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74-fs nanotube-mode-locked fiber laser

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We report an erbium-doped, nanotube mode-locked fiber oscillator generating 74 fs pulses with 63 nm spectral width. This all-fiber-based laser is a simple, low-cost source for time-resolved optical spectroscopy, as well as for many applications where high resolution driven by short pulse durations is required. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4757293>]

Optical pulses with durations ranging from nanoseconds to a few femtoseconds are necessary for basic research and applications.^{1–3} For a given wavelength, the ultimate duration is set by the oscillation period λ/c , with λ the wavelength and c the speed of light.^{4,5} Shorter pulses would not have the oscillatory character set by λ , thus would be unable to propagate.⁶ At $1.5\ \mu\text{m}$, $\lambda/c \sim 5$ fs, while at 800 nm this reduces to ~ 2.7 fs. Only by moving to shorter wave cycles, i.e., deep ultraviolet, with energies of 10 eV to 1 keV or beyond, one can break the femtosecond barrier (i.e., for 1 keV, $\lambda/c \sim 4.13$ attoseconds). For a review of attosecond pulse generation see Ref. 7.

In the visible to near infrared (NIR), very short pulses can be obtained with passive mode-locking.^{2–4} Over the years, a number of cavity designs have been investigated^{2,3,5} and an ever increasing research effort is being devoted to techniques that are able to push the temporal resolution towards the light oscillation period.^{2,3,5} Fiber lasers are attractive due to their simple and compact design, efficient heat dissipation, and alignment-free operation.^{3,8} Ultrafast pulses can be achieved in fibre lasers by passive mode-locking, with the aid of an intensity-dependent element called saturable absorber (SA).^{2,8} Key parameters for a SA are its dynamic response (the shorter the pulse, the faster the loss modulation, provided the SA has a sufficiently short recovery time^{2,8}) and its wavelength range (the broader the bandwidth, the shorter the supported pulses, e.g., $\Delta\nu\Delta\tau \approx 0.44$ for a Gaussian pulse, where $\Delta\nu$ and $\Delta\tau$ are the full width at half maximum (FWHM) spectral and temporal widths^{2,8}). Nanotubes and graphene are promising SAs, at the center of a growing research effort due to their fast recovery times,^{9–20} broad operation range,^{28–30} low saturation intensity,^{28,30} low cost,²³ and easy fabrication.^{21–37} They were used to mode-lock fiber,^{28–43} waveguide,^{44,45} solid-state,^{46–48} and semiconductor lasers.⁴⁹

Pulses with durations ranging from 100 fs to less than 10 fs are necessary for time-resolved ultrafast dynamics, e.g., to monitor photophysical and photochemical relaxation processes.^{16,50,51} Pulses shorter than 100 fs covering the visible to mid-infrared (MIR) range can be created using optical parametric amplifiers (OPA) pumped by Ti:sapphire lasers.^{50–52} An OPA efficiently transfers energy from a narrow band pump pulse to a broadband signal pulse, with a significantly shorter duration than the pump pulse.⁵² However, OPAs are complex and expensive compared to fiber-lasers.⁵²

In fiber lasers, a typical approach to ultrafast pulse generation is soliton mode-locking.⁵³ In this regime, the intra-

cavity chromatic dispersion is balanced by nonlinear optical effects triggered by the high-intensity of the pulse itself.^{54,55}

The effects of nonlinearity and dispersion can cancel each other, apart from a constant nonlinear phase delay per unit propagation distance⁵⁵ (i.e., a $\pi/4$ nonlinear phase delay is accumulated over a propagation distance called soliton period $Z_0 \approx \frac{\tau^2}{2\beta_2}$, where τ [s] is the pulse width and β_2 [s^2/m] the group velocity dispersion (GVD) parameter).⁵⁶ The shortest solitons that can be stably supported have typically $Z_0 > L/2$, where L is the cavity length.⁵⁵ For 1 ps pulses, $Z_0 \sim 23$ m, assuming a typical GVD = $-26\ \text{fs}^2/\text{mm}$ for silica at $1.5\ \mu\text{m}$.⁵⁷ For quicker pulses, Z_0 becomes too short, e.g., for 100 fs, $Z_0 \sim 23$ cm, corresponding to $L < 50$ cm. For such short L , it is not easy to compensate the dispersion and nonlinearity. For example, the amount of dispersion required involves long fibers, which in some cases are replaced by other optical components (e.g., grating pairs) for large dispersions.⁵⁷ A strategy to overcome the limitations associated with solitons is to alternate segments of large normal and anomalous GVD fibers, so that the pulse maintains the minimum duration only over small portions of the cavity, being thus less susceptible to nonlinear optical effects.^{54,55} This leads to periodic broadening and compression of the intracavity pulses.^{54,55} This configuration is known as the stretched-pulse design.^{54,55} Compared to soliton mode-locking, the average τ can increase by an order of magnitude or more, which significantly reduces the intracavity average peak power P , since⁵⁶ $P \propto \frac{E}{\tau}$, where E [J] is the pulse energy. This in turn reduces the associated nonlinear optical effects.^{54,55} However, in fiber lasers, it is difficult to achieve the minimum pulse duration, as set by the optical cycle. On one hand, for few-optical-cycles pulse durations, fiber dispersion related effects become significant,⁵⁸ and it is necessary to provide dispersion compensation not only for the average group delay dispersion (GDD) (second-order dispersion) but also for the third-order dispersion (TOD), and possibly even higher orders.⁵⁸ On the other hand, shorter durations (i.e., approaching the oscillation period) are affected by the limited bandwidth of gain fibers.⁵⁹ For example, at $1.5\ \mu\text{m}$, the shortest duration to date in a mode-locked fiber laser is 37.4 fs,⁶⁰ longer than the limit set by the oscillation period ~ 5 fs, which would require a spectral width > 600 nm, much wider than an ~ 135 nm in Ref. 60.

