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.

Keywords:
Nanotubes, Graphene, Mode-locking, Lasers, Ultrafast Photonics

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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 1.1μm[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 broad-band gain, spectral range and gain bandwidth they enable[11–13]. E.g., 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 (~300-2300nm)[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~70dB (10^7)[11].

The search for alternative materials to be used as saturable absorbers (SA), 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],

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] and inside fibers[67, 92], evanescent 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)[42, 43, 47, 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 laser mode-locked by SWNTs has steadily improved. Table 1 summarizes representative output performances. For example, the average output power has increased from few hundreds μw (e.g.$\sim$260μw [50]) to few Watts[28, 108, 126], with peak powers reaching a few hundreds kW (e.g. 200kW 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 2μm [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, 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$_2$SiO$_5$[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].

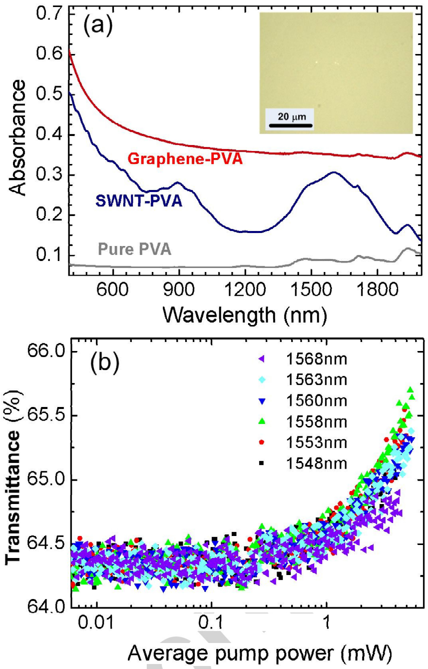

Figure 1: (a) Absorption of SWNT-SA and graphene-SA. Inset: micrograph of a graphene-SA. (b) Typical GSA transmittance as a function of input power at different wavelengths, adapted from Ref.[38].

The output spectra of SWNT mode-locked solid-state lasers have thus far covered 0.8[47, 127, 134], 1 [42–46, 175, 179],1.2[46, 48], 1.3[99–101, 155], 1.5[42, 46, 82, 122, 133] and 2μm[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, 126, 133]. The shortest pulse duration thus far achieved is 68fs at 1.5μm[42]. High-power up to 3.6W was reported at 1.06μm[126]. 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 1.5μm[46], showing that non-saturable losses may be decreased by optimizing the SWNT-SAs fabrication.

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]	115fs [95], 2.4ps [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]	113fs [69], 20ps-2ns selective [52, 53, 87]	0.177-21 selective [52, 53, 87], 328 [141]	0.1 [74, 90], 3.6W [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]	177fs [104], 1.15ns [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&1557 Switchable, 1545-1570 [42, 105, 106, 119, 150]	68fs [42], 6.2ps [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]	14fs [107], 1.5ps [56]	13.3 [66], 4GHz [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 [46, 48, 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]	62fs [127], 9.7ps [110]	5.3 [111], 1.69GHz [112]	50μw [60], 800 [155]
	P3HT	HiPCO [113, 114] CoMoCAT [115]	EDFL [113, 114], YDFL [115]	1070 [115], 1560 [113, 114]	113fs [114]	51 [113]	5 [114]
	PFO	//	Nd:YAG [158]	1064 [158]	8.3ps [158]	90 [158]	275 [158]
	PS	HiPCO [60]	EDFA [60]	1560 [60]	171fs [60]	7.63 [60]	0.050 [60]
	SU8	HiPCO [159]	EDFA [159]	1571 [159]	871fs [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]	124fs [163], 1.14ps [121]	5.2 [160]	0.1 [55], 1.5W [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]	190fs [164], 14ps [73]	3.18 [76], 19.4GHz [130]	16μw [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]	30fs [31], 1.14ps [85]	6.62 [85], 50 [59]	22μw [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]	1.1ps [169], 15ps [173]	79.7 [117]	280 [170], 3.6W [126]
Sol - gel glass		//	EDFL [176, 177]	1559-1563 [176, 177]	0.57ps[177],2.3ps [176]	2.96 [176]	2[177]
Solution	Cell	HiPCO [122, 123]	Er:glass [122],F$_2^-$:LiF [123]	1150 [123], 1540 [122]	$< 1ns$ [122]	//	//
	Micro-channel	HiPCO [67, 124]	EDFL [67, 124]	1566 [67]	0.9ps [124], 2.3ps [67]	2.56 [67], 5.26 [124]	15 [124], 22.4 [67]

