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# Ultrafast Raman laser mode-locked by nanotubes

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We demonstrate passive mode-locking of a Raman fiber laser using a nanotube-based saturable absorber coupled to a net normal dispersion cavity. This generates highly chirped 500 ps pulses. These are then compressed down to 2 ps, with 1.4 kW peak power, making it a simple wavelength-versatile source for various applications.

Mode-locked lasers have been widely deployed in time-resolved studies for nearly 50 years [1]. While many mode-locking approaches exist, allowing an extensive range of pulse durations and energies to be achieved, passive techniques are particularly attractive because of their inherent simplicity and lower production costs [2]. Over the past two decades, passively mode-locked fiber lasers have been extensively refined and developed, becoming a commercial success and an indispensable tool in many fields [1,2]. These systems have almost exclusively been based on rare-earth-doped gain media [1,2], taking advantage of the rapid technological growth of fiber-based amplifiers, driven largely by the telecommunications industry. However, such lasers have limited bandwidth, as rare-earth-doped gain fibers have typically narrow gain bandwidth ( $\sim$ few tens nm) [1].

In telecommunications, Raman-based amplification allows operation beyond the spectral limits of rare-earth devices [3]. Consequently, similar techniques can be applied to ultrafast fiber lasers. The most attractive feature of Raman-based amplification in silica fiber is that gain is available at any wavelength across the transparency window of the medium (300–2300 nm), given a suitable pump source [3]. With advances in high-power fiber laser pump technology and in cascaded Raman fiber lasers, efficient pump systems are now available throughout this entire band.

There have been a number of reports utilizing Raman gain in ultrafast sources [4–8]. However, to date, none of these systems has reached a level of performance comparable with state-of-the-art rare-earth-based lasers. In Ref. [4], dissipative four-wave mixing was used for mode-locking, generating a pulsed laser with a very high repetition rate. While this is useful for some applications, the high repetition rate limits the delivered peak power. Nonlinear loop mirrors [5,6] and nonlinear polarization evolution [8] have also been used to provide saturable absorption, but such systems suffer from instabilities due to fluctuations in ambient temperature, and often exhibit poor self-starting performance [5,6,8].

Recently, a Raman mode-locked laser using a semiconductor saturable absorber mirror (SESAM) was reported

[7]. While the use of a SESAM improves self-starting and robustness against environmental perturbations, there is limited spectral operation from a single device. In addition, the fabrication cost of SESAMs at nonstandard wavelengths is high. Availability of a broadband saturable absorber (SA) to achieve mode-locking at any desired wavelength across the transmission window of silica is an essential prerequisite to fully exploit the flexibility of Raman amplification in ultrafast sources across the visible and near-IR. Recent interest in the application of nano materials, in particular carbon nanotubes (CNTs) [9–12] and graphene [10,13–15], as SAs in mode-locked lasers have moved the field a step closer to a fully universal device [9–15]. For example, a single CNT-based SA was used to mode-lock fiber lasers, offering wide wavelength-tunable pulses [11] and broad spectral coverage from 1 to 2  $\mu$ m [9].

Here, we report a passively mode-locked laser combining both Raman gain and a CNT-based SA in a normally dispersive cavity, showing the potential of this flexible approach. Mode-locked lasers based on rare-earth media and operating with a net normal dispersion map, generating so-called dissipative solitons [12,16,17], were suggested as a means of overcoming the limits on pulse energy imposed by the soliton regime. Such systems were shown to support highly linearly chirped pulses [12,16,17], with durations up to several nanoseconds [12]. Thus they can be amplified and compressed without temporal and spectral degradation due to nonlinearities. Here we show that this approach is applicable to Raman-based systems, with pre-chirped pulses, generated in a normally dispersive seed oscillator, amplified and compressed to 2 ps, with 1.4 kW peak power.

The all-fiber geometry is shown in Fig. 1. The cavity consists of 100 m single-mode highly nonlinear fiber (OFS Raman Fiber), with an enhanced germanium oxide (GeO) concentration for an increased Raman gain coefficient ( $2.5 \text{ W}^{-1} \text{ km}^{-1}$ ), core pumped through a wavelength-division multiplexer (WDM) by a CW 15 W Er-doped fiber amplified spontaneous emission source at 1555 nm. No synchronous pumping is necessary in this system.

