Wideband Tunable, Nanotube Mode-locked Fiber Lasers

F. Wang[1], A. G. Rozhin[1], V. Scardaci[1], Z. Sun[1], F. Hennrich[2], I. H. White[1], W. I. Milne[1], A. C. Ferrari[1] *

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[1]Department of Engineering, University of Cambridge, 9 JJ Thomson Avenue, Cambridge, CB3 0FA, UK

[2]Institut für Nanotechnologie Forschungszentrum Karlsruhe, 76021 Karlsruhe, Germany

Ultrashort lasers with spectral tuning capability have widespread applications in fields such as spectroscopy, biomedical research and telecommunications[1, 2, 3]. Mode-locked fiber lasers are convenient and powerful sources of ultrashort pulses[4]. The key component to achieve wide tunability is a broadband saturable absorber, acting as a passive optical switch inside the laser cavity[5]. Semiconductor saturable absorber mirrors (SESAMs) are the present standard technology[4, 5, 6]. They are single or multiple quantum-wells embedded in a mirror structure [6]. However, their operating range is limited to a few ten nanometers[7, 8]. Also, for the technologically important spectral regions around 1.3-1.5 μm, the fabrication of wideband SESAMs is challenging[9, 10]. Single-wall carbon nanotubes (SWNTs) are ideal saturable absorbers because of their outstanding properties such as sub-picosecond recovery time, low saturation power, polarization insensitivity, mechanical and environmental robustness[11, 12, 13, 14, 15, 16, 17]. In principle, a combination of different diameters and chiralities could allow the operating wavelengths of SWNTs-based devices to cover

*acf26@eng.cam.ac.uk

a much broader bandwidth than any other system. Here, we engineer a SWNTs-polycarbonate film with a wide bandwidth (>300 nm) around 1.55 μm. We then use it to demonstrate a 2.4 ps Er^{3+} fiber laser tunable from 1518 to 1558 nm. This proves that SWNTs are ideal for compact, broadly tunable mode-locked lasers, and paves the way to their practical implementation.

The development of compact, diode-pumped, ultrafast fiber lasers as alternatives for bulk solid-state lasers is fast progressing, and is fueled by the recent advance of mature fiber technology[4]. This poses extra requirements on the existing mode-locking techniques, such as wide operating bandwidth, low saturation fluence, fiber-compatibility, etc. At present, the dominating technology in passively mode-locked lasers is based on semiconductor saturable absorber mirrors (SESAMs)[6]. Conventional SESAMs are III-V semiconductor multiple quantum wells grown on distributed Bragg reflectors (DBRs)[3]. Their fabrication involves complex molecular beam epitaxy and treatments such as post-growth ion-implantation, to reduce their relaxation time to sub-picosecond[6]. Furthermore, SESAMs are based on a resonant nonlinearity which tends to limit wavelength tunability [18]. Their operating bandwidth is further limited by the bottom DBR section, which has a finite bandwidth for high reflectivity[18]. For example, the bandwidth of conventional $Al_xGa_{1-x}As/AlAs$ SESAMs is limited to about 60 nm by the bottom Bragg mirrors [19]. Wider reflection bandwidth in excess of 200 nm was achieved using novel material pairs with larger refractive index difference (e.g. AlGaAs/CaF_2) [19], or replacing the DBRs with metallic mirrors [20]. But, so far, no tunable mode-locked laser was reported using these novel structures. In addition, problems, such as lattice mis-match and poor thermal properties, seriously limit the quality of SESAMs grown for wavelengths above 1.3 μm[9, 10]. Tradeoffs between design parameters have to be made in order to obtain targeted device characteristics[8]. So far, the widest tuning range demonstrated in a SESAM is 125 nm, achieved for a Yb-fiber laser operating at 1μm. However, two SESAMs with complementary spectral properties had to be used[21]. Thus, much simplified and cost-effective saturable absorbers with wideband tunability are needed.

