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2.6 ps pulse from passively mode-locking $Nd:Gd_{0.64}Y_{0.36}VO_4$ laser based on graphene oxide

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1. Introduction

Recently, it has been shown that the laser pulses with duration of picoseconds provide more excellent quality in micromechanical process than those of femtoseconds, which were considered as the best solution in the past. The plasma effect is avoided and the thermal damage is reduced on digging, incision and milling some hard materials using laser with picosecond pulses. Modelocking technology provides a method to obtain laser pulses from femtosecond to nanosecond. Compared with the active device that needs complex extracavity electronic driver, saturable absorber (SA) exhibits advantages of fast recovery time, stability and compactness. Till now, many kinds of semiconductor materials, e.g. GaAs; doped crystals, e.g. Cr4+:YAG; and color-center crystal, e.g. LiF:F⁻², have been studied in passively Q-switching and modelocking lasers [1–3]. Semiconductor saturable absorber mirror (SESAM) is the most popular and mature device that used in generating mode-locking laser in commercial. However, the energy gap of guantum-well material and the critical lattice-match to the substrate limit its absorption wavelength range [4,5]. In recent years,

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ABSTRACT

A diode-pumped passively mode-locking Nd:Gd_{0.64}Y_{0.36}VO₄ laser was accomplished by employing graphene oxide as saturable absorber and output coupler simultaneously, for which the graphene oxide was deposited onto the output mirror. Owing to the broadband gain spectrum of the Nd:Gd_{0.64}Y_{0.36}VO₄ crystal, cw mode-locking pulses with duration of ~2.6 ps centered at 1064.30 nm were obtained. The average output power was 1.3 W and the corresponded peak power was 8.8 kW.

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single-wall carbon nanotubes (SWCNT) and graphene have been proved to be promising materials as SAs [6-8]. The energy gap of SWCNT depends on the nanotube's diameter and chirality. The nanotubes with unsuitable diameters and the metallic nanotubes do not take part in the absorption process for a certain wavelength. Therefore, a broadband SWCNT SA may introduce large insertion loss in the cavity. For graphene, the band gap structure of Dirac point makes the saturable absorption independent of the incident wavelength. A graphene SA can be fabricated without any tailoring of the material. The electrons of valence band absorb the incident photons and excite into the conduction band and thermalize and cool down [7–9]. At high optical intensity, the electrons and holes fill the states near the edge of conduction and valence band, and further absorption is blocked. That means, saturation and the incident photons pass through without loss. The critical issue for its wide application is the solubility, which allows ease of wafer scale deposition. Compared with graphene, graphene oxide (GO) is strongly hydrophilic and water soluble, owing to oxygen functional groups [10–16]. The oxygen functional groups destroy the linear dispersion of the Dirac electrons and make GO insulating. But GO sheet contains not only insulating sp³ hybridized carbon matrix but also sp² hybridized carbon domains, which determines the saturable absorption property. The relaxation time of GO is characterized by a two-fold carrier dynamics process, which consists of a fast time of ~330 fs associated with intraband carrier-carrier scattering and a slow one of ~4.85 ps that reflects interband carrier-phonon







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scattering [13]. GO is attractive in mode-locking laser without the necessity of reduction to graphen (rGO). By now, GO has been studied in diode-pumped solid-state Nd:YAG [16], Nd:YVO₄ [17,18] and Nd:GdVO₄ [19,20] lasers as well as fiber lasers [21–25].

