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Ultrafast lasers mode-locked by nanotubes and graphene

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ABSTRACT

Ultrafast lasers play an increasingly important role in many applications. Nanotubes and graphene have emerged as promising novel saturable absorbers for passive mode-locking. Here, we review recent progress on the exploitation of these two carbon nanomaterials in ultrafast photonics.

1. Introduction

Ultrafast lasers are used in a variety of applications, ranging from optical communications [1] to medical diagnostics [2] and industrial materials processing [3]. Development of new gain media (e.g. Ti:sapphire [1–4]), and mode-locking technologies (e.g. Kerr-lens mode-locking [1–4] and Semiconductor Saturable Absorber Mirrors (SESAMs) [4–6]) have changed the outlook of ultrafast lasers over the past two decades. These advances, in particular the realization of new mode-locking technologies, have pushed the applications of ultrafast pulses to a realm broader than ever before. Nevertheless, current mode-locking technologies still suffer from drawbacks, e.g., Kerr-lens mode-locked lasers usually require external perturbations in order to start [1,3,5] and are extremely sensitive to misalignment [1,3]. SESAMs are complex quantum well devices, typically fabricated by molecular beam epitaxy on distributed Bragg reflectors [4–8]. Post-growth processing [4–7] (e.g. ion implantation [4–7]) is normally required to reduce their response time [4–7]. These limitations motivate research on new materials, novel designs and technologies.

Conventional lasers, including ion-doped solid-state, fiber, semiconductor, liquid and gas based, intrinsically have a limited wavelength range of operation [9], due to the limited transitions of the gain media [9]. For example, Ti:sappire lasers only work between 0.65 and 1.1 μm [9]. Nonlinear effects (e.g. optical parametric generation [10,11] and Raman scattering [11,12]) have been widely used for light amplification, in particular for ultrafast pulse amplification, due to broad-band gain, spectral range and gain bandwidth they enable [11–13]. For example, Raman amplification, whereby a signal at the Raman Stokes-shifted wavelength experiences amplification by stimulated Raman scattering, is often employed to reach beyond the spectral limits of rare-earth fibers [9,14]. Raman based amplification can potentially allow broadband gain at any wavelength across the transparency window of silica ($\sim$300–2300 nm) [9,14]. With advances in high-power fiber-laser pump technology and in cascaded Raman fiber lasers, high efficient pump systems are now available over this entire band, providing Raman gain coefficients exceeding $\sim$70 dB (10^7) [11].

The search for alternative saturable absorber (SA) materials, essential for passive mode-locking [3–5,7], has intensified, as traditional SAs (e.g. organic dyes [15], color filter glasses [16], ion-doped crystals [17]) have severe limitations in terms of stability and performance (e.g. slow response time [5], narrow operation wavelength [7,18], expensive fabrication and integration methods [5], low damage threshold [5]). Single wall carbon nanotubes (SWNTs) have emerged as new SA material with superior performance, such as sub-picosecond recovery time [18–25], mechanical [18,26] and environmental robustness [27,28]. SWNT mode-locked ultrafast lasers have been demonstrated for various applications (e.g. industrial measurements [29], material processing [28], optical sampling [30], data-pattern recovery [30], optical frequency metrology [31,32], and optical coherence tomography [33]). Graphene has also come to the fore as a new SA with ultrafast recovery time [34–39] and ultra-broadband operation (Fig. 1(a,b)) [18,38–41].

2. SWNT mode-locked lasers

SWNT based SAs (SWNT-SAs) have been successfully implemented in a variety of laser: solid-state [42–48], fiber [18,27,28,49–72], semiconductor [73] and waveguide [74,75]. Various strategies have been implemented to fabricate SWNT-SAs, Table 1. These include spray coating [49,50,73,76–82], direct growth/transfer [57,59, 83–86], optically driven deposition [27,28,51–53,61,62,68–70, 74,87–90], polymer composite (Fig. 1(a)) [27,28,51–53,61,62,68–70, 74,87–90], polymer fiber [91].

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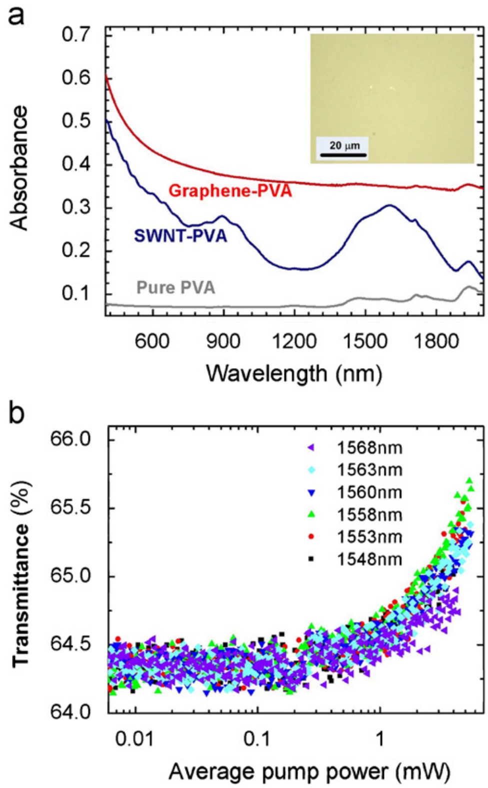

