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Mode locking of ceramic Nd:yttrium aluminum garnet with graphene as a saturable absorber

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The mode-locking of a ceramic Nd:yttrium aluminum garnet (YAG) solid-state laser (SSL) with solution processed graphene as saturable absorber (SA) was demonstrated. Transform-limited pulses with duration of 4 ps centered at 1064 nm were generated for a nondispersion compensated Nd:YAG SSL. *Z*-scan studies revealed that the graphene SA has a saturation intensity of 0.87 M W cm^{-2} and a normalized modulation depth of 17.4%. Our results illustrate the potential of using graphene as a mode locker for SSLs. © *2010 American Institute of Physics*. [doi:10.1063/1.3292018]

Graphene is a two-dimensional material that contains a single layer of carbon atoms. Due to its electronic structure, graphene possesses many unique optical properties. It was found that a single layer of graphene absorbs about 2.3% of the light incident on it, and each additional layer adds another 2.3% to the overall absorption.[1] Graphene has no band gap,[2] indicating that its absorption is wavelength insensitive. In addition, pump probe studies on epitaxial graphene revealed that it has saturable absorption with an ultrafast recovery time.[3] The implication of these results is that graphene could be used as a wavelength insensitive saturable absorber (SA). Zhang *et al.*[4] and Bao *et al.*[5] have recently demonstrated mode-locking of an erbium-doped fiber laser at about 1550 nm using graphene as a SA. When graphene is exposed to high intensity illumination, states in both the conduction and valence bands near the Dirac point are filled by the electrons and holes, respectively. Pauli blocking prevents electrons in the valence bands to be further excited, achieving saturated absorption and high transparency.[5]

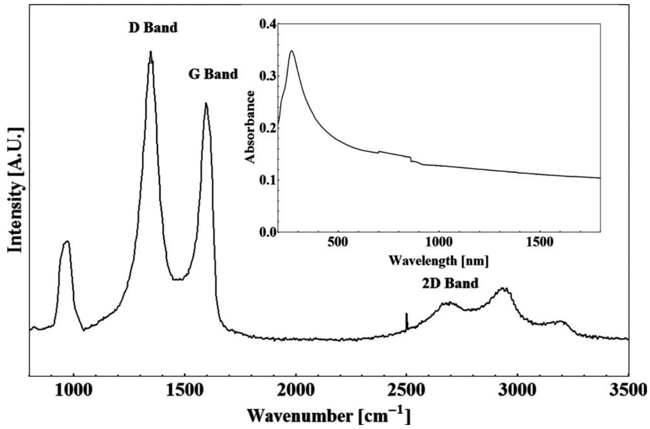

FIG. 1. Raman signature of graphene obtained from reducing graphene oxide. Inset: Linear absorption of graphene coated onto a piece of quartz substrate.

Mode locking of lasers is frequently demonstrated using the semiconductor saturable absorption mirror (SESAM).[6] However, fabrication of SESAM requires the usage of complicated deposition techniques. In addition, SESAMs have a narrow operating wavelength range[7] and their modulation depths are difficult to modify. Recently, single wall carbon nanotubes (SWCNTs) have also been used as a mode locker.[8] SWCNT mode lockers typically contain nanotubes of different diameters and both the semiconducting and metallic nanotubes. Metallic nanotubes and nanotubes with unsuitable diameters do not take part in the saturable absorption process. Moreover, SWCNTs tend to cluster, which end up as scattering sites leading to additional losses.[8] These problems can be sidestepped using graphene as a SA. Graphene can be solution processed[9] and coated onto different types of substrates. The number of layers of graphene deposited onto a piece of substrate can be controlled, thus varying the amount of light absorbed and the modulation depth. Graphene being a zero band gap material can function as a SA over a wide range of wavelengths without having to tune any material parameters. Finally, graphene does not form bundles like SWCNTs. In this letter the mode locking of a diode pumped ceramic Nd: YAG laser with graphene as a SA is demonstrated.

Graphene on a quartz substrate was obtained from solution graphene oxide via a reduction process using hydrazine, as reported in Ref. 9. Raman spectroscopy was carried out on

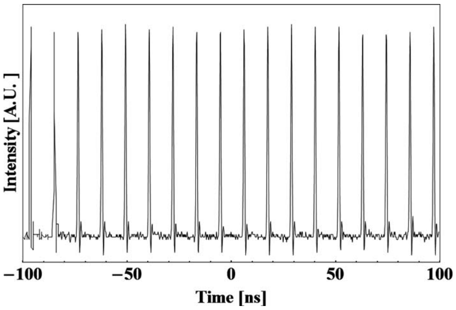

FIG. 2. cw mode locked pulse train.

