Diode-pumped Nd:BaY$_2$F$_8$ picosecond laser mode-locked with carbon nanotube saturable absorbers

Antonio Agnesi,[1,*] Luca Carrà,[1] Federico Pirzio,[1] Giancarlo Reali,[1] Alessandra Toncelli,[2] Mauro Tonelli,[2] Sun Young Choi,[3] Fabian Rotermund,[3] Uwe Griebner,[4] and Valentin Petrov[4]

[1]*Dipartimento di Elettronica dell'Università di Pavia, Via Ferrata 1, 27100 Pavia, Italy*
[2]*CNR NEST (Consiglio Nazionale delle Ricerche-National Enterprise for nanoScience and nanoTechnology) and Dipartimento di Fisica dell'Università di Pisa, Largo B. Pontecorvo 3, 56127 Pisa, Italy*
[3]*Division of Energy Systems Research, Ajou University, 443-749 Suwon, South Korea*
[4]*Max-Born-Institute for Nonlinear Optics and Ultrafast Spectroscopy, 2A Max-Born-Str., D-12489 Berlin, Germany*
**Corresponding author: antonio.agnesi@unipv.it*

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Picosecond pulse generation near 1-μm wavelength has been achieved with a Nd:BaY$_2$F$_8$ (Nd:BaYF) laser mode-locked using a single-walled carbon nanotube saturable absorber (SWCNT-SA). The laser was operated at its main 1049-nm transition, generating 8.5-ps pulses with ≈70-mW output power for ≈570-mW absorbed pump power. This is the first demonstration of cw mode-locking in the picosecond regime with Nd-doped crystals and SWCNT-SAs. The requirements on the SWCNT-SA for successful mode-locking in relatively narrow-band neodymium lasers are reviewed and their implications are discussed.

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1. INTRODUCTION

SWCNT-SAs have been successfully employed in a variety of solid-state and fiber lasers for generation of ultrashort pulses with passive mode-locking in a broad range of wavelengths extending from 1 to 2 μm [1,2]. This new class of saturable absorber (SA) devices has attracted great interest owing to the much easier and cost-effective fabrication process with respect to the well-established semiconductor saturable absorber mirror (SESAM) technology. Indeed, preparing a SWCNT-SA for a specific laser transition requires only choosing the SWCNT with the suitable diameter and chirality among those readily available commercially. For mode-locking of solid-state lasers, SWCNTs are usually deposited on glass substrates or dielectric mirrors (SWCNT-SAMs) as SWCNT/polymer dispersions.

To date, ultrafast solid-state lasers mode-locked with SWCNT-SAs operating in the femtosecond regime have been demonstrated in wide-bandwidth laser systems such as Nd:glasses [3]; Yb:KYW [4] and Yb:KLuW [5]; Er/Yb:glass [3]; Cr^{4+}:forsterite [6]; and Cr^{4+}:YAG [7]. Some of these reports also demonstrated chirped picosecond pulse generation in resonators without additional dispersion compensation.

Apparently, lasers based on Nd^{3+}-doped crystals have not been investigated for cw mode-locking with SWCNT-SAs: the only report we are aware of is of a flashlamp-pumped Q-switched mode-locked 1.34-μm Nd:GdVO$_4$ laser [8].

Employing the SWCNT-SA devices previously used for mode-locking Yb:KYW [4] and Yb:KLuW lasers [5] we decided to investigate their performance with several active materials, specifically cw diode-pumped Nd:glass: Nd:BaY$_2$F$_8$ (Nd:BaYF), Nd:YAG, and Nd:YVO$_4$.

Our choice of Nd:glass was motivated by the fact that it had already been successfully mode-locked in the femtosecond regime (we readily achieved results comparable to or better than those of [3]), while lasers with Nd^{3+}-doped crystals might be more difficult to mode-lock because of the larger emission cross section and the narrower fluorescence bandwidth. Indeed among these crystals, we were able to achieve cw mode-locking only using Nd:BaYF, whose spectroscopic features are intermediate between those of Nd:glass and of Nd:YAG and Nd:YVO$_4$. For comparison, Table 1 summarizes the main properties of neodymium materials investigated in this research. According to the generally accepted condition for mode-locking starting [9,10] in large emission-cross-section and narrow-bandwidth laser materials, both gain saturation and the fluctuation pulse duration of the free-running field contribute to make mode-locking more difficult. Therefore, SWCNT-SAs with larger modulation depth and smaller non-saturable loss should be employed in this case.