Single wall carbon nanotubes (SWNTs) are at the centre of nanotechnology research. They are direct bandgap materials, with a gap dependent on tube diameter and chirality [22]. A variety of possible uses are suggested for this unique form of carbon, such as transistors, composites and field emission devices. However, many key applications in electronics require individual tubes with given chirality for their optimum performance. Nowadays, it is not possible to grow such well-defined SWNTs. Here, we use the optoelectronic properties of SWNTs for photonics, by exploiting SWNTs with a range of chiralities and diameters, thus turning one of the major disadvantages of SWNT technology into the key element for success of our work.

SWNTs are a new class of saturable absorbers [11], with large optical nonlinearity [23], ultrafast carrier relaxation time[24] and high damage threshold [25]. They are compatible with optical fibers, and operation at $\sim$1.5 μm has been demonstrated [15, 27, 25, 28, 29, 30, 26, 16, 17]. In a tunable laser, it is very important that the mode locker maintains a sufficiently large modulation depth over a wide spectral range. This has been one limiting factor for achieving wider tunability in SESAMs-based lasers[10]. In the case of SWNTs, the variation of nonlinear absorption is determined by the amount of tubes in resonance with the incident light. However, despite a wavelength de-tuning of up to ($\sim$200 nm) from the peak resonance wavelength, appreciable saturable absorption of SWNTs can still be observed [23]. This implies great potential for wideband tunable lasers. Previous SWNTs-based mode-lockers were based on SWNTs solutions [13], SWNTs layers grown or spray-coated over optical parts [27] or SWNTs-polymer composites [15, 26]. The latter allows homogeneous dispersion and easy integration.

We fabricate a SWNTs-polycarbonate composite with a broad ($\sim$ 300nm) absorption band in the 1.5 μm spectral region, as described in Methods, and shown in Fig.1(a). This is achieved by using SWNTs with diameters from 1 to 1.3 nm. Note that pure polycarbonate does not exhibit any absorption around 1550 nm (Fig. 1(a)). The mode-locker is assembled by sandwiching the free-standing SWNTs composite between two fibre ferrules inside a physical contact ferrule connector (FC/PC), as described in Methods. The estimated mode diameter on the composite is 10 μm.

We first investigate the wavelength-dependent saturable absorption within the Er^{3+} fiber gain bandwidth, as described in Methods. Fig. 1(b) plots the measured loss as a function of average pump power at different pump wavelengths. All loss curves show a saturation depth about 0.3dB, against a linear loss of 2.65 dB (or, equivalently, a 4% increase in transmittance from a small-signal transmission of 54.3%). A slight decrease of modulation depth is detected towards shorter wavelengths, possibly caused by the mismatch between the pump wavelengths and the peak absorption of our SWNTs ($\sim$1550 nm). Further wavelength de-tuning would lead to a more pronounced decrease in the modulation depth. The data are then fitted according to a