Fig. 1. (a) Absorption of SWNT and graphene polymer composite SA. Inset: micrograph of a graphene polymer composite. (b) Typical GSA transmittance as a function of input power at different wavelengths, adapted from Ref. [38].

Different approaches have been used to integrate SWNT-SAs in lasers, such as free-space coupling (Fig. 2(a)) [49,50], deposition on fiber ends [55,161] and inside fibers [67,92] as well as eveanescent field interaction [56,66,81,84,93], Thus far, the most popular way to integrate SWNT-SAs into fiber lasers is to sandwich a SWNT polymer composite between two fiber connectors (Fig. 2(b)) [18,27,28,51–53,61,62,68–70,74,87–90], since this offers ease of integration into various lightwave systems [18,27,28,51–53,61,62,68–70,74,87–90]. A large range of host polymers, e.g. polycarbonate (PC) [51,64,75,94,95], polyvinyl alcohol (PVA) (Fig. 1(a)) [27,28,52,53,61,62,68–70,74,87–90, 96–98], Carboxymethyl cellulose (CMC) [47,42,43,71,99–104], Polyimide (PI) [42,63,105,106], Polydimethylsiloxane (PDMS) [65,66,107–109], Polymethyl methacrylate (PMMA) [44–46,48,60, 92,110–112] and poly(3-hexylthiophene) (P3HT) [113–115], have been used. SWNTs utilized in SAs have been prepared using a variety of growth techniques, e.g. laser ablation (LA) [27,28,42,49–51,61–64,68–70,74,75,90,94,96–98,102,105], arc discharge (AD) [44,45,71,103,104,110,116,117] and various chemical vapor deposition (CVD) methods [118,119] (such as Cobalt–Molybdenum catalyst (CoMoCAT) [42,52,53,87–89,115], High Pressure Carbon Monoxide (HiPCO) [43,46–48,55,60,65–67, 76–78,81,86,92,95,99–101,107,108,111–114,120–125] and alcohol catalytic chemical–vapor deposition (ACCVD) [59,83–85]), allowing the selection of different diameters and diameter distributions.

Since the first demonstration in 2003 [49], the performance of ultrafast lasers mode-locked by SWNTs has steadily improved. Table 1 summarizes representative output performances. For example, the average output power has increased from few hundred μw (e.g. ~260 μw [50]) to few Watts [28,108,126], with peak powers reaching a few hundred kW (e.g. 200 kW in Ref. [120]). A large range of output parameters, such as wavelength, pulse duration, and repetition rate have been achieved. Thus far, the demonstrated wavelengths range from 0.78 [47,127] to 2 μm [57,71,110,128]. The output pulse durations range from a few ns [52,53] to sub-20 fs [107]. The repetition rate spans from few tens kHz [52,53,129] to a few tens GHz [73,130]. Wavelength-tunable lasers based on SWNTs have also been reported [47,51,63,98, 128,131–134].

2.1. SWNT mode-locked solid-state lasers

Solid-state lasers, mainly using doped glass [3,10] or crystalline host materials [3,10] as gain media, are the most commonly used in various applications (e.g. industry, research and military [3,10,11]). They typically consist of a free-space cavity, formed by mirrors and a solid-state gain medium [178]. A variety of solid-state gain media have been coupled with SWNT-SAs to mode-lock solid-state lasers. These include Nd:glass [42,43,152,157], Nd:$GdVO_4$ [99,135,171], Nd:YVO_4 [117,126,137,155,172], Nd:YAG [100,101,158], Nd:YLF [170], Nd:$LuYVO_4$ [136], Er:glass [42,82,122], Yb:Sc_2SiO_5 [173], Yb:$KLu(WO_4)_2$ [44,45], Yb:KYW [46], Yb:$LuYSiO_5$ [174,175], Yb:$LuScO_3$ [179], Cr:YAG [46,133], Cr:LiSAF [134], Cr:forsterite [46,48] and Tm:$KLu(WO_4)_2$ [110]. In particular, SWNT-SAs have recently been reported to mode-lock solid-state Ti:sapphire lasers [47,127]. This is an important step, since Kerr-lens mode-locked Ti:sapphire lasers dominate the sub-200fs market. However, Kerr-lens mode-locking is not self-starting [1,11] and usually requires critical cavity alignment [1,3–5].

