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## Sub 200 fs pulse generation from a graphene mode-locked fiber laser

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Ultrafast fiber lasers with broad bandwidth are in great demand for a variety of applications, such as spectroscopy, biomedical diagnosis, and optical communications. Sub 200 fs pulses are required for ultrafast spectroscopy with high temporal resolution. Graphene is an ideal ultrawide-band saturable absorber. We report the generation of 174 fs pulses from a graphene-based fiber laser.

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Ultrafast fiber lasers have many applications, ranging from basic research to materials processing and medicine.<sup>1,2</sup> Compared to other lasers, they have a simple and compact design, efficient heat dissipation, and high-quality pulse generation.<sup>3</sup> The latter is typically achieved through a nonlinear device called saturable absorber (SA).<sup>2-5</sup> The SA technology is currently dominated by semiconductor SA mirrors.<sup>2,3,6</sup> However, these require complex fabrication and packaging, and have limited bandwidth.<sup>2,3,6</sup> Single wall carbon nanotubes (SWNTs) and graphene are ideal SAs, having fast recovery times, low saturation intensity, low cost, and easy fabrication.<sup>7-22</sup> On one hand, broadband operation can be achieved using a distribution of tube diameters.<sup>7</sup> On the other hand, this is an intrinsic property of graphene, due to the gapless linear dispersion of Dirac electrons.<sup>8,9,18,23-25</sup>

Soliton-like mode-locking is the conventional method to generate ultrashort pulses in fiber lasers, down to several hundred femtoseconds.<sup>7,9-12,26</sup> Using this technique, ~400 fs pulses were reported for SWNT-based SAs<sup>17,19</sup> and graphene-based SAs.<sup>9,27</sup> The area theorem<sup>28</sup> for solitons states that the product of pulse energy ( $E$ ) and duration ( $\tau$ ) is fixed by the cavity dispersion and nonlinearity:<sup>29</sup>  $E \times \tau \propto \beta_2/n_2$ , with  $\beta_2$  the group velocity dispersion (GVD) coefficient and  $n_2$  the nonlinear refractive index of the propagating medium. Thus, for a given system design, there is a trade-off between  $E$  and  $\tau$ . One approach to mitigate these effects is to alternate segments of large normal (positive) and anomalous (negative) GVD fiber. In this way, the average pulse width in one cavity round trip can increase by an order of magnitude or more, significantly decreasing the intracavity average peak power, compared to soliton-like operation.<sup>29</sup> This is referred to as dispersion-management. It decreases nonlinear optical effects due to high intensity, reducing pulse duration.<sup>30</sup>

Here, we report a dispersion-managed fiber laser mode-locked by a graphene-based SA (GSA). The cavity comprises sections with positive and negative dispersion, provided by an erbium doped fiber (EDF) and a single mode fiber (SMF). We get ~174 fs pulses with 15.6 nm spectral width, much shorter than reported thus far for GSAs.<sup>9,27</sup>

Graphite flakes are exfoliated by mild ultrasonication with sodium deoxycholate surfactant.<sup>9,31</sup> A dispersion enriched with single layer graphene (SLG) and few layer graphene is then mixed with an aqueous solution of polyvinyl alcohol (PVA). After water evaporation, a graphene-PVA