To make an ultrafast laser, besides its cavity design, the SA recovery time is another parameter to consider. Isolated single wall carbon nanotubes (SWNTs) have recovery times for the first optical transition (E_{11}) ~ 10 –100 ps,^{17–19} while

shorter times (few hundreds fs) are reported for bundled tubes.^{9,11,20} When embedded in polymer composites, the recovery time is still of the order of hundreds of fs.^{10,12} In the case of graphene, the relaxation time due to carrier-carrier scattering is $\sim 10\text{--}50$ fs.^{13–16} It is possible to achieve pulses shorter than the SA recovery time,^{2,8,61,62} with the final pulse-width mainly defined by the laser cavity design. Typical examples are lasers exploiting soliton effects, such as that in Ref. 62, where the SA is only needed to stabilize the pulse. Even without soliton effects, the SA recovery time can be as long as 30 times the final pulse duration (see, e.g., Ref. 61). In this case, the main role of the SA is to attenuate the leading wing of the pulse, which shifts backwards in each cavity round-trip.⁶¹ This limits the time over which the noise behind the pulse can be amplified⁶¹ and, as a result, the pulse remains short.⁶¹ In the case of nanotube and graphene mode-locked fibre lasers, pulses as short as 113 fs^{32,33} and 174 fs³⁶ were reported. Considering the few hundreds fs^{9–12,20} recovery time of nanotubes-composites, and the possibility of generating pulses with durations shorter than the SA recovery time,^{2,8,61,62} there should be, in principle, no fundamental obstacle in breaking the 100 fs barrier.

Here, we nanotube-mode-lock a fiber oscillator, generating 74 fs pulses. This is achieved by using an all-fiber stretched-pulse laser design. We get a spectral width of 63 nm, much larger than reported thus far for nanotubes and graphene based fiber oscillators.^{32,33,36}

In order to get shorter pulses than those reported in Refs. 32 and 33, we combine a highly-doped erbium doped fiber (EDF) able to provide broadband gain, with a shorter cavity length than Ref. 32 and a short Z_0 . Fig. 1 is the schematic dispersion distribution along our laser cavity. This has a ring configuration, as indicated by the dotted line in Fig. 1(b), and consists of segments of alternating large normal and anomalous dispersion fiber, Fig. 1(a). The net dispersion is kept slightly anomalous, to achieve a stretched-pulse design.^{54,55} We use a 1.3 m EDF with $\beta_2 = 48$ ps²/km as gain medium. The rest of the cavity consists of 1.78 m single mode fiber (SMF) Flexcore 1060 with $\beta_2 = -7$ ps²/km and 3.22 m SMF-28 with $\beta_2 = -22$ ps²/km. We estimate the GVD by inserting the fibres into a SWNT-mode-locked soliton-like fiber

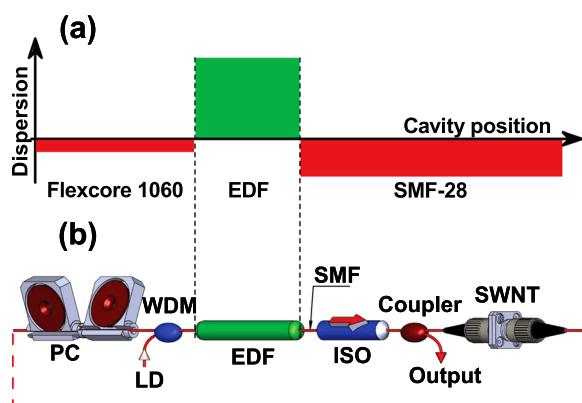


FIG. 1. (a) Scheme of dispersion distribution in our laser. Normal dispersion is provided by an EDF. Anomalous dispersion provided by a combination of SMFs: Flexcore 1060 and SMF-28. (b) Laser setup: LD-laser diode, WDM-wavelength division multiplexer, EDF-erbium-doped fiber, SMF-single mode fiber, ISO-isolator, PC-polarization controller.