The output spectra of SWNT mode-locked solid-state lasers have thus far covered 0.8 [47,127,134], 1 [42–46,179,175], 1.2 [46,48], 1.3 [99–101,155], 1.5 [42,46,82,122,133] and 2 μm [110]. SWNTs are normally coated on high reflectivity mirrors [42,44,46,82,134], and then employed as cavity mirror. High transmittance substrates (e.g. pure quartz [45,100]) coated with SWNTs (Fig. 2 (a)) have also been used [45,100,133,126]. The shortest pulse duration thus far achieved is ~60 fs [42,127]. High-power up to 3.6 W was reported at 1.06 μm [126,135]. Compared to fiber lasers, optimization of the SA non-saturable losses is crucial to mode-lock solid-state lasers [3,46], since their gain is lower, mainly due to the limited gain medium length (several mm) [3,10]. For wide-band operation, SWNTs covering a variety of diameters need be combined to form the SA device. These tubes, however, tend to bundle and curl [18,46], thus contributing to high non-saturable losses [18]. Recently, a single SWNT-SA was used to mode-lock solid-state lasers at 1, 1.2 and 1.5 μm [46], showing that non-saturable losses may be decreased by optimizing the SWNT-SA fabrication.

2.2. SWNT mode-locked fiber lasers

Fiber lasers are attractive alternatives to bulk solid state lasers due to their efficient heat dissipation [3,13] and alignment-free format [3], the latter being a key advantage for end-users. Furthermore, their typical gain can reach several tens dB [11], a few orders of magnitude higher than solid state lasers [10,11]. Thus, they do not need particular optimization of non-saturable losses for their operation, as the loss can be compensated by the large gain. Indeed, there has been a far greater research effort on exploiting SWNT-SAs in fiber lasers, as evident from Table 1, with a steady improvement in performance. A typical mode-locked fiber laser setup is shown in Fig. 2(c). This unidirectional ring cavity design allows easy self-starting due to decreased spurious reflections [180]. The maximum reported average output power is ~250 mW [84], with 6.5 nJ output pulse energy at ~1.5 μm [84]. In Ref. [84] SWNTs were transferred on a D-shaped fiber to enable evanescent field interaction, a technique also reported in Refs. [56,66,81,93]. Normal-dispersion fiber lasers [28,52,53,88,145,112] have also been demonstrated,

Table 1
Pulsed lasers exploiting SWNT-SAs. λ: wavelength; τ: pulse width; f: repetition rate; P: average output power.