composite is obtained.<sup>9,13</sup> Absorption measurements show a featureless spectrum from 500 to 2000 nm, Fig. 1. The UV peak is a signature of the van Hove singularity in the graphene density of states.<sup>32</sup> The PVA only shows significant absorption for shorter wavelengths.<sup>33</sup> Figure 2(a) plots a typical Raman spectrum of an exfoliated flake on Si/SiO<sub>2</sub>. Besides the G and 2D peaks, it has D, D', and D+D' bands. The G peak corresponds to the  $E_{2g}$  phonon at the Brillouin zone center.<sup>34</sup> The D peak is due to the breathing modes of  $sp^2$  rings and requires a defect for its activation by double resonance (DR).<sup>34,35</sup> The 2D peak is the second order of the D peak. This is a single band in SLG, whereas it splits in multilayer graphite,<sup>34</sup> reflecting the evolution of the band structure. The 2D peak is always seen, even when no D peak is present, since no defects are required for the activation of two phonons with the same momentum, one backscattering from the other. DR can also happen intravalley, i.e., connecting two points belonging to the same cone around  $\mathbf{K}$  or  $\mathbf{K}'$ . This gives rise to the D' peak. The 2D' is the second order of the D' peak. We do not assign the D and D' intensity in Fig. 2(a) to the presence of a large amount of structural defects, otherwise they would be much broader, and G, D' would merge.<sup>35</sup> We rather ascribe them to the edges of our submicrometer flakes.<sup>36</sup> Although 2D is broader than in pristine graphene, it is still a single Lorentzian. Thus, even if we have multilayers, these behave as a SLG ensemble. Figure 2(c) is the Raman spectrum of a reference PVA. The spectrum of the graphene-PVA composite can be seen as a superposition of Figs. 2(a) and 2(c). Therefore, embedding the flakes into PVA preserves their structure.

Power-dependent absorption is measured with a SWNT-based mode-locked fiber laser, delivering 450 fs pulses at 1558 nm with 38 MHz repetition rate.<sup>17</sup> The signal is di-

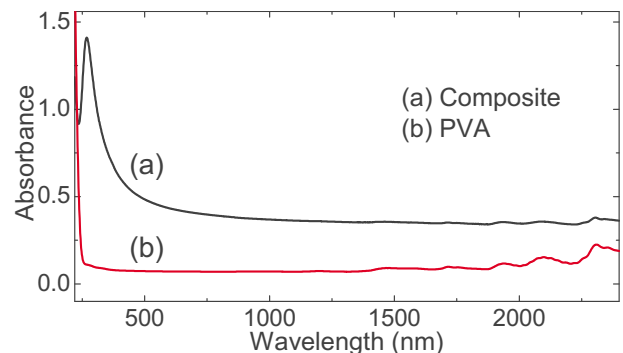


FIG. 1. (Color online) Absorption spectra of (a) composite and (b) PVA.

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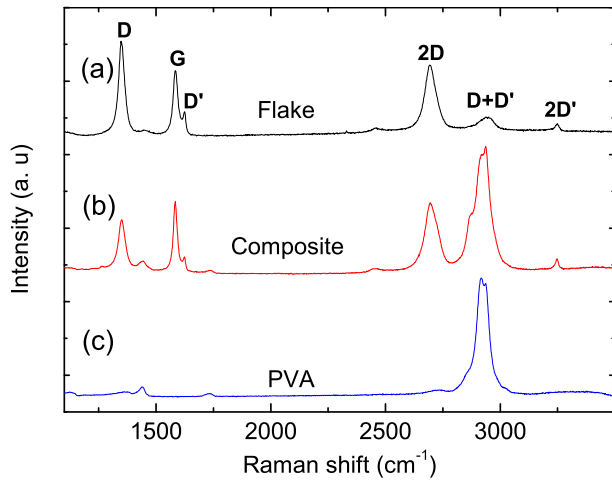
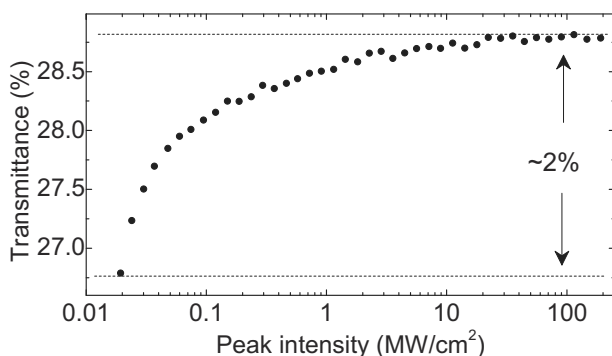