2. EXPERIMENTS

The laser cavity was an astigmatically compensated X-folded resonator shown in Fig. 1. The pump diode was a 100×1 μm^2 broad-area emitter with a maximum output power of 1 W at 805 nm. It was collimated by an 8-mm focal aspheric lens L1, expanded by a factor of 15 in the

Table 1. Emission Cross Sections and Gain Bandwidths of Neodymium-Doped Materials Discussed Here

	Nd:glass (phosphate Schott LG760)	Nd:BaYF (E//b)	Nd:YAG	$Nd:YVO_4$ (E//c)
Emission cross section [$\times 10^{20}$ cm^2]	4.5	8	33	160
Fluorescence bandwidth FWHM [nm]	24.3	2.8	0.6	0.8

slow-axis direction with a cylindrical lens telescope (C1, C2), and eventually focused in the laser crystal by a 75-mm focal achromat L2. The pump spot size measured with a CCD camera was $\approx 62\ \mu m \times 28\ \mu m$ along the (horizontal) slow- and fast-axis, respectively. The M^2 parameters for both axes were 19 and 1.5, respectively.

The 4-mm long, 1.8%-doped Nd:BaYF crystal is the same used in our previous experiments with SESAM mode-locking [11,12]. The Nd:BaYF crystal was Brewster-cut with parallel end-faces for propagation along the z axis with polarization parallel to the y-axis (which is both an optical and crystallographic axis).

The laser crystal did not require any active cooling for the present low-power diode-pumping.

The waist radius of the X-shaped cavity was measured to be $40\ \mu m \times 30\ \mu m$. The distances between cavity mirrors were: M1-M2=115 mm, M2-M3=265 mm, and M1-OC=380 mm. An output coupler (OC) with power transmissivity 0.8% was chosen, achieving up to 150 mW with 26% slope efficiency in cw operation.

For mode-locking experiments, the flat HR-mirror M3 was replaced by a flat SWCNT-SA mirror (SWCNT-SAM) with measured saturable loss of 0.21%, non-saturable loss of $\approx 0.7\%$, $F_a \approx 5\ \mu J/cm^2$ saturation fluence, and with biexponential fast ($\approx$150 fs) and slow ($\approx$1 ps) relaxation times. Detailed descriptions of the characterization techniques employed for the SWCNT-SAM are given in [4].

Q-switching mode-locking was readily observed, while cw mode-locking was achieved within a range of M1-M2 separation $\approx$115–118 mm. This corresponds to a mode radius on the SWCNT-SAM of $\approx 150-165\ \mu m$. Indeed,

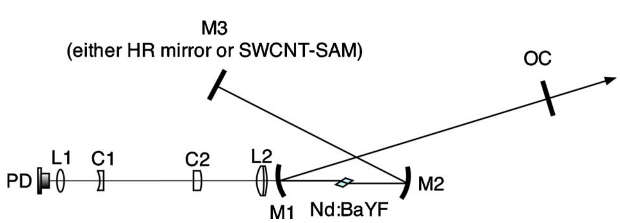

Fig. 1. (Color online) Resonator layout. PD, pump laser diode; L1, aspheric lens; L2, achromat lens; C1, C2, cylindrical lenses (15× telescope); M1, M2, concave mirrors, 100-mm curvature, high-reflectivity (HR) at 1050 nm, high-transmissivity at 800–810 nm; M3, either flat mirror, HR at 1050 nm, or SWCNT-SAM (HR mirror with SA film coating); OC, output coupler mirror, 30′ wedge.

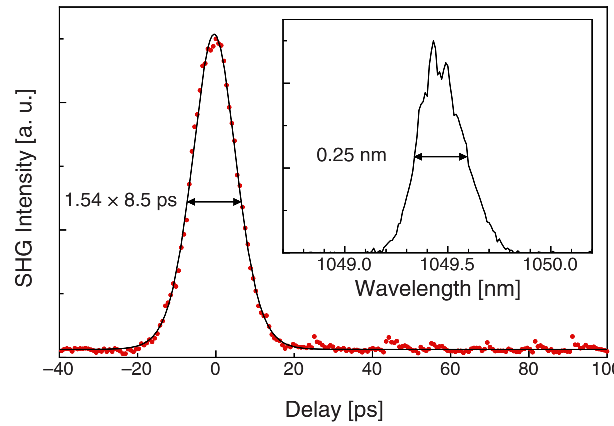

Fig. 2. (Color online) Autocorrelation of the mode-locking pulses at 1049 nm and spectrum (inset).

larger spot sizes resulted in Q-switched mode-locking, while smaller ones could produce local damage on the SWCNT film on SWCNT-SAM, depending on the position of the beam incidence.