SAs		SWNT types	Laser types	Laser parameters			
				λ (nm)	τ	f (MHz)	P (mW)
Polymer composites	PC	LA [51,64,75], HiPCO [95]	EDFL [51,64,95], waveguide laser [75]	1518–1558 tunable [51], 1560 [64,75,95]	115 fs [95], 2.4 ps [51]	15 [51], 39 [95]	0.36 [51], 3.4 [95]
	PVA	CoMoCAT [52,53,87,88], LA [27,28,61,62,68–70,74,90,96–98], AD [135,136], HiPCO [125]	$Nd:YVO_4$ [137], $Nd:GdVO_4$ [135], $Nd:LuYVO_4$ [136], YDFL [52,53,87,138], BDFL [88], EDFL [27,28,61,62,68–70,90,96–98,125,139–142], Waveguide laser [74]	1058–1060 [52,53,138], 1530–1563 tunable [98], 1532–1563 [27,28,61,62,68,69,74,90,96,97,125,139,140,142], 1601 [70]	113 fs [69], 20 ps–2 ns selective [52,53,87]	0.177–21 selective [52,53,87], 328 [141]	0.1 [74,90], 3.6 W [135]
	CMC	CoMoCAT [42], HiPCO [43,47,99–101,143,144], LA [102], AD [71,103,104,145,146]	Ti:sapphire [47,143], Nd:glass [42,43], $Nd:GdVO_4$ [99], Nd:YAG [100,101], YDFL [144–146], EDFL [102–104,144,146–149], TDFL [71]	780–820 tunable [47,143], 1000–1068 [42,43,144–146], 1320–1340 [99–101], 1550–1565 [102,104,144,146–149], 1930 [71]	177 fs [104], 1.15 ns [145]	37 [71], 110 [47]	3.4 [71], 450 [47]
	PI	LA [42,63,105,106,150], HiPCO [65]	Er:glass [42], EDFL [63,105,106,118,119,150]	1532–1562 tunable [63], 1532 and 1557 Switchable, 1545–1570 [42,105,106,119,150]	68 fs [42], 6.2 ps [150]	0.13 [150], 85 [42]	0.4 [150], 114 [106]
	PDMS	HiPCO [32,66,107–109,128,151]	EDFL [32,66,93,107–109,151], YDFL [56], TDFL [72,128]	1035 [56], 1530–1565 [66,93,107,108,151], 1885 [72], 1866–1916 tunable [128], 1000–1750 [107,108]	14 fs [107], 1.5 ps [56]	13.3 [66], 4 GHz [109]	1 [93], 11.5 [108]
	PMMA	AD [44,45,110,152], HiPCO [46,48,60,92,111,112,127,134,153–155]	EDFL [60,91,92,111,112,154], Yb:KLuW [44,45], Yb:KYW [46,131], Nd:BaYF [156], Nd:Glass [152,157], $Nd:YVO_4$ [155], Cr:forsterite [48,46,153], Cr:LiSAF [134], Ti:sappire [127], Cr:YAG [46,133], Tm:KLuW [110]	780–825 tunable [127], 868–882 tunable [134], 1035–1045 tunable [131], 1061–1075 tunable [157], 1048–1080 [44,131,152,156], 1240–1250 [48,153], 1342 [155], 1435–1505 [133], 1560–1567 [60,91,92,111,112,154], 1944 [110]	62 fs [127], 9.7 ps [110]	5.3 [111], 1.69 GHz [112]	50 μw [60], 800 [155]
	P3HT	HiPCO [113,114] CoMoCAT [115]	EDFL [113,114], YDFL [115]	1070 [115], 1560 [113,114]	113 fs [114]	51 [113]	5 [114]
	PFO	//	Nd:YAG [158]	1064 [158]	8.3 ps [158]	90 [158]	275 [158]
	PS	HiPCO [60]	EDFA [60]	1560 [60]	171 fs [60]	7.63 [60]	0.050 [60]
	SU8	HiPCO [159]	EDFA [159]	1571 [159]	871 fs [159]	21.27 [159]	1 [159]
Grown/ deposited/ transferred SWNTs	Optically driven deposition	HiPCO [55,120,121,160], CoMoCaT [55]	YDFL [55], EDFL [55,120,121,160–163]	1070 [55], 1532–1567 [55,120,121,160–162]	124 fs [163], 1.14 ps [121]	5.2 [160]	0.1 [55], 1.5 W [120]
	Spray-coating	HiPCO [76–78,81,130], LA [49,50]	PDFL [76], TDFL [80], EDFL [30,49,50,77,78,80,81,132,164], EYDFL [79,130], Er:Yb:glass [82], Semiconductor laser [73]	1294 [76], 1506 [80], 1550–1571 [30,49,50,73,77–82,130], 1605 [80]	190 fs [164], 14 ps [73]	3.18 [76], 19.4 GHz [130]	16 μw [73], 63 [82]
	Grown/ transferred	ACCVD [59,83–85], HiPCO [57,86,165–168]	YDFL [57], EDFL [31,57,59,83–86,165–168], TDFL [57,167]	1050 [57], 1550–1565 [31,57,59,83–86,165,167,168], 1990 [57,167]	30 fs [31], 1.14 ps [85]	6.62 [85], 50 [59]	22 μw [85], 250 [84]
	Drop-casting	AD [117,126,169–171], CVD [172–174]	Nd:YLF [170], $Nd:YVO_4$ [117,126,172], $Nd:GdVO_4$ [171], Yb:SSO [173], $Yb:LuYSiO_5$ [174,175]	1045/1059 [174], 1047–1064 [126,169–171,173,175]	1.1 ps [169], 15 ps [173]	79.7 [117]	280 [170], 3.6 W [126]
Sol–gel glass		//	EDFL [176,177]	1559–1563 [176,177]	0.57 ps [177], 2.3 ps [176]	2.96 [176]	2 [177]
Solution	Cell	HiPCO [122,123]	Er:glass [122], F_2^-:LiF [123]	1150 [123], 1540 [122]	<1 ns [122]	//	//
	Micro-channel	HiPCO [67,124]	EDFL [67,124]	1566 [67]	0.9 ps [124], 2.3 ps [67]	2.56 [67], 5.26 [124]	15 [124], 22.4 [67]

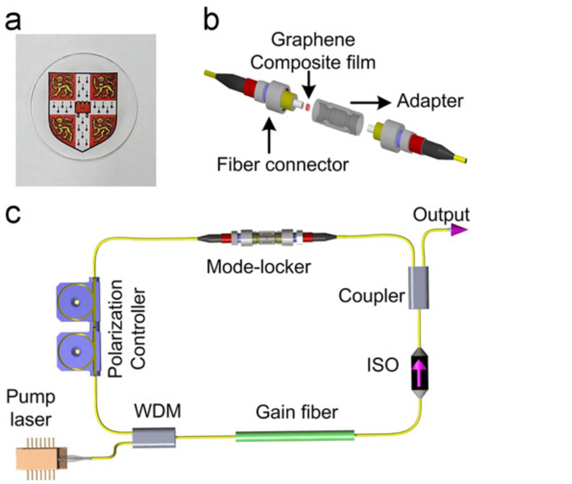

Fig. 2. (a) Graphene polymer composite coated on a quartz substrate. Its transparency to light (indicative of low loss) makes this suitable for integration in solid-state and waveguide lasers (b) Integration of a graphene saturable absorber polymer-composite film between two fiber connectors for fiber lasers. Note that these integration methods (as shown in (a) and (b)) also apply to SWNT-SA integration. (c) Typical passively mode-locked fiber laser setup. WDM: wavelength division multiplexer; ISO: isolator.