FIG. 2. (Color online) Raman spectra of (a) flake, (b) composite, and (c) PVA.

rected to the sample via an attenuator. A double-channel power meter measures both input and output power. The GSA is then placed between two fiber connectors. The output is monitored while the input power is varied, Fig. 3. The maximum peak intensity  $I_{\text{peak}} \sim 337 \text{ MW/cm}^2$  is reached for an average pump power  $\sim 5.15 \text{ mW}$ . The insertion loss is  $\sim 5 \text{ dB}$  (i.e.,  $\sim 28\%$  transmittance), acceptable for the single-pass gain of a fiber laser.<sup>7</sup> The transmittance increases by 2% when the SA saturates at  $I_{\text{peak}} \sim 100 \text{ MW/cm}^2$ . The modulation depth is 2%, comparable to SWNT-SAs.<sup>7,15,16</sup> Further improvement of modulation depth and nonsaturable loss should be possible by enriching the amount of SLG in the composite.<sup>18</sup>

Figure 4(a) is a schematic laser setup. We use 1.25 m highly doped EDF as gain medium, pumped by a 980 nm laser diode (LD) through a fused wavelength division multiplexer (WDM). The laser output is directed through the 20% port of a coupler. The total cavity length is  $\sim 7.6 \text{ m}$ . We use a EDF with  $\beta_2 = 48 \text{ ps}^2/\text{km}$ , estimated by inserting it into the SWNT-mode-locked soliton-like fiber laser described above, and measuring the shift ( $\Delta\lambda$ ) between sidebands and central wavelength of the soliton pulse spectrum.<sup>37</sup> The rest of the cavity consists of a combination of SMFs (Flexcor 1060 and SMF-28) with anomalous GVD, as indicated in the dispersion map of Fig. 4(b). The measured total intracavity GVD is  $\sim -0.052 \text{ ps}^2$ , typical for dispersion-managed cavities.<sup>29</sup> The laser operation is monitored with a second harmonic generation (SHG) autocorrelator and an oscilloscope. An optical spectrum analyzer probes the output.



Transmittance as a function of peak intensity.

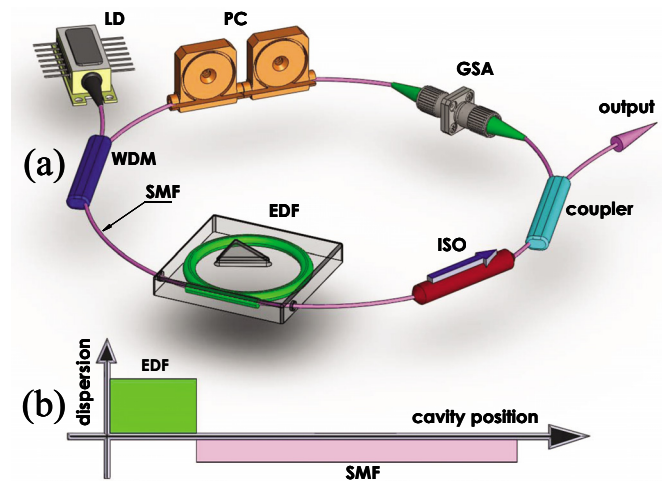


FIG. 4. (Color online) (a) Dispersion-managed fiber laser setup. Laser diode (LD), wavelength division multiplexer (WDM), single mode fiber (SMF), erbium-doped fiber (EDF), isolator (ISO), graphene saturable absorber (GSA), and polarization controller (PC). (b) Dispersion map. EDF and SMF provide positive and negative dispersions.