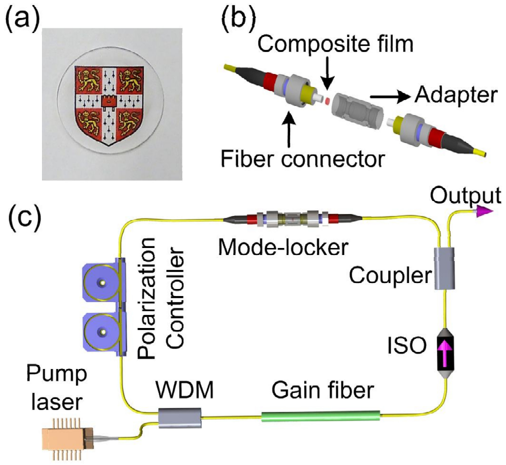

Figure 2: (a) Graphene coated on a quartz substrate. (b) Integrated GSA fiber device for fiber lasers. Note that these integration methods (as shown in (a) and (b)) also apply to SWNT-SA integration. (c) Typical GSA mode-locked fiber laser setup. WDM: wavelength division multiplexer; ISO: Isolator.

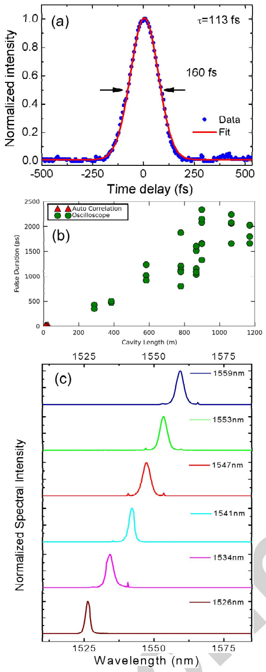

Figure 3: (a) Autocorrelation trace of output pulses with Gaussian fit. (b) Output pulse duration as a function of cavity length, adapted from Ref.[52]. (c) Tunable mode-locked fiber laser spectra, adapted from Ref.[40].

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$\sim$250mW[84], with 6.5nJ output pulse energy at$\sim$1.5μm[84]. In Ref.[84] SWNTs were transferred on a D-shaped fiber to enable evanescent field interaction, a technique also reported in Refs.[56, 66, 81, 93]. Normal-dispersion fiber lasers[28, 52, 53, 88, 112, 145] have also been demonstrated, with 155mW average output power and 3nJ pulse energy at$\sim$1.03μm[56]. Ref.[102] reported 63nJ pulse generation, the highest to date from a SWNT-SA mode-locked laser. Ref.[181] theoretically predicted that SWNT mode-locked fiber laser could achieve up to 330nJ.

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

Several fibres have been used to date, including Ytterbium (Yb) doped (YDFL)[52–57, 146], Erbium (Er) doped (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 2μm[57, 71, 72]. Ref.[51] firstly demonstrated wavelength-tunable devices. Later, by using a single SWNT-SA device, Ref.[57] achieved mode-locking at 1,1.5 and 2μm, in YDFL, EDFL, TDFL. 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 ∼1 and 1.54μm, coupled through a shared SWNT-SA. Refs.[185–188] reported Raman fiber lasers mode-locked by nanotubes at 1.1[187, 188] and 1.6μm [185, 186],when pumped at 1 and 1.5μm, 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 supercontinuum generation[31, 107, 108, 130, 189]. An output average power of up to 1.6W 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]. 11.5W pulses were reported with three cascaded amplifiers[108]. After the grating compressor, 135fs pulses were generated with 5.7W output power and 160nJ pulse energy in Ref.[108]. Note that in these amplification experiments[28, 108], the output power is juts 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, i.e. 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 14fs pulses by seeding a SWNT mode-locked fiber laser into a nonlinear fiber, with output spectrum covering from 1 to 1.75μm.