with 155 mW average output power and 3 nJ pulse energy at $\sim$1.03 μm [56]. Ref. [102] reported 63 nJ pulse generation, the highest to date from a SWNT-SA enabled ultrafast laser. Ref. [181] theoretically predicted that SWNT mode-locked fiber laser could achieve up to 330 nJ.

The shortest pulse achieved thus far from SWNT-SA mode-locked fiber lasers is $\sim$84 fs [182], by using a stretched-pulse design [69], i.e. alternating normal and anomalous dispersion, to obtain periodic stretching and compression of the intracavity pulses in the resonator [69,183]. Thus, the average pulse width in one cavity round trip can increase by an order of magnitude compared to the typical solition design [69,183], and ultrafast (e.g. 77 fs from a nonlinear polarization evolution (NPE) mode-locked Erbium (Er) doped fiber laser (EDFL) [183]) pulses are achievable due to the reduced nonlinear effects [69,183]. A typical autocorrelation trace of a stretched-pulse fiber laser is shown in Fig. 3 (a). Selectable pulse duration, from 20 ps to 2 ns, was also demonstrated by changing cavity length (shown in Fig. 3(b)) [52,53]. A short cavity design [79,130] and harmonic mode-locking (where multiple pulses circulate in the laser resonator at an integer multiple of the fundamental frequency [11]) can allow high repetition rate. Ref. [130] reported SWNT mode-locked pulses up to $\sim$20 GHz, using a cavity formed by a $\sim$5 mm fiber sandwiched between two mirrors.

Several gain fibres have been used to date, including Ytterbium (Yb) doped (YDFL) [52–57,146], EDFL [27,28,54,55,57,59,61,62,64, 67–69,109,112,125,146,184], Er and Yb co-doped (EYDFL) [54,130], Bismuth (Bi) doped (BDFL) [88,89], Praseodymium (Pr) doped (PDFL) [76] and Thulium (Th) doped (TDFL) [57,71,72]. Amongst them, EDFLs are the most popular, since they allow easy excitation of soliton pulses in single mode fibers [3], and all necessary components are economically available from the fiber-telecom market [3]. The achieved wavelength range covers 1 [52–57], 1.1 [88,89], 1.3 [76], 1.5 [27,28,49,50,57–69], 1.6 [70], and 2 μm [57,71,72]. Ref. [51] first demonstrated wavelength-tunable devices. Later, by using a single SWNT-SA device, Ref. [57] achieved mode-locking at 1, 1.5 and 2 μm, in YDFL, EDFL and TDFL, respectively. Wide-band operation requires the combination of SWNTs with different diameters [18,51,57]. Ref. [146] demonstrated the synchronization of two all-fiber mode-locked lasers, operating at $\sim$1 and 1.54 μm, coupled through a shared