The mode-locked Nd:BaYF laser operated at the main transition near 1049 nm, yielding $\approx$8.5-ps pulses at 194 MHz and 70-mW output power. The threshold for cw mode-locking was very close to the maximum output power available with the present setup. Though the laser was not self-starting, the stable cw mode-locking regime could easily be initiated by gently tapping one cavity mirror.

Figure 2 shows the autocorrelation trace and Fig. 3 the RF spectrum showing no Q-switching instabilities. The pulses were measured to be reasonably Fourier-limited with a time-bandwidth $\Delta\nu \times \Delta\tau \approx 0.58$.

The same resonator was used to assess the ability of the SWCNT-SAM to achieve cw mode-locking with the other Nd^{3+}-doped gain media. Nd:glass readily allowed cw mode-locking with femtosecond pulses or even chirped picosecond pulses in the positive dispersion regime (these results will be reported elsewhere [13]). Instead, neither Nd:YAG nor $Nd:YVO_4$ crystals evidenced the slightest tendency to mode-locking or at least Q-switching mode-locking.

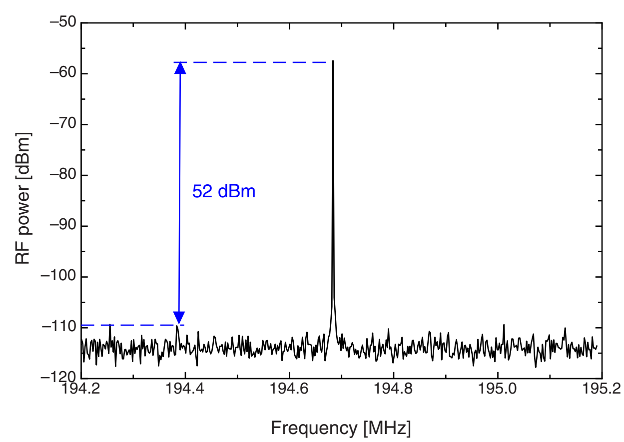

Fig. 3. (Color online) Radio frequency spectrum (bandwidth resolution 100 Hz).

3. DISCUSSION

These observations suggest that, given the characteristics of the SWCNT-SAM available, mode-locking was severely hindered in the case of Nd:YAG and $Nd:YVO_4$, while Nd:BaYF was barely suitable for cw mode-locking.

This increasing difficulty to evolve free-running spontaneous fluctuations into either cw mode-locking or even Q-switching mode-locking when choosing gain media with larger emission cross section and smaller gain bandwidth can be explained as follows.

Besides considering the self-starting mode-locking requirements, a necessary condition ensuring the existence of a positive gain window for field fluctuations was discussed in [9,10]. Random noise fluctuations of the instantaneous power $P(t)$ have durations $\tau_p < T_R/m$, T_R being the round-trip time (≈5 ns in our experiments) and m the number of longitudinal modes related to the gain bandwidth of neodymium lasers. For Nd:glass m is $<10^2$ [10], and even smaller in Nd:YAG and $Nd:YVO_4$, owing to a ≈20-times reduction of the fluorescence bandwidth. Therefore, in our experiments τ_p is >50 ps and the SWCNT-SAM behaves as a fast absorber during the early stages of the mode-locking process:

$$q(t) \approx \frac{q_0}{1 + \dfrac{P(t)}{A_a I_a}}, \tag{1}$$

where $q(t)$ is the loss at time t, q_0 the saturable loss, A_a the mode area, and $I_a = F_a/\tau_a$ the saturation intensity, and where τ_a is the saturable absorber relaxation time. Since $\tau_p \gg \tau_a$ we may consider the slower relaxation time component $\tau_a \approx 1$ ps. The instantaneous loss change due to the transit of the low-energy fluctuation in the SA is then