Not only the properties of SA but also those of gain medium should be considered in mode-locking lasers. A new Nd-doped crystal Nd: $Gd_xY_{1-x}VO_4$, which can be grown by fractionally replacing the Y-ions in Nd:YVO₄ by Gd-ions or by fractionally replacing the Gd-ions in Nd:GdVO4 by Y-ions, has emerged as promising laser crystal [26,27]. The mixed crystal not only inherits the general properties of Nd:GdVO₄ and Nd:YVO₄ but also has some new features. The properties such as fluorescence bandwidth, stimulated emission cross-section, upper-state lifetime and thermal conductivity can be adjusted by tuning the Gd/Y composition ratio. This characteristic has attracted the people with much interest in investigating properties with different *x* value. It has been demonstrated that Nd:Gd_{0.64}Y_{0.36}VO₄ has the smallest stimulated emission cross-section and the longest upper-state lifetime $(7.5 \times 10^{-19} \text{ cm}^2, 138 \,\mu\text{s})$ among the series including Nd:YVO₄ $(11 \times 10^{-19} \text{cm}^2, \sim 98 \,\mu\text{s})$ and Nd:GdVO₄ $(7.9 \times 10^{-19} \text{cm}^2, 120 \,\mu\text{s})$ [26-28]. Small stimulated emission cross-section combined with long upper-state lifetime implies high energy storing capacity. The fluorescence bandwidth of Nd:Gd_{0.64}Y_{0.36}VO₄ is broader than that of either Nd:YVO₄ or Nd:GdVO₄ [29]. The reported gain bandwidth is \sim 1.3 nm at 1064 nm [30]. That means, pulses as short as sub-picosecond can be supported in mode-locking operation. In this paper, we report obtaining a diode-pumped passively modelocking Nd:Gd_{0.64}Y_{0.36}VO₄ laser using GO. ~2.6 ps pulses with repetition rate of 57 MHz. The corresponding pulse energy and peak power were 23 nJ and 8.8 kW, respectively.

2. Graphene oxide saturable absorber mirror (GO SAM)

The GO used in our experiment was prepared via the oxidation of graphite with the method of modified Hummers. The as-prepared GO was sonicated to disperse the GO sheets and dialysed and remove the residual ions. The final GO dispersion was by free drying to obtain GO powder. The AFM analysis shows that the thickness of GO sheets is ~ 2 nm, which means it is a single layer, as shown in Fig. 1. The diameter is in the range of 2-3 µm. About 10 milligrams of GO powder was poured into 10 ml 0.1% sodium dodecyl sulfate (SDS) aqueous solution. Then the GO aqueous solution was ultrasonically agitated for 10 h. After the ultrasonic process, the dispersed GO solution was centrifuged to remove large graphene oxide clusters. The upper portion of the centrifuged solution was decanted and diluted. A piece of K9 glass coated with reflectivity of 95% at 1064 nm was used as the substrate. The GO aqueous solution was dripped onto the substrate and dried in a vacuum oven at 30€ for 48 h. With this structure, the GO SAM served output coupler as well as SA. Fig. 2 shows the Raman spectrum of GO obtained with



Fig. 2. Raman spectrum and pump-probe trace (insert) of GO.

a 532 nm laser excitation. The 1G (1619 cm^{-1}) peak corresponds to the first scattering of E_{2g} photons at Brillouin zone center. The 1D (1377 cm^{-1}) peak originates from the attachment of hydroxyl and epoxide groups on the carbon basal plane. The weak and broad 2D (2744 cm^{-1}) peak indicates that there were rare graphene. The relative intensity ratio I_G/I_D indicates the number of sp³ hybridized carbon atoms to that of sp². The ratio is ≈ 1.3 . This value is comparable to that in [16], but larger than that in [11]. The nonlinear response of the GO was probe measured with an ultrafast laser system (Tsunami femtosecond Laser, Spectra-Physics Com.), which provided 100 fs laser pulses at 1 kHz repetition rate at 1060 nm. The Fig. 2(insert) shows the pump–probe trace with exponential function fit. The fast relaxation time is about 760 fs and the slow one is about 4.25 ps.

3. Experiment and discussion

The laser configuration is shown in Fig. 3. The pumping source was a fiber coupled diode laser emitting at 880 nm. The fiber had a core diameter of 400 μ m and numerical aperture of 0.22. The pump beam was imaged to 670 μ m in diameter inside the gain medium by a coupling system. The gain medium was an a-cut Nd:Gd_{0.64}Y_{0.36}VO₄ with concentration of 0.3 at.% and dimensions of $3 \times 3 \times 6$ mm³. The crystal was indium foil wrapped and mounted in a copper heat sink, which was cooled by water kept at about 20€. The length of the cavity was about 2630 mm with 660 mm from M₁ to M₂, 800 mm from M₂ to M₃, 890 mm from M₃ to M₄ and 280 mm from M₄ to the GO SAM. The radii of M₁ and M₄ are 200 mm and 500 mm, respectively. M₂ and M₃ are plane mirrors. All the mirrors are antireflection coated at 880 nm and high reflection coated at 1064 nm. The calculated cavity mode on the GO SAM was around 20 μ m \times 27 μ m.

The output behavior of the laser was investigated with careful adjustment. The incident pump power means the power measured



Fig. 3. Schematic of the experiment setup.

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