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99 fs Nd:Glass Laser Mode-Locked with Carbon Nanotube Saturable Absorber Mirror

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The mode-locked operation of Nd:silicate glass with both single-walled carbon nanotubes (SWCNTs-SAM) and semiconductor saturable absorber mirrors (SESAMs) has been investigated. SWCNT-SAM mode-locking has been optimized in Nd-doped bulk lasers for the first time, yielding 99-fs pulses at 1070 nm. For comparison, by using a SESAM instead of SWCNT-SAM, pulses as short as 87 fs and a tuning range of 30 nm were achieved. Related to the applied 200-mW single-mode laser diode as pump source the lasers were characterized by high efficiency and low threshold. The optimized low-power pump setup yielded a remarkable slope efficiency of 46.5% in the cw regime.

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Compact diode-pumped femtosecond lasers operating at a wavelength of ~1 µm are cost-effective solutions for many applications that require only a few milliwatts of average power, such as diagnostic techniques in biophotonics, THz generation and detection, and seeding of ultrafast amplifiers. With the low laser threshold allowed by the four-level nature of the material, Nd:glass offers a valid alternative to ytterbium lasers for low pump powers, as single-mode diode-pumped ~100-fs oscillators at 1 µm have recently demonstrated.[1,2] Presently, the most expensive and critical component in these laser resonators is the semiconductor saturable absorber mirror (SESAM), needed to start-up and stabilize the soliton mode-locking. Recently, single-walled carbon nanotube saturable absorbers (SWCNT-SAs) have attracted significant attention as potential substitutes of SESAMs for the mode-locking of ultrafast lasers. Besides some intrinsic advantages (broad spectral operating wavelength and easy tunability, possibility to realize both transmission- and reflection-type absorbers on a wide variety of substrates, very fast recovery time, low saturation fluence and absence of two-photon absorption),[3,4] one of the most favorable SWCNT characteristics is the reduced cost and complexity of manufacturing apparatus and techniques, if compared with those of SESAM.

Mode-locking with SWCNT-SAs was first introduced and implemented in fiber lasers.[5] Because of their high single-pass gain, fiber lasers tolerate much larger amounts of non saturable losses than bulk lasers, relaxing the design of the saturable absorber. Nevertheless, some refinements in the synthesis techniques and a careful control of the carbon nanotube dispersion and deposition processes made SWCNT-SAs with the combination of low linear loss, small modulation depth and fast recovery time available. In particular, a low linear loss is required for the passive mode-locking of femtosecond bulk lasers.[6]

SWCNT-SAs allowing passive mode-locking with pulse durations of ~100 fs and even shorter have been reported at 1.57 µm in Er/Yb:glass[7] and Cr:YAG,[6] near 1.25 µm in Cr:forsterite[8] and in the 1 µm range with Yb:KYW[9] and Yb:KLuW.[10] Apparently, SWCNT-SA mode-locking of Nd:glass has been largely overlooked: only a preliminary result of ≈200 fs pulse duration is mentioned in the literature.[7] In this paper, we report, for the first time to the best of our knowledge, sub-100 fs mode-locking of Nd:glass with SWCNT-SA mirror (SWCNT-SAM) in a remarkably efficient low-pump-power oscillator. For comparison, we also discuss the results obtained with the same experimental setup employing a commercial SESAM as a mode-locker.

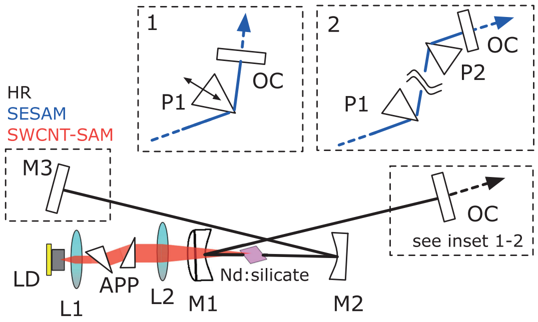

Fig. 1. Resonator layout. LD: pump laser diode; L1: aspheric lens (4.5-mm focal); APP: anamorphic prism pair; L2: spherical singlet lens (50 mm focal); M1: concave mirror, 50 mm curvature, high reflectivity (HR) at 1000–1100 nm, high transmissivity at 800–810 nm; M2: concave mirror, 100 mm curvature, HR; M3: flat mirror: HR, SESAM or SWCNT-SAM depending on the experiment; P1, P2: FS prisms; OC: output coupler, 30′ wedge.