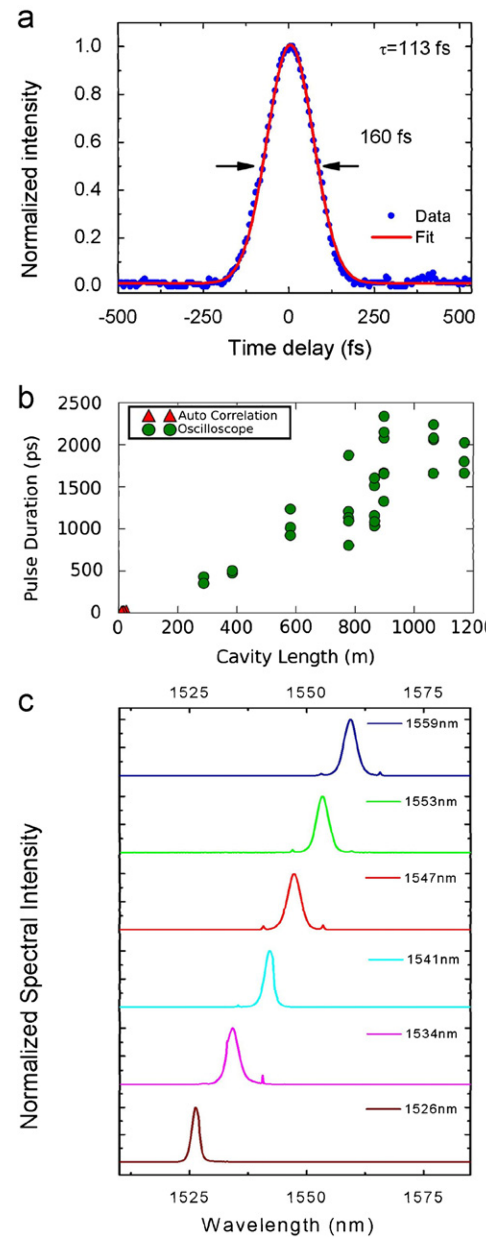

Fig. 3. (a) Autocorrelation trace of output pulses with Gaussian fit. (b) Output pulse duration as a function of cavity length, adapted from Ref. [52]. (c) Tunable mode-locked fiber laser spectra, adapted from Ref. [40].

SWNT-SA. Refs. [185–188] reported Raman fiber lasers mode-locked by nanotubes at 1.1 [187,188] and 1.6 μm [185,186], when pumped at 1 and 1.5 μm, respectively.

Pulses from SWNT mode-locked fiber lasers have also been used for further investigations, e.g. nonlinear compression [65], amplification [28,31,32,108,120] and super-continuum generation [31,107,108,130,189]. An output average power of up to 1.6 W has been achieved by using direct amplification of a SWNT enabled chirp-pulse oscillator [28], with the potential for further scaling of output power/pulse energy [28]. 11.5 W pulses were reported with three cascaded amplifiers [108]. After the grating compressor, 135 fs pulses were generated with 5.7 W output power and 160 nJ pulse energy in Ref. [108]. Note that in these amplification experiments [28,108], the output power is just limited by the pump power, i.e. even higher power could be possible by increasing the pump power. Ref. [31] seeded the output pulses from a SWNT mode-locked oscillator into an amplifier, followed by a highly nonlinear fiber, to build a fiber laser based frequency comb (i.e. a light source with optical spectrum consisting of equidistant lines [11]). Supercontinuum generation (a nonlinear process to strongly broaden the spectrum of light [11]) has been demonstrated with 30-fs output pulses [31]. Using supercontinuum generation, Ref. [107] reported 14 fs pulses by seeding a SWNT mode-locked fiber laser into a nonlinear fiber, with output spectrum covering from 1 to 1.75 μm.

2.3. SWNT mode-locked semiconductor laser

Semiconductor lasers usually exploit direct band gap semiconductors as gain medium [11]. Since such lasers can be electrically pumped [11], they have been widely employed in a range of common devices, from home entertainment [190] (e.g. CD/DVD players) to telecommunications [11]. Furthermore, semiconductor lasers are attractive also because of their inherent simplicity and compactness [11]. The output pulse repetition rate (f_{rep}) is linked to the cavity length (L) by $f_{rep} = c/(2\,n\,L)$, where c is the speed of light, and n is the refractive index of the cavity material [1]. Therefore, mode-locked semiconductor lasers typically offer high (> GHz) repetition rate due to their relatively short (~ a few mm) cavity length. Consequently, mode-locked semiconductor lasers are particularly suitable for high-speed optical communications [1]. Ref. [73] first demonstrated a SWNT-SA mode-locked semiconductor laser with repetition rate up to 17.2 GHz, using a semiconductor optical amplifier to provide gain at ~1.5 μm. The full width at half maximum (FWHM) of the pulses was 0.73 nm at ~1570 nm, with a duration ~14 ps [73].

2.4. SWNT mode-locked waveguide lasers

Solid-state waveguide lasers are built on planar or channel waveguides in polymer, glass or crystalline substrates [11]. Passive mode-locking of waveguide lasers is also inherently simple and compact. Compared to traditional semiconductor lasers (having an upper-state lifetime in the order of ns [190]), solid-state waveguide lasers are more suitable for high-energy pulse generation, because the gain materials typically have longer upper-state lifetime (~ ms) [11]. Therefore, more energy can be stored inside the gain material for high-energy pulse generation [10,11]. A variety of devices, such as waveguides [191], couplers [192], gratings [193], optical amplifiers [194] and oscillators [194] have been fabricated by ultrafast inscription in transparent substrates, a technique not needing any photolithographic process, and allowing three-dimensional device fabrication [194].

Ref. [74] first demonstrated mode-locking with a SWNT-SA in an active waveguide laser fabricated by ultrafast laser inscription. An Er and Yb co-doped phosphate glass waveguide was used, providing net gain over the whole telecom C Band and 7.3 dB (~5.4) peak gain at 1535 nm [74]. The laser generated 1.76 ps transform-limited pulses [74].