$$\Delta q(t) \approx -q_0 \frac{P(t)}{A_a I_a}. \tag{2}$$

Therefore, a positive gain window for the fluctuation exists, provided that the net gain averaged over $P(t)$ is >0; that is,

$$\langle \Delta g - \Delta q \rangle_{P(t)} = \frac{\displaystyle\int_{-\infty}^{+\infty} [\Delta g(t) - \Delta q(t)] P(t) dt}{\displaystyle\int_{-\infty}^{+\infty} P(t) dt} > 0, \tag{3}$$

where

$$\Delta g(t) \approx -g_{cw} \frac{\displaystyle\int_{-\infty}^{t} P(s) ds}{A_g F_g} \tag{4}$$

is the variation of gain due to saturation during the transit of the perturbation $P(t)$, g_{cw} is the saturated gain of the laser, and A_g and F_g are the mode area and saturation fluence of the gain medium, respectively. The condition for successful mode-locking initiation is therefore

$$q_0 > \frac{F_a A_a}{F_g A_g} \frac{g_{cw}}{\tau_a} \left[\frac{\dfrac{1}{2}\left(\displaystyle\int_{-\infty}^{+\infty} dt P(t) \right)^2}{\displaystyle\int_{-\infty}^{+\infty} dt P^2(t)} \right] \approx \frac{F_a A_a}{F_g A_g} g_{cw} \frac{\tau_p}{\tau_a}, \tag{5}$$

where τ_p can be assumed as the FWHM of the fluctuation $P(t)$.

Considering the parameters of Nd:glass in Table 1 and our experimental setup, Eq. (5) yields $q_0 > 2 \times 10^{-5}$, which is easily fulfilled, while for Nd:YAG a much higher modulation depth is required, $q_0 > 0.1\%$, comparable to that of SWCNT-SAMs usually employed for low-gain solid-state femtosecond lasers and used in our experiments. The situation is slightly better for Nd:BaYF and still worse for $Nd:YVO_4$ as suggested by cross section values and gain bandwidths summarized in Table 1.

The condition on the critical output power P_c required for stable cw mode-locking is readily fulfilled for the Nd:BaYF laser owing to the small value of F_a for the SWCNT-SAM [14],

$$P_c = \frac{T_{oc}}{T_R} \sqrt{F_a A_a F_g A_g q_0}, \tag{6}$$

yielding $P_c = 41$ mW, considering the parameters of our experimental setup.

At first sight, tighter focussing in the SA might improve the mode-locking tendency. However, the drawback is the damage threshold of the SWCNT polymer film, which can safely handle average intracavity power only up to <10 kW/cm^2 [3,4].

One possibility to enable successful mode-locking with this kind of SWCNT-SAs and Nd^{3+}-doped crystals consists of reduction of non-saturable losses and increase of modulation depth q_0, maintaining the same g_{cw}. The situation is somehow relaxed for SESAMs, since the slow component τ_a is often much longer (≈10 ps), and the average power limit is much higher (at least an order of magnitude better), according to our direct experience.

It is worth mentioning that the relatively low damage threshold of the particular sample used in our experiments seems related to strong bundling [2] of nanotubes, as well as to the polymer concentration. Preliminary saturable absorption investigations on the latest series of SWCNT films with lower polymer concentration show a damage threshold significantly improved. For example, though working at a longer wavelength of 1.2 μm, Cr:forsterite lasers withstanding average intensity of <80 kW/cm^2 have been demonstrated [7].

Higher modulation depth of the SA would also yield shorter pulses [15], with performance closer to the 2.6-ps pulsewidth recently achieved with SESAMs in the Nd:BaYF laser [12].

4. CONCLUSIONS

We have demonstrated, for the first time to our knowledge, cw mode-locking in a diode-pumped picosecond laser based on a Nd^{3+}-doped crystal and a SWCNT-SAM. The Nd:BaYF laser generated ≈8.5-ps pulses and 70 mW at low pump power ≈1 W. With the same setup we also investigated Nd:YAG and Nd:YVO_4 crystals, which did not exhibit any mode-locking tendency. We suggest that this is a consequence of the strong gain saturation occurring in the early stages of pulse evolution, preventing the creation of a positive gain window given the small modulation depth of the SWCNT-SAM employed. Our study further suggests that a higher ratio between saturable and non-saturable losses is required to successfully mode-lock narrow bandwidth, high-gain neodymium lasers.

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