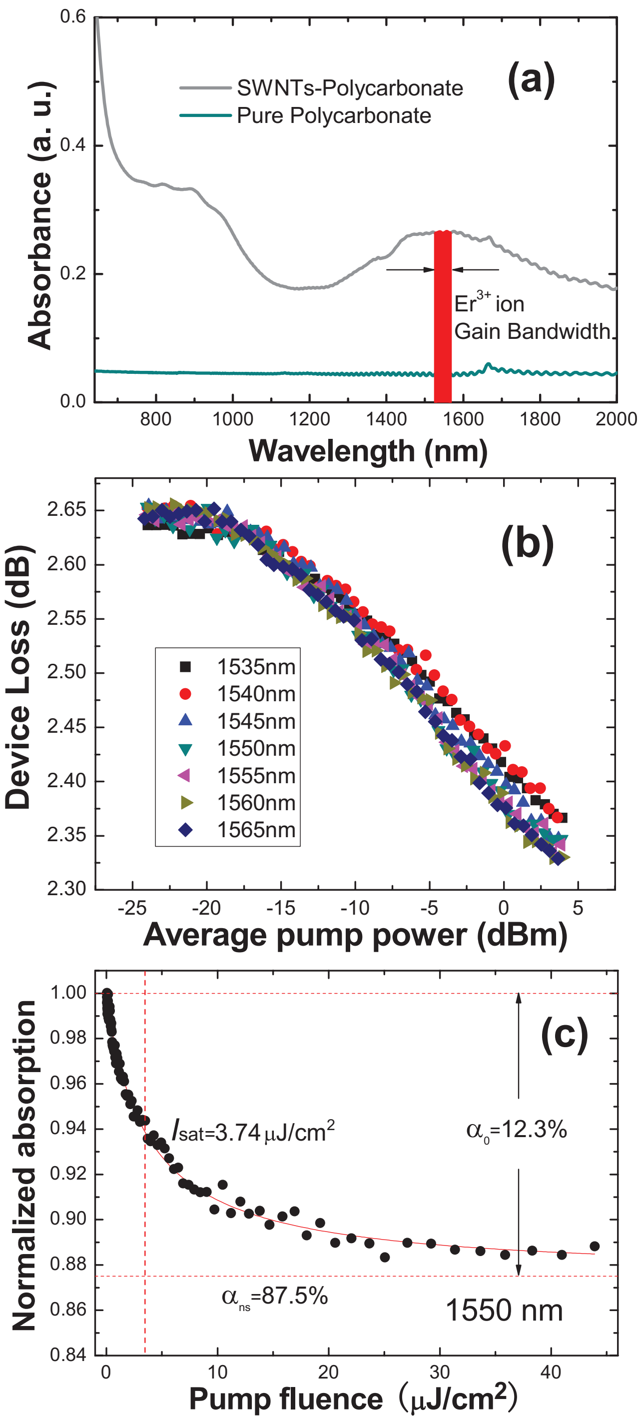

Figure 1: **Optical characterizations of the composite films**(a) Absorption spectrum of the SWNTs composite and pure polycarbonate. The red stripe shows the spectral gain region of the Er^{3+} doped fiber (b) Nonlinear absorption measurements of our SWNTs saturable absorber. The pump power ranges from 3μW to $\sim$3mW. (c) Typical normalized absorption of the SWNTs-saturable absorber as a function of pump pulse fluence. Data correspond to pump at 1550 nm (red line: fitted curve according to Eq.1)

simple two-level saturable absorber model [31]:

$$\alpha(I) = \frac{\alpha_0}{1 + I/I_{sat}} + \alpha_{ns} \tag{1}$$

where $\alpha(I)$ is the intensity-dependent absorption coefficient, α_0, α_{ns} and I_{sat} represent the saturable absorption, non-saturable absorption and saturation fluence, respectively. This gives an average saturable absorption (modulation depth) of 12.2% and an average saturation fluence of 4.28 μJ/cm^2 in the spectral range investigated. A typical nonlinear absorption curve for 1550 nm excitation is shown in Fig.1(c). The modulation depth and saturation fluence for our devices are very comparable to those of SESAMs [6], but the non-saturable loss is larger. This is tolerable for fiber lasers with a relatively big single round-trip gain coefficient[25]. Yet, it degrades the mode-locking performance, resulting in less output power or longer pulses. In our case, the main contribution to the non-saturable loss is the linear coupling loss between the two fibre ends, due to mode divergence inside the film[32]. On the other hand, the scattering losses from the SWNTs do not play a major role, since the bundle sizes in the composites are much smaller than the incident optical wavelength[33]. Thus, by further engineering the film thickness, devices with minimized non-saturable loss could be obtained.