Recently, bismuthate glasses have been employed as waveguide gain media, as they can be doped with sufficient concentrations of Er-ions for high gain. Ref. [195] reported that a bismuthate waveguide amplifier can exhibit a peak net gain ~16 dB (~40) at 1533 nm and a wide and flat gain spectrum [195], favorable for ultrafast pulse generation [11]. Ref. [75] achieved 320 fs pulses in an Er-doped bismuthate glass waveguide laser mode-locked by a SWNT-SA, with an output spectral width ~8.9 nm at 1.55 μm [75].

3. Graphene mode-locked lasers

Graphene is at the center of an ever growing research effort due to its unique electronic properties [230–236]. Near-ballistic transport at room temperature [231,237] and high mobility [234–238] make it a potential material for nanoelectronics [238–241], especially for high frequency applications [238–241]. Furthermore, its optical properties are ideal for transparent conducting films [39,242,243] and electrodes [39,244], photodetectors [39,245,246] and optical modulators [39,247,248].

The ultrafast nonlinear properties of graphene have been intensively investigated [34–37]. Two relaxation time scales are typically observed. A faster one (< 100 fs), usually associated with carrier–carrier intra-band collisions and phonon emission [34,36,37], and a slower one (~ ps), corresponding to electron inter-band relaxation and cooling of hot phonons [34–37,39,249]. Graphene is thus an ultrafast SA material [18,38,39]. Ref. [18] first reported a GSA mode-locked laser. Subsequently, a variety of lasers mode-locked by graphene were demonstrated [38–41,200,201,203–205,207,208,214,216,226], as shown in Table 2.

Graphene has been sourced in various ways, such as liquid phase exfoliation [18,38,40,199], CVD [200,201,203–205], carbon segregation [218], graphene oxide, GO, [39,208,221]), reduced GO [214,216] and micro-mechanical cleavage [39,209–211]. As shown in Table 2, several approaches (e.g. sandwiching [18,38], free-space coupling [216,227], placement inside Photonic Crystal Fibers (PCFs) [215], evanescent field interaction [214]) have been used to integrate GSAs into cavities, mostly following previous approaches used for SWNT-SAs. Sandwiching a GSA between two fiber connectors (Fig. 2 (b)) is thus far the most common approach for GSA integration [18,38,40,41,200,201,203–205,207,208,210].

Compared to traditional SAs (e.g. SESAMs) and SWNT-SAs, the major advantage of using graphene is the intrinsic wide-band operation. Thus far, GSAs have been used to produce pulses at 1 [216], 1.25 [226], 1.5 [18,38–41,199–201,203–205,207,208,210,214], and 2 μm [225,250]. Similar to SWNT-SAs, GSAs have been mostly combined with EDFLs [18,38,40,41,199–201,203–205,207,208,210,214], not because GSAs have any preference for a particular wavelength, but because EDFLs can easily produce soliton pulses in single mode fibers [3], and all necessary components are economically available from the optical telecom market [3]. Ref. [40] reported GSA mode-locked fiber lasers tunable in the 1525–1559 nm range (Fig. 3 (c)), only limited by the filter used in the cavity [39,40]. Ref. [205] reported ~240 fs tunable pulse generation using fiber lasers under different operation regimes (e.g. from all-anomalous to all-normal dispersion). Stretched-pulse design was employed, generating sub-200fs pulses [196]. Ref. [251] reported 163nJ pulse generation. Refs. [216,221,220,224–227] also reported pulses using solid-state lasers mode-locked by GSAs. 94-fs tunable (~1.22–1.27 μm) pulses have been achieved with a GSA mode-locked solid-state Cr:forsterite laser [226]. High-power (~1 W) pulses have been demonstrated with a GSA mode-locked Nd:YVO_4 solid-state laser [220,221].

4. Outlook

Currently, solid-state and fiber lasers are the most common for high output power/pulse energy applications [1,3]. Amongst various solid-state laser configurations, a thin-disk design can significantly reduce thermal effects and nonlinearities [252], enabling high average power and energy pulses [252,253]. SWNT-SAs and GSAs could be used in thin-disk designs for this purpose. The main challenge is the relatively large non-saturable loss of these SAs, which can be addressed by further device optimization (e.g. enrichment in semiconducting nanotubes [18]).

The output peak power of fiber lasers is restrained by enhanced nonlinear effects [1,3,254]. Recently, large-mode-area fiber based ultrafast lasers working in a dissipative soliton regime have been demonstrated for high average power pulses [255,256],

Table 2
Pulsed lasers using GSAs. LPE: liquid phase exfoliation; GO: graphene oxide; FG: functionalized graphene. CS: carbon segregation. MMC: micro-mechanical cleavage. RGO: reduced GO.