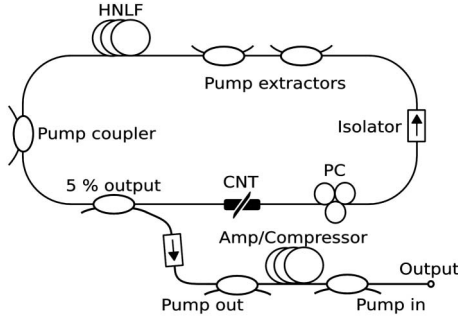


Fig. 1. Experimental setup. HNLF, highly nonlinear fiber; PC, polarization controller; CNT, carbon-nanotube-based saturable absorber.

The CNT-polymer SA device is prepared by solution processing [10]. We use carbon nanotubes grown by catalytic chemical vapor deposition [18]. After purification by air oxidation at 450 °C, followed by HCl washing, the remaining carbon-encapsulated catalytic nanoparticles are removed [19]. Analysis of the purified samples by transmission electron microscopy (TEM) reveals the presence of ~90% double-wall carbon nanotubes (DWNTs), ~8% single-wall carbon nanotubes, and ~2% triple-wall carbon nanotubes [19]. The diameter distribution for DWNTs is ~0.8–1.2 nm for the inner and ~1.6–1.9 nm for the outer diameters, as determined by Raman spectroscopy and TEM. This wide diameter distribution can potentially enable broadband operation, essential for the large wavelength coverage offered by Raman amplification. The purified nanotubes are then dispersed using a tip sonicator (Branson 450 A, 20 kHz) in water with sodium dodecylbenzene sulfonate surfactant, and mixed with aqueous polyvinyl alcohol (PVA) solution to obtain a homogeneous and stable dispersion free of aggregates. Slow evaporation of water from this mixture produces a CNT-PVA composite ~50  $\mu\text{m}$  thick. Optical microscopy reveals no CNT aggregation or defect in the composite, thus avoiding scattering losses.

Self-starting mode-locked operation is initiated and maintained by integrating our CNT-based SA into the cavity between a pair of fiber connectors. A polarization insensitive in-line optical isolator and fiber-based polarization controller stabilize mode-locking. Light is extracted from the unidirectional cavity through a 5% fused fiber coupler. To prevent high levels of undepleted pump power damaging the passive cavity components, a second WDM is used to couple out residual pump light.

Stable quasi-CW mode-locking is supported over a range of pump powers above the lasing threshold at 9.5 W. Increasing pump power leads to spectrum broadening and breakup of the single-pulse operation. The spectrum, the temporal pulse intensity profile, and the radio frequency (RF) trace of the fundamental harmonic of the cavity are reported in Fig. 2. The pulses are 500 ps [Fig. 2(a)], and the average output power for stable mode-locking is ~0.08 mW, with a single pulse per round trip. The Raman laser operates at the first Stokes order of 1666 nm from a pump at 1555 nm [Fig. 2(b)]. The spectrum has a square shape, with a -6 dB bandwidth of ~1.6 nm. The square-shaped spectrum is a recognizable feature of lasers operating in the dissipative soliton regime [12]. Such systems generate pulses carrying a large

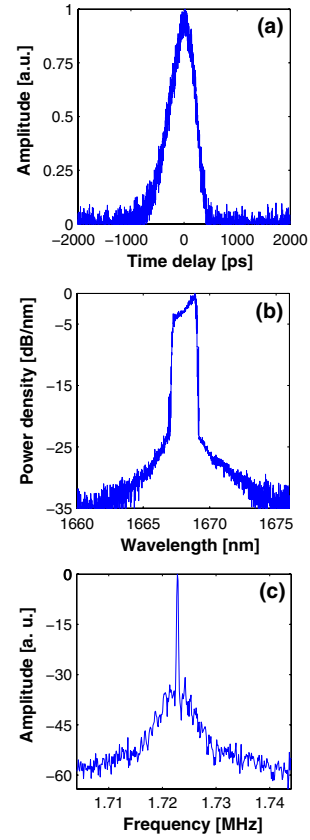


Fig. 2. (Color online) (a) Temporal pulse intensity profile, (b) spectrum, and (c) RF trace at the output of the oscillator.

