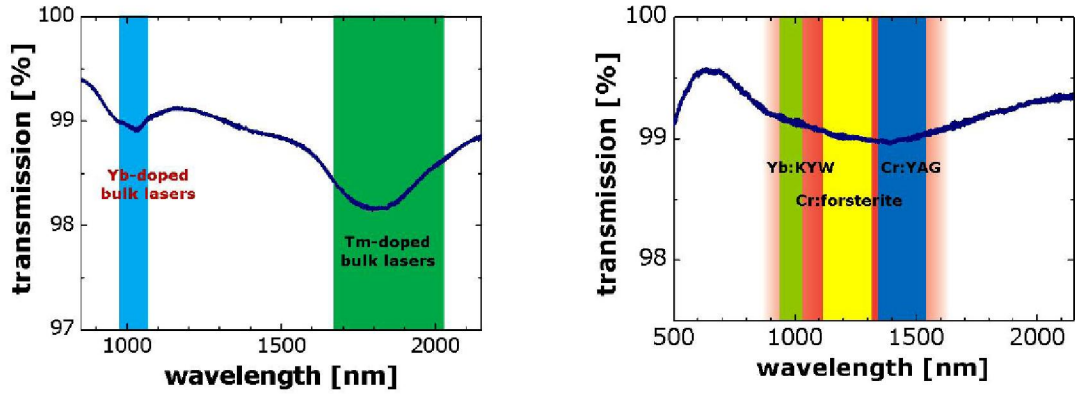


Fig. 1 Transmission spectra of SWCNT-SAs, applicable for different bulk solid-state lasers near 1-2  $\mu\text{m}$ .

Further development in sample fabrication, in which differently-grown SWCNTs with different chiralities and diameters were mixed and dispersed in DCB allowed to extend the operation bandwidth of saturable absorbers. We could fabricate octave-spanning SWCNT-SAs. This single device were applied for laser mode-locking at wavelengths between 1-2  $\mu\text{m}$ . Note that the optimized  $E_{22}$  transition of HiPCO SWCNTs, which is located around 800 nm, enabled the fabrication of saturable absorbers for mode-locking of the widespread Ti:sapphire laser.

### 3. Mode-locking of bulk solid-state lasers

The first bulk laser mode-locking with a SWCNT-SA was demonstrated near 1.5  $\mu\text{m}$  with Er/Yb:glass [3]. This successful demonstration motivated further mode-locking investigations on other bulk lasers in adjacent wavelength regions. Recently, we demonstrated SWCNT-SA mode-locking of Yb-doped bulk lasers in the 1  $\mu\text{m}$  range and of Cr:forsterite and Cr:YAG lasers near 1.25 and 1.5  $\mu\text{m}$ , respectively, which delivered about 100 fs pulses [5-8]. We also achieved SWCNT-SA mode-locked operation of a Tm-doped KLuW laser in the wavelength range around 2  $\mu\text{m}$  [9]. Figure 2 shows typical mode-locked laser spectra and the corresponding autocorrelation traces.

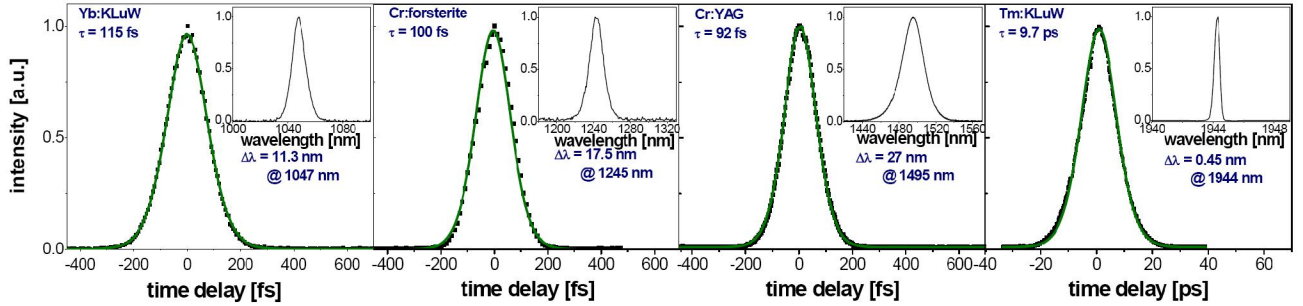
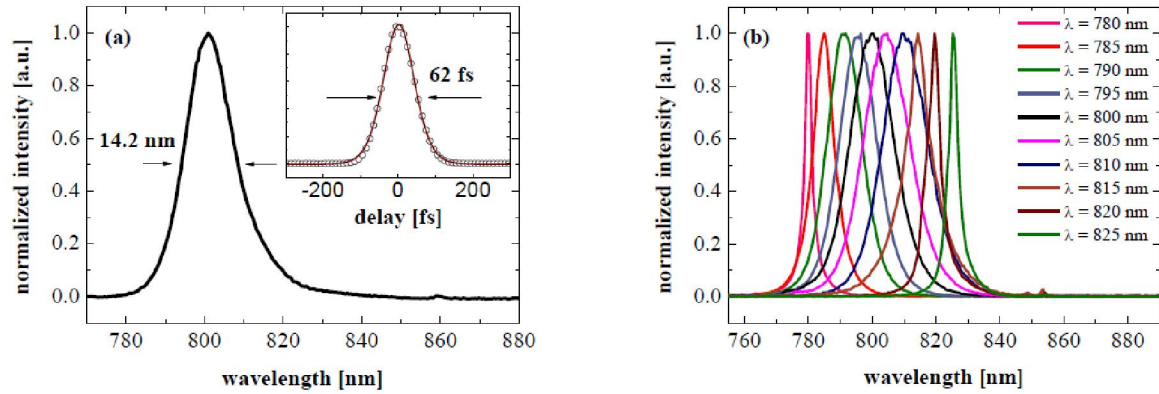


