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### **Supplementary Information**

# Direct growth of graphitic nano-layer on optical fiber for

# ultra-fast laser application

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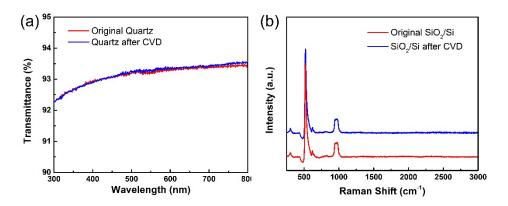
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#### Growth condition without the rf power

Neither graphene nor any carbonaceous materials were deposited on the optical fibers, quartz slide, or  $SiO_2/Si$  ( $SiO_2$  300 nm in thickness) wafer with the growth condition in the original file but without rf power for 40 min. Used the air as the baseline, the transmittance spectra were shown in Fig. S1(a), the transmittance of the quartz before and after thermal chemical vapor deposition (CVD) process was almost the same. Note that the little difference between the two lines probably caused by the stray light in the instrument.

The Raman scattering spectrum of the original  $SiO_2/Si$  wafer and substrates processed by thermal CVD were shown in Fig. S1(b). No G and 2D bands (signature of graphene) were observed around 1584 and 2695 cm<sup>-1</sup>, respectively. Indicating that no graphitized structure formed on the surface of the  $SiO_2/Si$  wafer.

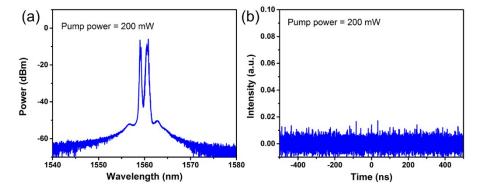


**Fig. S1.** Quartz slide and SiO<sub>2</sub>/Si wafer before and after thermal CVD process. (a) Transmittance spectrum; (b) Raman scattering spectrum.

Integrating the optical fiber processed by thermal CVD into the fiber laser. Fig. S2 shows the measured emission spectrum and the laser intensity as a function of time for a pump power of 200 mW, respectively. No matter how adjusting the intracavity polarization state, only continuous wave laser operation was observed and no pulsed operation was observed

from 50 mw to 200 mW. These results confirmed that no GNL was deposited on the fiber-

end and the mode-locked fiber laser was induced by the GNLSA.



**Fig. S2.** (a) Emission spectrum of the mode-locked fiber laser with an optical fiber processed by thermal CVD; (b) laser intensity as a function of time for a pump power of 200mW.

Kinetically, graphene is very difficult to be deposited with methane using thermal CVD at 700 °C. The activation energy for direct growth of graphene on a dielectric substrate was reported as 1.03 eV with a 50 W rf plasma versus 2.0-2.6 eV with thermal CVD.<sup>1</sup> Through linear fit of the Arrhenius plot of growth rate vs. substrate temperature, an activation energy was determined to be ~0.76 eV with a 200 W rf plasma in our PECVD apparatus.<sup>2</sup>

#### Morphology of VG grown on optical fiber-end

The high temperature PECVD growth results in a continuous flower-like three-dimensional network structure of VG, as shown in Fig. S3. The VG was growth with a mixture of  $CH_4$  (0.3 sccm) and  $H_2$  (9.7 sccm), and the reactor kept at 900 °C and rf power of 200 W for 45 min.

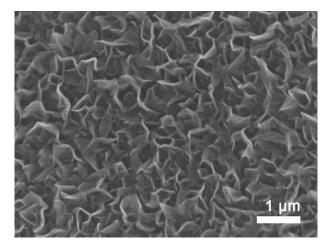


Fig. S3. SEM image of a fiber-end with directly grown VG.

### References

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- 2. D. Zhu, H. Pu, P. Lv, Z. Zhu, C. Yang, R. Zheng, Z. Wang, C. Liu, E. Hu and J. Zheng, *Carbon*, 2017, **120**, 274-280.