and predominantly linear chirp [12], and are suitable for compression [12]. The RF trace [Fig. 2(c)] shows a significant pedestal, containing 0.57% of the total energy, indicating that the cavity is prone to long-term temporal instabilities and fluctuations of the pulse-to-pulse energy. This relatively high noise contribution, compared to state-of-the-art rare-earth-based systems, is expected due to the high level of pump power. On the other hand, the narrow linewidth of the peak at 1.72 MHz, corresponding to the round-trip time of the cavity, indicates low pulse timing jitter.

Significant interest in normal dispersion mode-locked lasers is stimulated by the possibility of overcoming the pulse energy limits imposed by soliton propagation, with a linearly chirped-pulse structure, commonly known as a dissipative soliton, now routinely generated in all-fiber geometries [12,16,17]. In particular, giant-chirp oscillators (GCOs) have been proposed as a means of pre-chirping the pulse frequencies directly in the oscillator to simplify the chirped-pulse amplification design [12,16]. Further information regarding all-normal dispersion operation in mode-locked lasers can be found in Refs. [20–23]. In our setup, after the oscillator, a second, 10 km Ge-doped fiber with a zero dispersion wavelength at 1320 nm is used as a combined amplifier and compressor (Fig. 1). The residual pump out of the seed oscillator is used to counterpump the Ge-doped fiber for a compact GCO-type master oscillator power fiber amplifier solution.

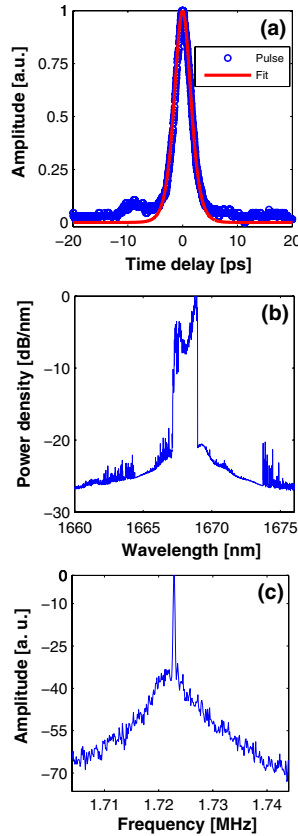


Fig. 3. (Color online) (a) Autocorrelation pulse trace, (b) spectrum, and (c) RF trace after compression.

The autocorrelation trace of the compressed and amplified pulse, its spectrum, and the RF trace of the fundamental cavity harmonic after compression are shown in Fig. 3. The pulses are successfully compressed nearly 250 times from 500 to 2 ps, and no significant pedestal is observed on the autocorrelation traces. The average output power after amplification and compression is  $\sim 5$  mW, corresponding to the 18 dB gain provided by the amplifier, which requires 6 W supplied by the undepleted power from the oscillator stage. Importantly, the spectral shape is preserved [see Fig. 3(b)], indicating linear compression, although noticeable degradation of the optical signal-to-noise ratio due to the amplified spontaneous signal in the amplifier fiber is apparent. The RF trace of the amplified and compressed signal is naturally centered at 1.72 MHz and shows a 6% increase of the noise pedestal when compared to the input signal. The pulse peak power after compression is  $\sim 1.4$  kW. Scalability of the peak power to higher levels should be possible by decoupling the amplifier from the compressor.

In conclusion, we reported an all-fiber passively mode-locked Raman laser utilizing a broadband CNT-based SA. The 500 ps pulses provided by the oscillator were amplified and simultaneously compressed to 2 ps with 1.4 kW peak power. The oscillator was implemented in a cavity with dominant normal dispersion, allowing generation of high energy pulses, up to  $\sim 3$  nJ, after amplification. This confirms the flexible approach of combining Raman gain and a CNT-SA, both broadband devices, for the development of ultrashort sources across the complete transparency window of silica fibers.

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