The resonator layout is shown in Fig. 1, including all its variants for cw and mode-locked operation. The pump source was a single-mode 200 mW laser diode (Intense Ltd.), emitting at 805 nm with a narrow 0.05 nm linewidth. With respect to our previous investigation,[2] anamorphic prisms were employed to circularize the elliptical pump beam and optimize its overlap with the resonant mode inside the active medium. The maximum power delivered to the active medium was 156 mW. The broad absorption spectrum of the 3% doped Nd:silicate glass (Schott LG680), centered at 805 nm, yields about 90% pump absorption in the 4-mm-thick Brewster-positioned glass plate.

The pump beam was characterized with a charge coupled device (CCD) camera scanning along the propagation axis near the focal plane, yielding waist radii $w_{px} \times w_{py} = 14.1 \times 14.4\,\mu m^2$ in air and beam quality parameters $M_x^2 = 1.0$ and $M_y^2 = 1.1$. The resonator beam waist radius

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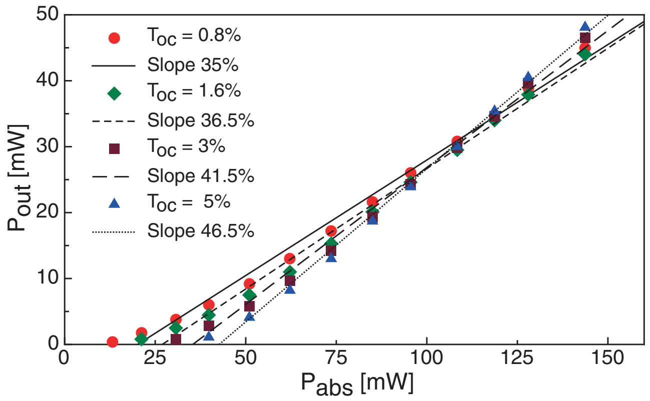

Fig. 2. Output power (P_{out}) as a function of the absorbed pump power (P_{abs}) of the Nd:glass laser in the cw regime.

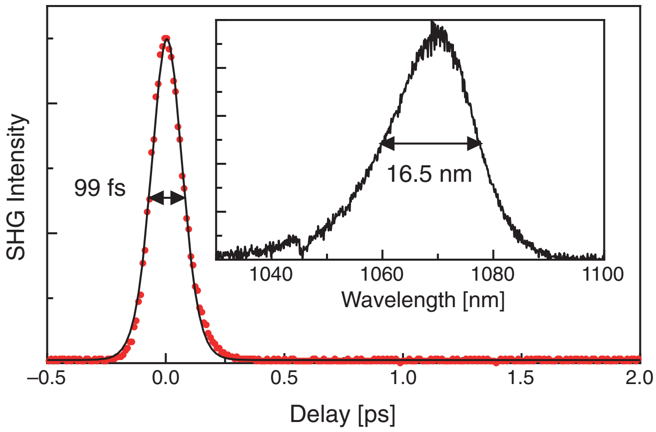

Fig. 3. Autocorrelation trace and spectrum of the mode-locked Nd:glass laser obtained with SWCNT-SAM.

was calculated to be $\approx 15-20\,\mu$m within the stability range.