The laser setup is schematically shown in Fig.2. A 1-meter highly-doped Er^{3+} fibre is used as the gain medium. It is pumped by a 980 nm diode laser via a wavelength-division-multiplexing (WDM) coupler. Two isolators are placed at both ends of the amplifier section to maintain unidirectional laser operation. A tunable filter (nominal operating wavelength 1535-1565 nm) with a 3-nm passband is placed after the isolator at the output of the amplification section. Light is then extracted from the cavity via a 50/50 coupler. A polarization controller is used to optimize the mode-locking conditions and the SWNT-film is placed between this and the isolator at the input of the amplification section. The total length L of the cavity is $\sim$13.3 meters. We can thus estimate the expected repetition rate to be $\sim$15 MHz, from $f_r = c/(nL)$, where f_r is the repetition rate, c the velocity of light in vacuum, and n the average refractive index of the cavity ($n \sim 1.5$).

The mode locking results are as follows. With the filter in the cavity, the threshold pump power for continuous wave (CW) lasing is $\sim$15 mW at 1550 nm. When the pump power is increased to $\sim$45 mW, stable mode-locking can be initiated by introducing a disturbance to the polarization controller. Once stable output is achieved, no further polarization controller adjustment

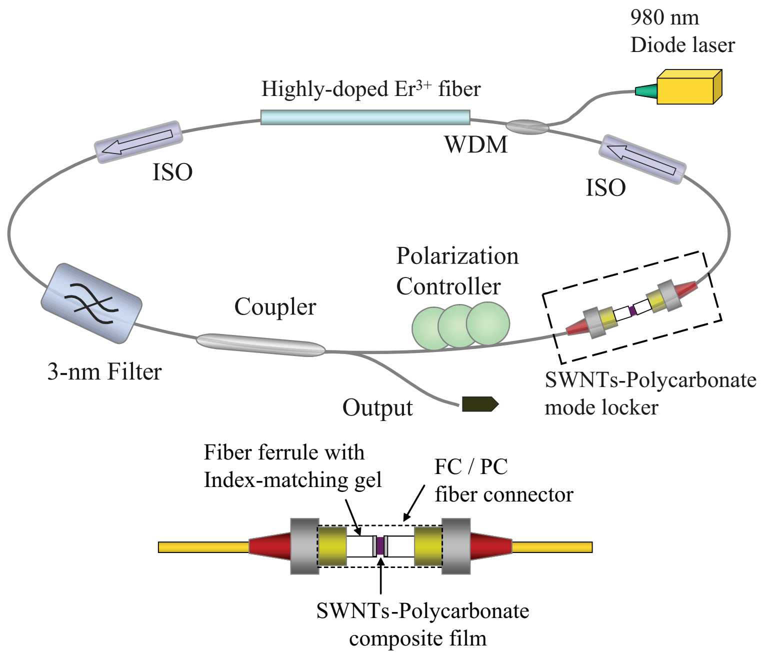

Figure 2: **Laser setup** ISO:isolator;WDM:Wavelength division multiplexer.

is needed and it is possible to decrease the pump power to ~35 mW while maintaining mode-locking. For optimal polarization controller settings, the laser can self-start with excellent repeatability. The measured repetition rate is 15.01 MHz, in agreement with the design parameters. As the pump power exceeds ~50 mW, multiple pulses per round-trip are observed. We attribute them to overdriven nonlinear effects, such as self-phase modulation, when a single pulse with relatively high peak power is circulating in the cavity[21].

Wavelength tuning is provided by the intra-cavity filter. By changing its tilt angle it is possible to tune the filter passband from 1535 to 1565nm. To deduce the maximum tuning range, we also study wavelengths outside the nominal bandwidth of the gain medium and the filter. To enhance the gain coefficient outside the typical amplifying wavelength range (1530~1560nm) of the Er^{3+} fiber, more pump power is provided to the gain fiber. We find that a tuning range of 40 nm, from 1518 nm to 1558 nm, is possible with the current setup. Figs.3a,b illustrate the output spectra and autocorrelation traces at several wavelengths within the tuning range. Due to the use of the bandpass filter, the output spectra (Fig. 3(a)) have a high signal-to-noise-