Fig. 2 SWCNT-SA mode-locked laser spectra and the corresponding autocorrelation traces.

Most recently, we could demonstrate passive mode-locking of a Ti:sapphire laser employing a SWCNT-SA specially designed and fabricated for wavelengths around 800 nm. Mode-locked pulses as short as 62 fs were generated at a repetition rate of 99.4 MHz. We achieved output powers from the SWCNT-SA mode-locked laser as high as 600 mW with a slope efficiency of 26%. The nonlinear response of the SWCNT-SA was measured at 800 nm by non-collinear cross-polarized pump-probe spectroscopy. The pump-probe trace indicates a nearly instantaneous response of about 150 fs and a slow exponential decay of  $< 1.2$  ps. These intraband and interband recovery times are comparable to the values measured for the  $E_{11}$  interband transition of HiPCO SWCNTs, which was utilized for Cr-doped bulk lasers in our mode-locking experiments. Figure 3 shows the mode-locked laser spectrum and the corresponding autocorrelation curve. The operation wavelength could be slightly tuned in the spectral range between 780-825 nm (fig. 3(b)).



**Fig. 3** SWCNT-SA mode-locked Ti:sapphire laser: (a) spectrum and autocorrelation trace and (b) tunability.

## 4. Conclusion

We fabricated novel saturable absorbers based on SWCNTs which can be applied for mode-locking different bulk solid-state lasers in a wide spectral range between 800 nm and 2  $\mu$ m. These devices are not only cheaper and simpler to manufacture in a reproducible manner, but also offer very attractive properties such as low saturation fluence, high damage threshold, and extremely fast relaxation, which make them a promising alternative for replacing semiconductor-based SAs.

## 5. Acknowledgments

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## 6. References

1. U. Keller, K. J. Weingarten, F. X. Kärtner, D. Kopf, B. Braun, I. D. Jung, R. Fluck, C. Hönninger, N. Matuschek, and J. Aus der Au, *IEEE J. Sel. Top. in Quantum Electron.* **2**, 435 (1996).
2. S. Y. Set, H. Yaguchi, Y. Tanaka, and M. Jablonski, *J. Lightwave Technol.* **22**, 51 (2004).
3. T. R. Schibli, K. Minoshima, H. Kataura, E. Itoga, N. Minami, S. Kazaoui, K. Miyashita, M. Tokumoto, and Y. Sakakibara, *Opt. Express* **13**, 8025 (2005).
4. K. H. Fong, K. Kikuchi, C. S. Goh, S. Y. Set, R. Grange, M. Haiml, A. Schlatter, and U. Keller, *Opt. Lett.* **32**, 38 (2007).
5. J. H. Yim, W. B. Cho, S. Lee, Y. H. Ahn, K. Kim, H. Lim, G. Steinmeyer, V. Petrov, U. Griebner, and F. Rotermund, *Appl. Phys. Lett.* **93**, 161106 (2008).
6. A. Schmidt, S. Rivier, G. Steinmeyer, J. H. Yim, W. B. Cho, S. Lee, F. Rotermund, M. C. Pujol, X. Mateos, M. Aguiló, F. Díaz, V. Petrov, and U. Griebner, *Opt. Lett.* **33**, 729 (2008).
7. W. B. Cho, J. H. Yim, S. Y. Choi, S. Lee, U. Griebner, V. Petrov, and F. Rotermund, *Opt. Lett.* **33**, 2449 (2008).
8. 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).

9. W. B. Cho, A. Schmidt, J. H. Yim, S. Y. Choi, S. Lee, F. Rotermund, U. Griebner, G. Steinmeyer, V. Petrov, X. Mateos, M. C. Pujol, J. J. Carvajal, M. Aguiló, and F. Díaz, *Opt. Express* **17**, 11007 (2009).