Continuous wave operation starts at  $\sim 18 \text{ mW}$  pump power, single-pulse mode-locking at  $\sim 25 \text{ mW}$ . The repetition rate is 27.4 MHz. The output power is  $\sim 1.2 \text{ mW}$  for  $\sim 28 \text{ mW}$  pump, with pulse energy  $\sim 44 \text{ pJ}$  and  $I_{\text{peak}} \sim 282 \text{ MW/cm}^2$ . Figure 5(a) shows that the sidebands, expected for soliton operation, are small, as a result of pulse stretching/compression.<sup>38</sup> The full width at half maximum bandwidth is  $\sim 15.6 \text{ nm}$ , larger than previous GSA-fiber lasers.<sup>9,18,27,39</sup> Figure 5(b) plots the SHG autocorrelation trace. For a  $\text{sech}^2$  profile, we get  $\sim 174 \text{ fs}$  pulses, much shorter than previous GSA-mode-locked lasers.<sup>9,18,27,39</sup> The time-bandwidth product is  $\sim 0.335$ , slightly higher than 0.315, expected in the case of transform-limited  $\text{sech}^2$  pulses, possibly due to uncompensated third-order dispersion distorting the intracavity pulse, thus limiting the minimum pulse width.<sup>38</sup> To get even shorter pulses, SAs with higher modulation depth are required.<sup>2</sup> Higher power could be feasible by evanescent field interaction with the GSAs.<sup>18,39</sup>

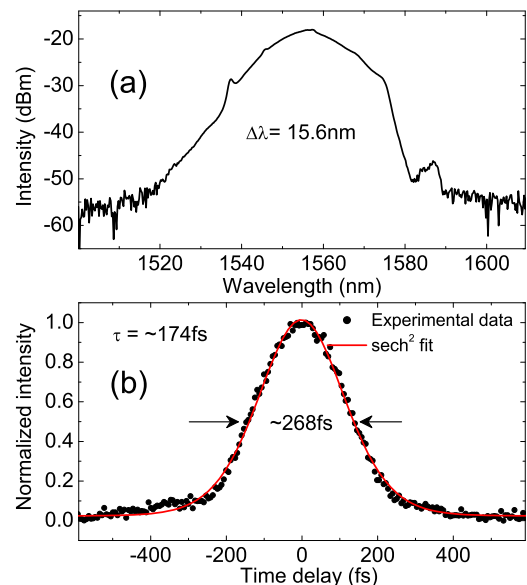


FIG. 5. (Color online) (a) Optical spectrum with bandwidth  $\Delta\lambda = 15.6 \text{ nm}$ . (b) Autocorrelation with  $\text{sech}^2$  fit.

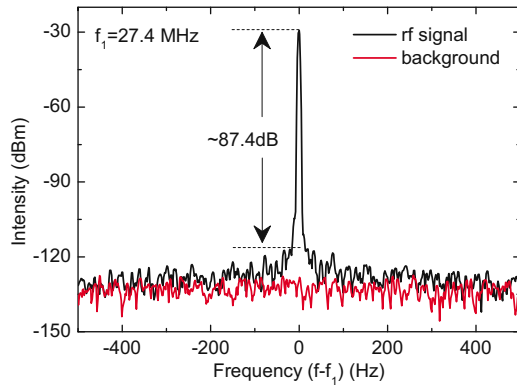


FIG. 6. (Color online) rf spectrum around the fundamental repetition rate  $f_1=27.4$  MHz with 10 Hz resolution.

Stability is important for applications. We measured the radio frequency (rf) spectrum using a photodetector connected to a rf spectrum analyzer around the fundamental repetition rate ( $f_1=27.4$  MHz), Fig. 6. The signal-to-noise ratio is  $>80$  dB, highlighting the low-amplitude fluctuations of our laser, thus stable mode-locking.<sup>40</sup>

In conclusion, we demonstrated a graphene-based SA mode-locker for generation of pulses shorter than 200 fs. The flexibility offered by the fiber laser design, along with the easy SA preparation, can lead to simple, low-cost, ultrafast light sources.

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