Laser type	Coupling means	Fabrication method	Laser parameters				Ref.
			λ (nm)	τ	f (MHz)	P (mw)	
EDFL	Sandwiching	LPE	1557	800 fs	//	//	[18]
			1559	464 fs	19.9	//	[38]
			1525–1559 tunable	1 ps	8	1	[40]
			1560	174 fs	27.4	1.2	[196]
			1562	630 fs	19.9	//	[41]
			1522–1555 tunable	2 μs	0.036–0.1	3.4	[197]
			1519–1569 tunable	4.6 μs	0.008–0.029	2.4	[198]
			1532	850 fs	5.27	//	[160]
			1565	190 fs	42.8	0.4	[199]
		CVD	1565	756 fs	1.79	2	[200]
			1576	415 fs	6.84	50	[201]
			1561	1.23 ps	3	3	[202]
			1594	2.1,71 ps	//	//	[203]
			1570–1600 tunable	40–140 ps	1.5	//	[204]
			1570–1600 tunable	240–655 fs, 70–150 ps	//	//	[205]
			1538	206 ns	0.031–0.236	7.8	[206]
		FG	1559	743 fs	//	//	[39]
			1590	700 fs	6.95	50	[207]
			1570–1600 tunable	1.08 ps	6.95	//	[208]
		MMC	//	3.2 ps	10.9	3	[209]
			1566	0.88 ps	6.22	//	[210]
			1561	480 fs	7	//	[211]
		RGO	1566.1/1566.3	3.7−18 μs	0.003–0.065	1.1	[212]
			1572.6	//	91.5	//	[213]
	Evanescent field		1561	1.3 ps	6.99	15.5	[214]
	PCF	FG	1561	4.85 ns	7.68	4.3	[215]
Nd:YAG	Free-space	RGO	1064	4 ps	88	100	[216]
		RGO	1064	260 ns	0.167	1389	[217]
		CS	1064	161–400 ns	0.3–0.66	105	[218]
$Nd:LuVO_4$			1064	56–131 ns	0.89	474	[219]
$Nd:YVO_4$		GO	1063	//	75	1000	[220]
			//	//	88	1200	[221]
$Nd:GdVO_4$			1064	105–1435 ns	0.3–0.7	2300	[222]
			1065	16 ps	43	360	[223]
Yb:KGW			1031	428 fs	86	504	[224]
$Tm:YAlO_3$			2023	~10 ps	71.8	268	[225]
Cr:forsterite		CVD	1222–1227 tunable	94 fs	74.6	230	[226]
Er:Yb:glass			1552	260 fs	88	4.5	[227]
YDFL	Sandwiching		1069	580 ps	0.9	0.37	[228]
		LPE	1064	70–250 ns	0.14–0.257	12	[229]

reaching MW peak powers [257]. In principle, large-mode-area fiber lasers mode-locked with SWNTs and graphene may deliver better performances (e.g. high average power, high peak power, system simplicity). For example, coating SWNT-SAs and GSAs on the fiber surfaces to enable evanescent-wave interaction [56,84] or inside the fibers [67] (e.g. holes of PCFs [92,215]) can preserve the alignment-free waveguide format of such fiber lasers, by removing the free-space components, which are necessary for traditional SA (e.g. SESAMs [257]) coupling. These integration strategies can be applied to various laser designs, such as waveguide (e.g. laser inscribed [194] or polymer [258]) and semiconductor (e.g. vertical external cavity surface-emitting semiconductor lasers [259] and optically pumped semiconductor disk lasers [260]) for high power/energy pulse generation. This technology can also enable compact lasers with repetition rates up to hundreds of GHz [29,130]. Another option to increase repetition rate is via harmonic mode-locking [1].

The combination of wide-band gain materials (for example Ti:sapphire) and SWNT/graphene SAs could produce novel broadband tunable ultrafast sources. Note that GSAs can intrinsically operate at "full" bandwidth [18,38,39], while the output wavelength or tuning spectral range of a traditional laser will be ultimately constrained by the gain medium. Nonlinear effects (e.g. optical parametric generation [261,262] and Raman scattering [9,14]) can be used to broaden the spectral range. They can provide broadband gain, potentially covering from ultraviolet [263] to terahertz [264]. The recent demonstration of broadband Raman gain [185–188] and broadband SWNT-SAs/GSAs shows the possibility of getting broader output spectra than ever before.

External amplification of SWNT and graphene mode-locked lasers [28,31,108,120] or coherent combination of various lasers [265–267] could boost output power and energy. Nonlinear frequency conversion (e.g. harmonic frequency generation [1,268–271], parametric oscillation [261,272,273] and amplification [1,262], four-wave mixing [11], supercontinuum generation [31,107,108,263,189,130]) is also an useful way to expand the wavelength accessibility after the oscillator. External-cavity pulse compression (e.g. nonlinear compression [11,31,65,107,108]) could be used to generate shorter pulse down to a few optical cycles (e.g. 4.3-fs [274]).

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