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Femtosecond single-mode diode-pumped Cr:LiSAF laser mode-locked with single-walled carbon nanotubes

Antonio Agnesi [a], Federico Pirzio [a,*], Elena Ugolotti [a], Sun Young Choi [b], Dong-Il Yeom [b], Fabian Rotermund [b]

[a] University of Pavia, Electronics Department — Via Ferrata 1, I-27100 Pavia, Italy
[b] Division of Energy Systems Research, Ajou University 443-749 Suwon, Republic of Korea

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ABSTRACT

A low threshold Cr:LiSAF laser pumped with an inexpensive single-mode laser diode emitting 120 mW was passively mode-locked with a novel ultrafast saturable absorber mirror based on single-walled carbon nanotubes (SWCNT-SAM). Pulses as short as 122 fs were achieved, tunable across 14 nm. A second pump diode coupled in polarization allowed to shorten the pulse duration to 106 fs, with up to 24-mW output power.

1. Introduction

$Ti:Al_2O_3$ laser technology has been leading the commercial market of ultrafast lasers for 20 years, owing to unique properties of such gain medium, in particular excellent thermo-mechanical characteristics and nearly octave-spanning fluorescence spectrum. These allowed generation of pulses as short as 5 fs [1] directly from a laser oscillator, an outstanding achievement still unmatched by any other solid-state laser media. However, especially for low-power operation, alternative laser technology near 800-nm wavelength has been investigated in another class of vibronic materials. Indeed, Cr^{3+} doped colquiirites such as Cr: LiSAF, Cr:LiSGAF, Cr:LiSCAF and Cr:LiCAF are leading candidates for developing low-cost, tunable, continuous wave (cw) or femtosecond laser sources around 800 nm [2] thanks to their attractive properties such as: direct diode pumping, broad emission bandwidth, high quantum efficiency and long fluorescence lifetime. This latter characteristic is particularly promising for the developing of low threshold devices, very attractive since the poor thermal properties of colquiirites limit power levels available without active cooling. Among these materials Cr:LiSAF is the most extensively studied for low-pump-power applications [3,4] since it has the higher emission cross section, the broader bandwidth and the lower scattering losses.

Kerr-lens mode-locked Cr:LiSAF oscillators were earlier reported generating 75-fs pulses with only 36 mW of pump power [3]. Owing to the ready availability of inexpensive single-mode red laser diodes at 150-mW output power, in the last few years there has been a significant effort also in power scaling of ultrafast diode-pumped colquiirites, with output power as high as ≈250 mW and pulse widths as short as 26 fs (with lower output power), demonstrated using SEmiconductor Saturable Absorber Mirrors (SESAMs) [5,6].

Indeed, employing saturable absorbers is the most effective means to induce self-starting passive mode-locking, allowing construction of robust and reliable ultrafast sources. The state-of-the-art solution is represented by SESAMs, however new nanostructured materials such as single-walled carbon nanotube saturable absorbers (SWCNT-SAs) [6,7] and graphene [8,9] have been more or less extensively investigated in solid state lasers in the last few years. They benefit from a much simpler fabrication process that allows lower costs. Moreover, compared to SESAMs, they have favorable properties such as larger bandwidth, faster recovery time and saturation fluence comparable or even smaller. Currently it is reported their use with Ti:Sapphire lasers [10], Cr:Forsterite or Cr:YAG lasers, or other solid-state and fiber sources such as Nd-, Yb-, Tm-doped lasers [7,11,12]. In this work we present for the first time, to the best of our knowledge, the use of SWCNT-SAMs to operate a low-threshold Cr:LiSAF laser in femtosecond soliton mode-locking regime.

2. Pumping system and CW experiments

The active medium was a 3%-doped, 4-mm-long, Brewster-cut, Cr: LiSAF crystal. The very wide absorption bandwidth, extending from about 580 to 710 nm, allows for direct diode pumping with commercially available, low-power, single-mode visible red-emitting laser diodes. The technology of such lasers, thanks to the expansion of the optical

* Corresponding author. Tel.: +39 0382985225; fax: +39 0382422583.
E-mail address: federico.pirzio@unipv.it (F. Pirzio).

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data storage market, has been well developed in last decades, allowing for low-cost, reliable devices. However, narrow-stripe high-brightness devices required for effective diode pumping of ultrafast lasers are limited in power to 100–200 mW.

Pump power up-scaling is usually realized employing multiple laser diodes, typically either through polarization or wavelength multiplexing (taking advantage of the wide absorption bandwidth). In our experiments, initially we used a single Mitsubishi (101J27-01) high-brightness (measured $M^2 \approx 1.1$) laser diode rated at 120 mW maximum output power at 660 nm. Later, a second identical laser diode, was coupled in polarization in order to increase the available pump power. No reshape of the pump beam was required, thanks to the very low astigmatism of the laser diodes.

We carried out, at first, a characterization of the laser performances in cw regime, employing several different output couplers. The resonator layout is depicted in Fig. 1. A standard X-folded resonator was set up in order to compensate for cavity mode astigmatism introduced by the Brewster-placed active medium.

The π-polarized pump power absorbed by the crystal was about 110 mW. With the optimum 2% transmittance output coupler (OC), we obtained up to 27 mW output power with a 32% slope-efficiency in cw operation. A very low 16.5-mW absorbed pump power at threshold was measured with the 1%-transmission OC, later employed in the cw mode-locking experiments.