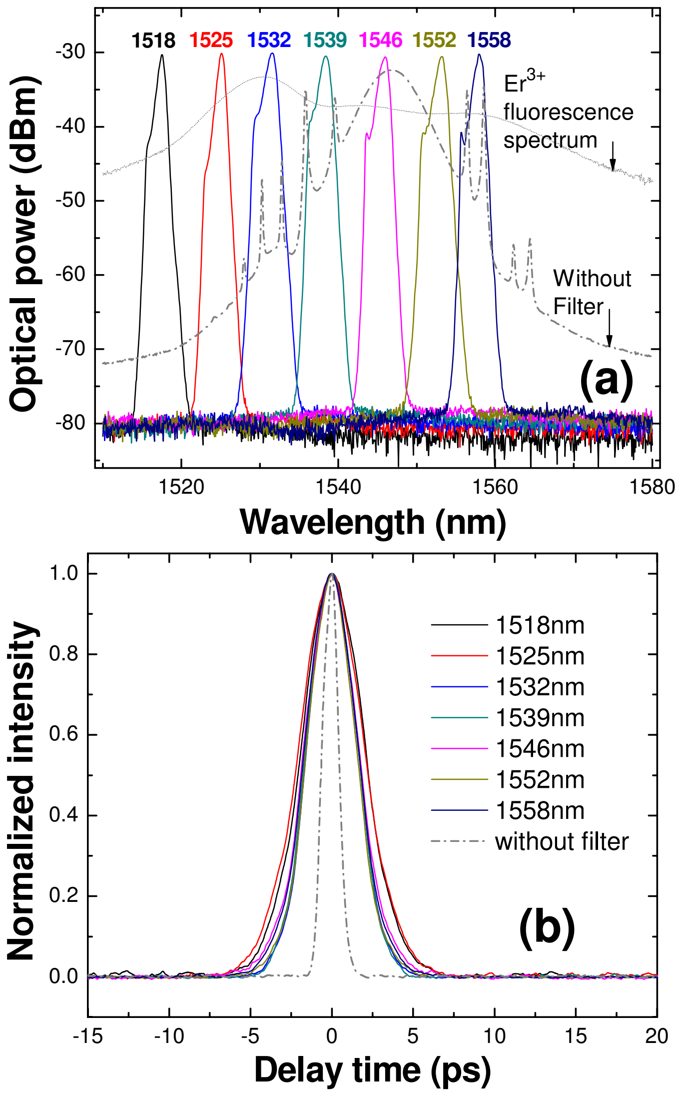

Figure 3: **Wavelength tuning**(a)Output spectra at different wavelengths. (Dashed line: optical spectrum without band-pass filter; dotted line: Er^{3+} fluorescence spectrum) (b) Autocorrelation traces of laser output at different central wavelengths. (Dashed line: laser output without band-pass filter)

ratio of ~50 dB (10^5 contrast). No sideband structure is observed due to the spectral limiting effect of the filter[34]. On the other hand, the filter also limits the mode-locking spectral width to an average value of 1.2nm, which keeps the pulse duration above 1ps. All the autocorrelation traces in Fig.3(b) do not show pedestals (low-intensity backgrounds), indicative of single pulse operation and reflection-free cavity design[5]. Our cavity uses an anomalous dispersion fibre, with an overall negative group velocity dispersion, in order to facilitate soliton-like pulse shaping through the interplay of group velocity dispersion and self-phase modulation [5]. In this case, the output pulses are expected to have a sech^2 lineshape[5]. Indeed the data in Fig. 3(b) are well fitted by a sech^2, giving an average pulse duration of 2.39 ps. The average output power is 0.36 mW, with a fluctuation of 15% across the entire range. Since we use a 50/50 coupler, the incident power onto the mode-locker is the same as the output power. Form the average output power and repetition rate, the energy per pulse is 24 pJ. Considering the mode diameter of 10μm, we get a fluence of ~30 μJ/cm^2, i.e. 7-10 times the saturation one, implying a full saturation of the SWNTs mode-locker. By comparison, the laser output when no filter is present (at a pump level of 35 mW) is also shown in Figs.3a,b as a dashed line. In this case, the laser mode locks at 1545 nm (corresponding to the effective gain maximum of the cavity) with typical sidebands arising from periodic cavity perturbations[5]. The pulse width is 706fs, shortened due to the elimination of intra-cavity spectral limiting effects.