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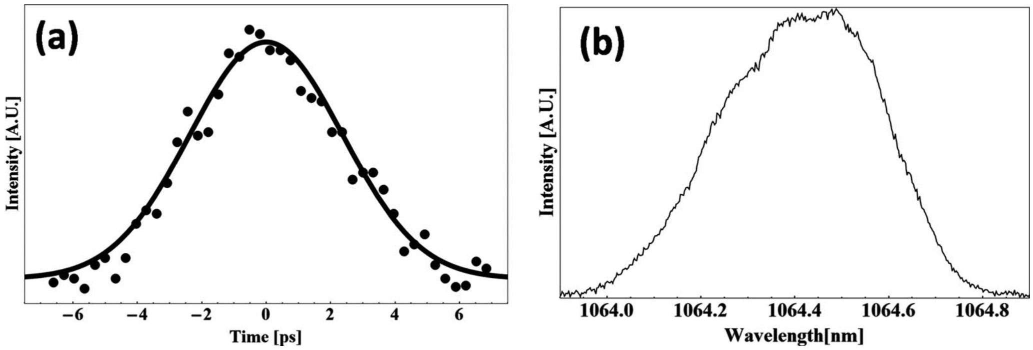

FIG. 3. (a) Autocorrelation trace of the cw mode locked pulses. Closed circles are data points and the solid line is a Gaussian fit. (b) The corresponding optical spectrum.

the as-fabricated samples and the Raman trace is shown in Fig. 1. The inset of Fig. 1 shows the linear absorption of a graphene sample. Due to the zero band gap of graphene, the linear absorption extends from the visible to the mid-IR region and shows little wavelength dependence. As such graphene can potentially serve as a SA for different lasers whose wavelengths are in this region. The ceramic Nd:YAG laser has a cavity design similar to the one shown in Ref. 10, but with the SESAM replaced by a high reflection mirror (HRM). A commercial $4\times3\times3$ mm^3 ceramic Nd:YAG crystal with an Nd^{3+} doping concentration of 0.5 at. % was used. The graphene SA was inserted between mirrors M3 and the HRM. To achieve cw mode locking, the distance between the SA and HRM was varied along the cavity axis. Figure 2 shows the mode locked pulse train obtained. The pulse to pulse separation corresponds to the cavity fundamental repetition rate of 88 MHz. The shortest autocorrelation trace measured had a full width at half maximum (FWHM) of about 5.6 ps as shown in Fig. 3(a). This implied that the mode locked pulse duration was about 4 ps assuming a Gaussian pulse profile. The FWHM of the optical spectrum shown in the inset of Fig. 3(b) is 0.42 nm. Therefore, the time-bandwidth product of the pulses is 0.44 indicating that it is transform limited. The laser emission has an average power of about 100mW.

The saturable absorption property of the graphene sample was measured using z-scan. The laser source used for z-scan was a homebuilt mode locked oscillator with a repetition rate of 88 MHz and pulse width of 500 fs operating at about 1040 nm. An average power of 1.34 mw is incident and focused to a spot size of 11 μm on the graphene sample at the beam waist. The data obtained from the z-scan was then fitted according to

$$T = A \exp\left(\frac{-B}{1+\frac{I}{I_{\text{sat}}}}\right), \qquad (1)$$

where T is the transmission, A is a normalizing constant, B is the linear limit of saturable absorption or the normalized modulation depth, I is the intensity and I_{sat} is the saturation intensity. Fitting of the z-scan data resulted in Fig. 4, which gives the values of B and I_{sat} as 17.4% and 0.87 M W cm^{-2}, respectively. These results compare well with those of Bao *et al.*[5] When the same experiment was repeated by replacing the HRM with a SESAM with 1% modulation depth, pulses of about 25 ps with an average output power of 1 W was obtained. The lower average output power under graphene mode locking was attributed to the large insertion loss of the current graphene mode locker. The shorter pulse width obtained could be understood from the large normalized modulation depth of the graphene. A larger normalized modulation depth would result in shorter mode locked pulses.[11]

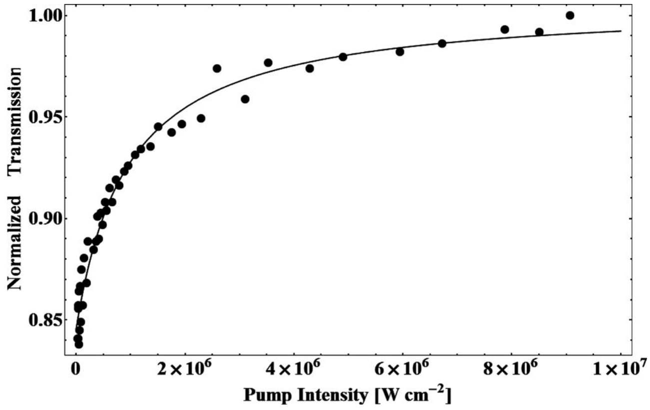

FIG. 4. Transmission of graphene with increasing pump intensity. Closed circles are data points while the solid line is the fitting obtained from Eq. (1).

In conclusion, mode locking of a diode pumped ceramic Nd:YAG laser with graphene as the SA was demonstrated. Transform-limited pulses with a pulse width of 4 ps and an average output power of about 100 mW were produced. z-scan of the graphene sample revealed that the graphene SA has a saturation intensity of 0.87 M W cm^{-2} and a normalized modulation depth of 17.4%. Our results illustrate the potential of using graphene as a mode locker for SSLs. Graphene can be used as a SA for lasers operating in the visible to the mid-IR.

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