A remarkable 46.5% slope efficiency was obtained in the cw regime, the highest reported for diode-pumped Nd:glass lasers to the best of our knowledge. The output power was as high as 48.1 mW for the optimum 5% output coupler (Fig. 2). The same resonator setup was used to perform the Findlay–Clay analysis to determine the total loss (saturable + non saturable) introduced by the SWCNT-SAM employed in mode-locking experiments, which turned out to be $\approx$1.5%. This value is in rather good agreement with the nonlinear reflectivity measurements of the same sample, which gave a saturable loss of 0.21% and a non saturable loss of 0.7%. The saturation fluence was measured to be $5\,\mu\mathrm{J/cm}^2$, whereas the nonlinear response was described as a biexponential response with fast ($<$150 fs) and slow ($<$1 ps) components. To fabricate the SWCNT-SAM, arc-discharge made SWCNTs were mixed with poly(methyl methacrylate) (PMMA) in dichlorobenzene (DCB) and the SWCNT/PMMA film was subsequently spin-coated onto a dielectric mirror designed for broadband lasers near 1 μm. The detailed fabrication procedure is described elsewhere.[13]

For investigation of the passive mode-locking regime, we employed a 0.4% output coupler and the mirror M3 was replaced by the SWCNT-SAM. A couple of fused silica (FS) prisms were used to obtain the net negative intracavity second-order dispersion required for soliton mode-locking.

We varied the cavity mode dimension on the SWCNT film by changing the length of the corresponding cavity arm M2–M3 and consequently adjusting the M1–M2 separation to manage the resonator stability. Best results were obtained with the following mirror separations: M1–M2 $\approx$ 84 mm, M2–SWCNT-SAM $\approx$ 400 mm, and M1–OC $\approx$ 800 mm (P1–P2 separation $\approx$ 650 mm), yielding a cavity mode radius on the saturable absorber $w_a \approx 80\,\mu$m. Stable 99 fs long pulses and 10 mW average output power with 16.5-nm-wide spectrum centered around 1070 nm (time-bandwidth product $\approx$ 0.43) were obtained (see Fig. 3). Under these conditions, the mode-locking regime was not self-starting but could be sustained for several minutes once it was built-up through a small perturbation. No local damage occurred to the SWCNT absorber layer: in any case, if interrupted, mode-locking could be restored by gently shaking a mirror mount or moving a prism. Accounting for the good sample homogeneity, pulse duration, mode-locked pulse train stability and average output power were not significantly influenced by the spot position on the SWCNT-SAM. Reducing the distance between the prisms (hence, reducing the amount of net negative dispersion), we observed even wider spectra (up to about 20 nm), but the mode-locking regime became very unstable and cw components in the blue tail of the spectrum around the fluorescence peak at 1060 nm appeared systematically, probably suggesting a modulation depth of the absorber insufficient to stabilize the mode-locking under these conditions as well as to guarantee at least a short-term stability with pulses as in Fig. 3. Indeed, when the laser resonator was not optimized to generate the shortest pulses with duration approaching 100 fs, i.e., the pulse spectrum was narrower than $\sim$10 nm, the mode-locking state lasted much longer: this is a further indication that mode-locking collapse is related to gain reduction due to broad-spectrum oscillation, and should be counterbalanced by an increase of the saturable loss.

We also investigated a cavity arrangement employing a single FS prism for dispersion control.[1,2] The mirror separations were as follows: M1–M2 $\approx$ 84 mm, M2–SWCNT-SAM $\approx$ 430 mm, M1–P $\approx$ 720 mm, and P–OC $\approx$ 40 mm. According to ABCD modeling as outlined in ref. 11, the separation between real and virtual prism in this setup was about 60 cm. With the straightforward central wavelength selection allowed by the single-prism setup, red-shift of the pulse spectrum and consequent suppression of cw residual components was easier. Spectra as broad as 13 nm centered around 1070 nm with 143-fs pulse duration (time-bandwidth product $\approx$ 0.49) were obtained. A central wavelength tunability of 12 nm, limited to the longer wavelengths (1068–1080 nm) of the Nd:silicate fluorescence spectrum, was obtained. The longer pulses are due to transverse wavelength dispersion in the gain medium.[2,11]