The current laser does not mode-lock at wavelengths beyond 1558 nm, while it does at shorter wavelengths outside the filter nominal operating spectral range (such as 1518 nm and 1525 nm). The fluorescence spectrum of the Er^{3+} fibre indicates that there is still bandwidth on the longer wavelength side. To verify this, we use the amplified spontaneous emission (ASE) from an Er^{3+} fiber amplifier as a wideband optical source to measure the filter pass-band characteristics. We find, as expected, that the longer wavelength pass-band exhibits much worse ripple effects than the shorter wavelength one,resulting in strong narrow band limiting effect. Combined with the data in Fig.1(b), we conclude that the nonlinear absorption of our mode-locker is not the limiting factor. Indeed, in principle our SWNT film could enable a much wider tuning range, e.g. the entire S (1460-1530nm),C (1530-1565nm) and L (1565-1624nm) bands of telecommunications [35], if gain fibers and tunable filters with sufficient bandwidth would be available.

Fig.4 summarizes some of the laser output trends and characteristics within the tuning range. An average pulse duration of 2.39 ps is obtained

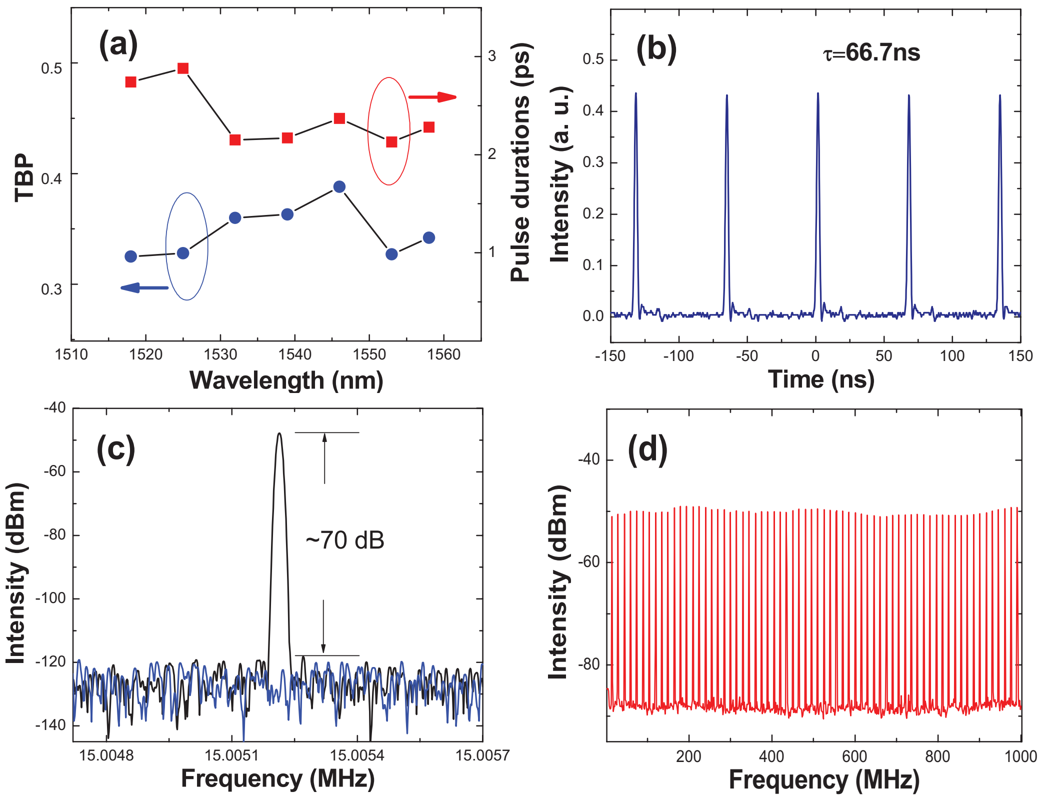

Figure 4: **Mode-locking characteristics** (a)Output pulse duration and TBP at different central wavelengths. (b)Oscilloscope trace of the typical laser output. (c) Fundamental of a typical RF spectrum of the laser output after optical-to-electrical conversion. The blue trace depicts the background when the laser is switched off. (d) Wideband RF spectrum up to 1 GHz.

for different wavelengths (Fig. 4 (a)). Fig 4a also plots the time bandwidth product (TBP), i.e. the product of the pulse temporal and spectral widths. Different applications demand the shortest pulse duration at a given spectral width. Fig 4(a) shows that the TBP ranges from 0.32 to 0.36, reasonably close to 0.315, which corresponds to the shortest pulse duration for a given spectral width, for transform-limited sech^2 pulses[5]. Fig.4(b) is the measured output pulse train, with the fundamental repetition rate of 15 MHz (period τ=66.7 ns). Fig. 4(c) shows the radio frequency (RF) power spectrum of the laser output after Optical-to-Electrical conversion using a fast photodiode. A ~70 dB peak-to-background ratio (10^7 contrast) is observed for the fundamental peak, which indicates excellent stability of the mode-locking regime. In Fig.4(d) the wideband RF spectrum up to 1 GHz is shown. The non-existence of spectral modulation indicates that the tunable laser is operating in the pure CW mode-locking regime, where the output pulse train is not subject to low-repetition-rate modulation arising from relaxation oscillations[6]