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 $f_{rep} = \frac{c}{2*n*L}$, 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 17.2GHz, using a semiconductor optical amplifier to provide gain at∼1.5μm. The full width at half maximum (FWHM) of the pulses was 0.73nm at ∼1570nm, with a duration∼14ps[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 of 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 7.3dB (∼5.4) peak gain at 1535nm[74]. The laser had 1.76ps 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 ∼16dB (∼40) at 1533nm and a wide and flat gain spectrum[195], favorable for ultrafast pulse generation [11]. Ref.[75] achieved 320fs pulses in an Er-doped bismuthate glass waveguide laser mode-locked by a SWNT-SA, with an output spectral width∼8.9nm at 1.55μm [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 carriercarrier 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 ultrafast SA material[18, 38, 39]. Ref.[18] first reported a GSA

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	800fs	//	//	[18]
			1559	464fs	19.9	//	[38]
			$1525-1559$tunable	1ps	8	1	[40]
			1560	174fs	27.4	1.2	[196]
			1562	630fs	19.9	//	[41]
			$1522-1555$tunable	2μs	$0.036-0.1$	3.4	[197]
			$1519-1569$tunable	4.6μs	$0.008-0.029$	2.4	[198]
			1532	850fs	5.27	//	[160]
			1565	190fs	42.8	0.4	[199]
		CVD	1565	756fs	1.79	2	[200]
			1576	415fs	6.84	50	[201]
			1561	1.23ps	3	2.5	[202]
			1594	$2.1, 71ps$	//	//	[203]
			$1570-1600$tunable	$40-140ps$	1.5	//	[204]
			$1570-1600$tunable	240-655fs,70-150ps	//	//	[205]
			1538	206ns	$0.031-0.236$	7.8	[206]
		FG	1559	743fs	//	//	[39]
			1590	700fs	6.95	50	[207]
			$1570-1600$tunable	1.08ps	6.95	//	[208]
		MMC	//	3.2ps	10.9	3	[209]
			1556	0.88ps	6.22	//	[210]
			1561	480fs	7	//	[211]
		RGO	1566.1/1566.3	3.7-18μs	0.003-0.065	1.1	[212]
			1572.6	//	91.5	//	[213]
	Evanescent field		1561	1.3ps	6.99	15.5	[214]
	PCF	FG	1561	4.85ns	7.68	4.3	[215]
Nd:YAG	Free-space	RGO	1064	4ps	88	100	[216]
		GO	1064	260ns	0.167	1389	[217]
		CS	1064	$161-400ns$	$0.3-0.66$	105	[218]
Nd:LuVO$_4$			1064	$56-131ns$	0.89	474	[219]
Nd:YVO$_4$		GO	1063	//	75	1000	[220]
			//	//	88	1200	[221]
Nd:GdVO$_4$			1064	$105-1435ns$	$0.3-0.7$	2300	[222]
			1065	16ps	43	360	[223]
Yb:KGW			1031	428fs	86	504	[224]
Tm:YAlO$_3$			2023	$\sim$10ps	71.8	268	[225]
Cr:forsterite		CVD	$1222-1227$tunable	94fs	74.6	230	[226]
Er:Yb:glass			1552	260fs	88	4.5	[227]
YDFL	Sandwiching		1069	580ps	0.9	0.37	[228]
		LPE	1064	70ns	$0.14-0.257$	12	[229]

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 indicated 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 PCF[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 2μm[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 1525-1559nm range (Fig.3 (c)), only limited by the filter used in the cavity[39, 40]. Ref.[205] reported$\sim$240fs tunable pulse generation using fiber lasers under different operation regimes (e.g. from all-anomalous to all-normal dispersion). Stretched-pulse design was employed, generating sub-200fs pulses[196]. Ref.[251] reported 163nJ pulse generation. Refs. [216, 220, 221, 224–227] also reported pulse generation using solid-state lasers mode-locked by GSAs. 94-fs tunable ($\sim$1.22-1.27μm) pulses have been achieved with a GSA mode-locked solid-state Cr:forsterite laser[226]. High-power ($\sim$1W) pulses have been demonstrated with a GSA mode-locked Nd:YVO$_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 fibers based ultrafast lasers working in a dissipative solution regime have been demonstrated for high average power pulses[255, 256], reaching megawatt 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, 130, 189, 263]) 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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