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Mode-Locked Fiber Lasers Using Adjustable Saturable Absorption in Vertically Aligned Carbon Nanotubes

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We report an adjustable saturable absorber based on vertically aligned single-walled carbon nanotubes (SWNTs). Using the low-temperature alcohol catalytic chemical vapor deposition (CVD) method, high-quality vertically aligned SWNTs were directly synthesized onto quartz substrates. The saturable absorption is adjustable by changing the decline angle of the substrate. We applied it as a mode-locker in the passively mode-locked fiber lasers, and optimized the operation by declining the vertically aligned SWNT sample. [DOI: 10.1143/JJAP.45.L17]

KEYWORDS: fiber laser, mode lock, pulse generation, saturable absorber, carbon nanotube

Passively mode-locked fiber lasers have been used in many applications in various fields, such as optical communications, optical signal processing, two-photon microscopy, laser surgery etc., due to their simplicity and their ability to generate transform-limited optical short pulse in sub-picosecond regimes.^{1,2)} Such lasers offer superb pulse quality and there is no need for costly modulators as required in actively mode-locked lasers. Instead, a passively mode-locked fiber laser employs a mode-locker, a device that possesses an intensity-dependent response to favor optical pulse formation over continuous-wave lasing. This is usually a fast saturable absorber, such as a semiconductor-based saturable absorber, or a fiber-based saturable absorber. Amongst these mode-lockers, the semiconductor-based multi-quantum-well (MQW) device, commonly referred as the semiconductor saturable absorber mirror (SESAM), has become the main device used in almost all commercial passively mode-locked fiber lasers.

Instead of SESAMs, we recently demonstrated a saturable absorber incorporating carbon nanotube (SAINT),^{3–7)} which offers many advantages such as ultra-fast recovery time (<1 ps), simplicity, robustness, etc. The SAINT can be directly formed onto the substrates or the cleaved fiber end.⁵⁾ In these studies, we used single-walled carbon nanotubes (SWNTs) whose axial directions are randomly oriented on the substrates, because it was not possible to control the direction during fabrication. Recently, some of the authors have succeeded in fabricating vertically aligned SWNTs by a simple chemical vapor deposition (CVD) process.^{8,9)}

In this paper, we present a passively mode-locked fiber lasers using the vertically aligned SWNTs. The vertically aligned SWNT has an advantage that the amount of the saturable absorption is adjustable by declining the substrate due to the absorption anisotropy against the direction of the incident electric field. We optimize the decline angle of the substrate so as to maximize the output power and minimize the pulse width.

Vertically aligned SWNTs were grown at 800°C and 10 Torr using an alcohol CVD process.^{8,9)} The CVD reaction was catalyzed by Mo/Co bimetal nanoparticles, which were

affixed to quartz substrates by a dip-coat method (0.01 wt %Mo/Co acetate). Subsequent annealing in air at 400°C oxidized the nanoparticles, preventing catalyst agglomeration in the high-temperature CVD environment. Flowing Ar/H_2 (3% H₂) during preheating of the reaction chamber reduced the catalyst in order to retrieve catalyst activity before the growth stage. When the CVD chamber reached 800° C, the Ar/H₂ flow was stopped, followed by the introduction of ethanol vapor. The reaction time was varied in order to observe samples at different stages of growth. The samples were characterized with a Hitachi S-4700 fieldemission scanning electron microscope (FE-SEM). Figure 1(a) shows a synthesized thick mat of vertically aligned SWNTs on a quartz substrate surface. An advantage of the low-temperature alcohol catalytic CVD method is that it can synthesize SWNTs directly onto various samples, including cleaved end of fibers. We used vertically aligned SWNT sample directly synthesized onto the quartz surfaces.