laser,⁴³ and measuring the shift $\Delta\lambda$ between sidebands and central wavelength of the soliton pulse spectrum.⁶³ The measured total intracavity GDD is ~ -0.003 ps², comparable to that typically reported for stretched-pulse lasers.^{32,33,54,55} The EDF is pumped by a 980 nm laser diode (LD) through a fused wavelength division multiplexer (WDM) made of Flexcor 1060 fiber, Fig. 1(b). Unidirectional lasing in the ring is achieved with an optical isolator (ISO). The laser output is directed through the 20% port of a coupler. For mode-locking optimization, a polarization controller (PC) is placed after the SWNT-SA. The total cavity length is ~ 6.3 m, shorter than ~ 11.17 m of Ref. 32.

The SWNT-SA is prepared as follows. ~ 0.03 wt.% laser ablation SWNTs with ~ 1.3 nm mean diameter⁶⁴ are ultrasonicated for an hour with 0.7 wt.% sodium-carboxymethylcellulose (Na-CMC) polymer²³ using a tip sonicator (Branson 450A, 20 kHz) with ~ 50 W power. These SWNTs have their lowest optical transition at ~ 0.8 eV,⁶⁵ resulting in an absorption peak ~ 1550 nm.⁶⁵ Na-CMC eliminates the requirement of surfactants to disperse SWNTs,⁶⁶ thus cutting one process step. Also, the optical absorption of the surfactant would contribute to non-saturable losses, with a consequent increase of the SA insertion losses. The dispersion is then centrifuged in a swing bucket rotor at 30 krpm using a Beckman Coulter Optima Max E and then spin-cast in a Petri dish. After water evaporation, we get a ~ 30 μm thick composite. The SWNT-based SA is then realized by cutting a ~ 2 mm² piece and placing it between two fiber connectors.

Power-dependent absorption is measured with an optical parametric oscillator (Coherent, Chameleon) delivering ~ 200 fs pulses with 80 MHz repetition rate at 1550 nm. The optical transmittance is determined by monitoring the input and output power on the SA. The nonlinear transmittance increases from $\sim 33\%$ to $\sim 50\%$ at saturation, with $I_{\text{peak}} \sim 203$ MW/cm², for 2.9 mW pump power. This gives $\sim 17\%$ modulation depth (Fig. 2).

Continuous wave (CW) operation starts at ~ 17 mW pump power. Self-starting mode-locking is observed at ~ 23 mW. This can be further optimized by adjusting the intracavity polarization. The repetition rate is 33 MHz, as determined by the cavity length. The typical output power is 1.2 mW for ~ 27 mW pump, with a pulse energy of 36 pJ. A typical spectrum is shown in Fig. 3(a). Resonant sidebands, signatures of soliton-like operation,⁶³ are reduced, as

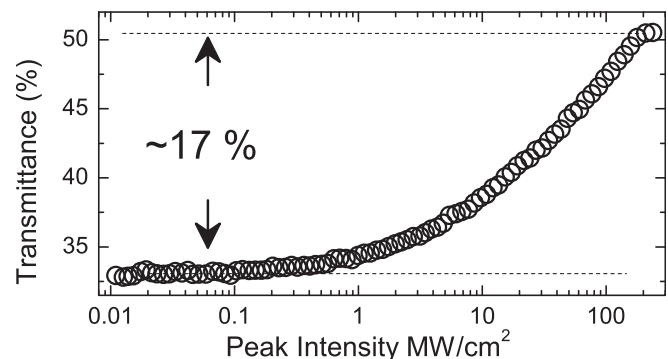


FIG. 2. Transmittance as a function of pump intensity.

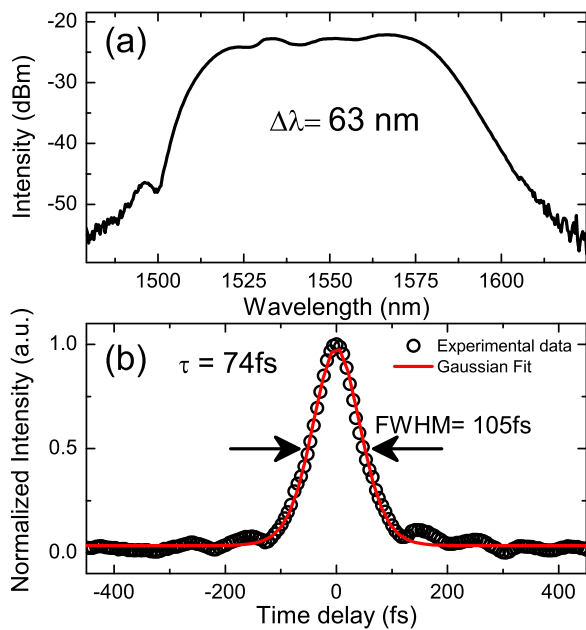


FIG. 3. (a) Optical spectrum with bandwidth $\Delta\lambda = 63$ nm. (b) Autocorrelation trace with Gaussian Fit.