In conclusion, our work exploits SWNTs with a range of diameters and chiralities, harnessing the resulting wideband absorption to produce the first SWNTs based wideband tunable fibre laser. This turns one of the major disadvantages of SWNTs, into the key element for success of this technology. It represents a significant step forward in applying these nanomaterials as cost-effective and viable alternatives to current semiconductor quantum well based absorbers. This paves the way to the use of SWNTs-composites in ultrafast pulse generation and as a functional building block for future nano-scale photonic integrated circuits.

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1 METHODs

1.1 SWNTs composite

The SWNTs-Polycarbonate composites is prepared as follows. SWNT powders are dispersed in dichlorobenzene (DCB) in presence of poly (3-hexylthiophene-2, 5-diyl) (P3HT) by ultrasonication (Diagenode SA, Belgium). The solution is then filtered through a 1 μm retention filter (Whatman) and centrifuged (Beckman Coultrer Optima MaxE Ultracentrifuge, MLA130 fixed angle rotor) to remove bundles and impurities. Pellets of polycarbonate are dissolved in the solution by ultrasonication. The final mixture is dried at room temperature to form a film with a thickness typically of 40 microns or more. No SWNTs bundles under the scrutiny of an optical microscope can be detected, thus ensuring minimized scattering losses from large bundles.

1.2 Power-dependent absorption measurements

Power-dependent absorption measurements are carried out as follows. The SWNTs mode-locker assembly is coupled to a 650 fs optical pulse source, tunable from 1535 nm to 1565 nm. This is achieved by filtering a commercial femtosecond fibre laser (200 fs, 76.9 MHz, TOPTICA) using a 3-nm band-pass filter. The laser beam is amplified by an Erbium Doped Fibre Amplifier (EDFA) and a 10% tap is used to monitor the input power to the mode-locker assembly, containing our composite, and two appropriately calibrated power-heads are programmed to read the input/output power simultaneously.

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