Subsequently, we replaced the SWCNT-SAM with a SESAM specified at 1% total losses (0.4% non saturable loss), $70\,\mu\mathrm{J/cm}^2$ saturation fluence and 1 ps recovery time (Batop GmbH). At first, we tested the cavity setup employing two FS prisms. In order to obtain stable mode-locking, it was necessary to reduce significantly the resonant mode size on the saturable absorber, as expected due to the typically

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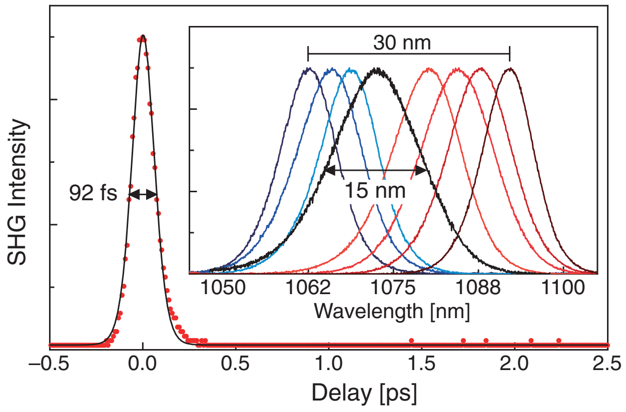

Fig. 4. Shortest pulse autocorrelation (corresponding spectrum width: 15 nm full width half maximum) obtained with SESAM in single-prism setup. Inset: central wavelength tunability in the mode-locked regime.

higher SESAM saturation fluence with respect to that of SWCNT-SAs.[9,10,12)] Keeping the separations in the OC cavity arm equal to those of previous double-prism cavity experiments with SWCNT-SAM, we reduced the M2–SESAM distance to $\approx$175 mm and adjusted the M1–M2 separation to $\approx$97 mm for cavity stability. Under these conditions, the ABCD model yields $w_a \approx 30\,\mu\text{m}$. The required reduction of mode area on the SESAM by a factor $\approx$7 compared with that on the SWCNT-SAM is an indirect confirmation of the saturation fluence ratio between the two saturable absorbers. Finely adjusting the cavity mirror alignment and tuning the dispersion, we obtained stable and self-starting 87 fs pulses with an average output power of 15 mW. The corresponding 14-nm-wide spectrum, centered around 1070 nm, yielded a time-bandwidth product $\approx$0.32 close to the Fourier limit for sech^2-shaped pulses.

SESAM was also employed in the single-prism cavity arrangement. Pulses as short as 92 fs (15-nm-wide spectrum) were generated at 1073 nm. A remarkable 30 nm tuning range (from 1062 to 1092 nm) with output powers exceeding 22 mW, at least 9-nm-wide spectra, and pulses shorter than 120 fs for any operating central output wavelength were obtained. In Fig. 4, both the autocorrelation trace of the shortest pulses and the spectral tunability are shown.

In summary, we have demonstrated the first sub-100 fs Nd:glass laser mode-locked by a SWCNT-SAM and have compared its performance with that achieved with a commercial SESAM. Furthermore, owing to the improved beam shaping of the single-mode laser diode yielding circular profile optimally matched to the Nd:glass resonator mode, remarkable results in terms of cw operation and SESAM mode-locking have been achieved. The 30 nm tuning range is the broadest ever reported for a femtosecond Nd:glass laser. Higher efficiency, shorter pulses with better mode-locking stability comparable to that allowed by SESAMs are expected with SWCNT-SAM as well, through improvement of the ratio between saturable and non saturable losses, allowing larger nonlinear modulation. The relatively small saturation fluence of the SWCNT-SAM ≈ 5–$10\,\mu\text{J/cm}^2$, according to the measurements reported in ref. 9, is a favourable parameter, relaxing the focusing requirements on the saturable absorber, simplifying the resonator design, and making the operation of SWCNT-SAM safe and highly reliable, especially for this class of low-pump-power femtosecond sources.

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