a result of the alternating dispersion, which limits phase-matched coupling.^{54,55,67}

The FWHM bandwidth is 63 nm, wider than usually reported for the Er^{3+} gain bandwidth,⁵⁹ and almost twice that previously achieved in SWNT-based fiber lasers.^{32,33} For our EDF segment, the gain bandwidth at 3 dB is ~ 30 nm, for a pump power ~ 30 mW. The autocorrelation trace is shown in Fig. 3. The FWHM is 105 fs. Assuming a Gaussian profile, as expected for stretched-pulse lasers,⁶⁸ deconvolution gives ~ 76 fs duration, shorter than earlier SWNT and graphene-mode-locked fiber lasers.^{32,33,36} The radio frequency (rf) spectrum in Fig. 4 gives a signal-to-noise ratio >70 dB (10^7 contrast), indicating low-amplitude fluctuations, thus stable mode-locking.⁶⁹

With a similar cavity design, but using nonlinear a polarization evolution mode-locker instead of nanotubes, Ref. 67 achieved 77 fs, consistent with our work. Note that the shortest pulse duration for EDF lasers, 37.4 fs,⁶⁰ was generated in a ~ 90 cm cavity, much shorter than ours, mode-locked by nonlinear polarization-evolution. Our 63 nm spectral width suggests that even shorter pulses may be generated. For this bandwidth, a transform-limited pulse would produce sub-60 fs pulses ($\Delta\tau \approx \frac{0.44 \lambda^2}{c \Delta\lambda} = 56$ fs, at $\lambda = 1.5 \mu\text{m}$). However, our measured time-bandwidth product (TBP) is ~ 0.57 , slightly higher than 0.44, expected in the case of transform-limited Gaussian pulses.⁵⁴ This may be due to uncompensated high-order dispersion,⁵⁸ which could distort the intracavity pulse, limiting the minimum pulse width.^{67,70} In particular, TOD is the major effect usually reported for cavities with small GVD.⁷⁰ The TOD is typically positive in normal single mode fibers at $1.5 \mu\text{m}$ (e.g., $\beta_3 \sim 0.12 \text{ps}^3/\text{km}$ for SMF-28,⁷¹ $\beta_3 \sim 0.1 \text{ps}^3/\text{km}$ for EDF⁷²), unlike second-order dispersion (i.e., GVD), which changes sign for the SMF-28 and EDF in our cavity. Therefore, the TOD is not compensated and is typically accumulated with the cavity length. Thus, even shorter pulses may be obtained by cutting the fiber length, since this may reduce the intracavity TOD.

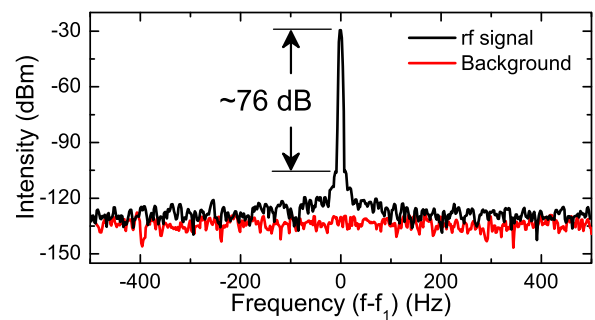


FIG. 4. Output rf spectrum, measured around the fundamental repetition rate $f_1 = 33$ MHz with 2 Hz resolution.

A shorter cavity is also helpful as it enables less nonlinear effects, since these depend on the interaction length.⁵⁸ Another approach to TOD compensation would be to use negative TOD components, such as specially designed fibers (e.g., photonic crystal fiber⁷³) and prism pair.⁷⁰ However, even after careful dispersion compensation, achieving pulses below 20 fs may still be limited by the EDF gain bandwidth.^{60,74}

In conclusion, we demonstrated SWNTs as mode-lockers for a stretched-pulse Er doped fiber laser generating pulses shorter than 100 fs. The output spectral width is 63 nm. This result has great potential in order to realize low-cost light sources needed for applications requiring ultra-short pulses, such as micro-machining, spectroscopy, and biomedical diagnostics.

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