The saturable absorption comes from the energy bandgaps in semiconducting SWNTs. The bandgap energy is determined by the tube diameter of SWNT. A tube diameter of \sim 1.2 nm gives absorption at wavelength around 1550 nm. It is known that SWNTs absorb light whose electric fields are parallel to the tube axis, thus it turns out that there should be no extonic absorption when the light is incident to the vertically aligned SWNTs perpendicularly from the top. As the substrate is declined, the absorption begins to be caused, and it is possible to adjust the amount of absorption. Figure 1(b) shows the absorption spectra of the vertically aligned SWNTs at the decline angles of 0, 15, 30, and 45 deg. The absorption band is found to be wideband from 1000 to 1600 nm, and the amount of absorption becomes larger as the SWNT sample is declined further. This was not possible in the previous randomly oriented SWNT samples, and it is useful for applications in saturable absorbers.

The schematics of the mode-locked fiber ring laser are shown in Fig. 2. An erbium-doped fiber amplifier (EDFA) is used as the laser gain medium. The output light from the EDFA is launched through a fiber collimator and focused onto the vertically aligned SWNT sample. The output light from the SWNT sample is collected and launched back into the fiber cavity via another fiber collimator. A 10-m-long

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Fig. 1. Vertically aligned SWNTs synthesized directly onto quartz substrate. (a) SEM image and (b) absorption spectrum.



Fig. 2. Mode-locked fiber ring laser incorporating a vertically aligned SWNT sample.

single-mode fiber (SMF) is inserted into the cavity to adjust the chromatic dispersion in the cavity and to stabilize the pulse formation. A polarization controller in the cavity is used to adjust the round-trip polarization states. The output of the laser is tapped through a 5% port of a 95 : 5 fiber coupler, whereas the other 95% port is used to feed back into the laser cavity. Optical isolators are inserted to prevent the back-reflection in the cavity and to ensure the unidirectional operation.

When the SWNT sample was not declined, the light beam was perpendicular to the substrate, and the laser would not start to be mode-locked even if the EDFA gain is at its maximum. This shows that there is no saturable absorption when the light beam is parallel to the axis of SWNTs. In the case of the sample decline angle of ~ 10 deg, the laser started to be mode-locked and produced multiple pulses in a round-trip time when the EDFA gain reached around 8.3 dB. Once mode-locked, the laser could produce the single pulse train by adjusting the polarization controller. The EDFA gain could be reduced to ~ 5.6 dB with maintaining mode-locking at a fundamental repetition rate of 6.62 MHz. The average

output power with the EDFA gain of $6.6 \,\text{dB}$ was $-16.6 \,\text{dB}$ m. The output spectrum, the autocorrelation trace, and the pulse waveform of the output pulses are shown in Figs. 3(a), 3(b), and 3(c), respectively. The autocorrelation trace and spectrum are well fitted by a Gaussian pulse profile. The inferred full-width half maximum (FWHM) width from the autocorrelation trace is estimated to be 1.14 ps, whilst the 3 dB spectral width is 3.05 nm. The uneven pulse amplitude in Fig. 3(c) is an artifact of a limited sampling point of the digital scope.

Figure 4 shows the average output powers and pulse widths as a function of the decline angle from 1 to 45 deg. It is found that the laser could operate at a wide range of decline angle from 1 to 45 deg, and around 30 deg was the optimum angle to maximize the output power and minimize the pulse width in this experimental setup. Decline angle beyond 45 deg was not possible due to the limitations imposed by the closely positioned collimators.

In conclusion, we reported an adjustable saturable absorber using vertically aligned SWNTs. Using the lowtemperature alcohol catalytic CVD method, high-quality vertically aligned SWNTs were directly synthesized onto quartz substrates. We confirmed that the saturable absorption is adjustable by changing the decline angle of the substrate. We applied it as a mode-locker in the passively mode-locked fiber lasers, and optimized the pulse width and output optical power by declining the vertically aligned SWNT sample.

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Fig. 3. Laser outputs at the decline angle of 10 deg. (a) Optical spectrum, (b) autocorreletion trace, and (c) pulse waveform.



Fig. 4. Pulse power and pulse width with different sample angles.

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