In order to increase the absorbed pump power, we later added a second identical laser diode. The two pump sources were polarization coupled employing a polarizing cube transmitting the π-polarized pump beam, from the first diode, and reflecting the σ-polarized one, from the second diode (Fig. 1). The Cr:LiSAF crystal exhibits different pump absorption cross-section for π and σ polarizations; in particular we measured a pump power absorption of 98.5% for the π-polarized diode and 90% for the σ-polarized. Moreover the σ-polarized pump radiation experiences 11% reflection losses at the crystal interface, Brewster-cut for π-polarization. Absorbed pump power at laser threshold and slope efficiency did not change significantly, whereas the addition of the second diode allowed to increase the maximum output power up to about 59 mW with the optimum 1% output coupling. In Fig. 2 the characterization of laser operation in cw regime with several different OCs is reported. We observed that the extra power, provided by the double pump diode system, caused some thermal quenching effects in the laser crystal that was simply standing on its mount with no special heat-sinks: after an initial transient,

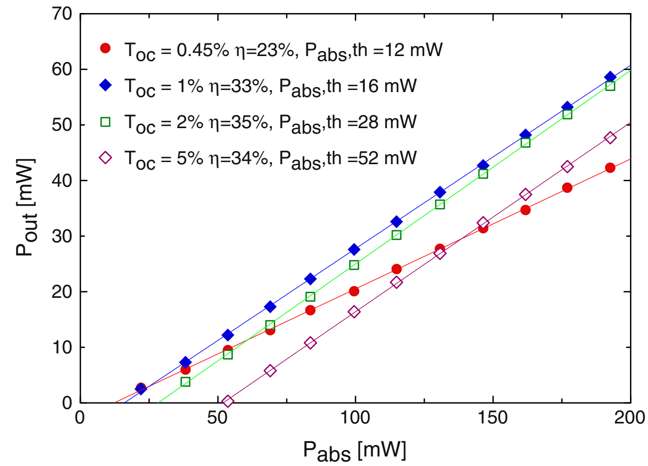

Fig. 2. Output power vs absorbed pump power (two pump diodes) with different OCs. Slope efficiencies (η) and absorbed pump power at lasing threshold ($P_{abs,th}$) are also reported.

average output power stabilized on the values reported in Fig. 2, that was few mWs lower than the starting level.

A dispersive Fused Silica (FS) prism was inserted in the cavity to assess the laser tunability (Fig. 1, inset 2). We measured the tuning range either employing as cavity end-mirror a standard, broadband HR mirror or the SWCNT-SAM later employed in cw mode-locking experiments. In both cases the tuning range extended from about 800 to about 935 nm, covering a significant fraction of the entire gain bandwidth and mainly limited by the dielectric optical coatings of the mirror set.

3. Mode-locking experiments

For cw soliton mode-locking we employed a classical two-prism intracavity group velocity dispersion (GVD) compensation scheme (Fig. 1, inset 3). The FS prism separation was experimentally set to about 43 cm for optimum net negative intracavity GVD.

SWCNT-SAMs, if compared to commercially available SESAMs, typically present a lower saturation fluence, that allows some simplification in the cavity design (i.e., no provision for an additional folding

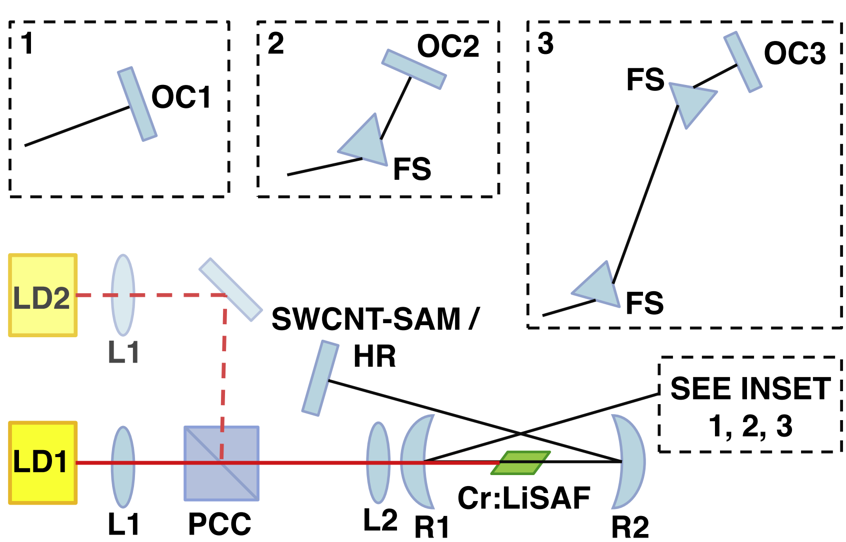

Fig. 1. Cavity layout. LD1: pump laser diode, π-polarized; LD2: second pump laser diode, σ-polarized; L1: aspheric lens (f = 6 mm); PCC: polarization coupling cube; L2: spherical lens (f = 50 mm); R1 and R2: curved mirrors HR at 800–900 nm AR at 660 nm (R1 = 50 mm, R2 = 100 mm); FS: fused silica prism; OC1: output coupler for cw experiments (T = 0.5%, 1%, 2%, 5%) (inset 1); OC2 = OC3: output couplers for central wavelength tuning (inset 2) and ML operation (inset 3), T = 1%, 30′ wedge.

mirror set [13]), since no tight focusing over the SAM is needed to obtain stable cw mode-locking regime.

SWCNTs employed in our experiments were synthesized by HiPCO conversion technique as described in Ref. [10], by dispersing purified SWCNTs from Unidym Inc. in dichlorobenzene (DCB) via ultrasonic agitation. For accurate segregation the solution was eventually centrifugated. The well-dispersed SWCNT/DCB solution was then mixed with PMMA solution, spin coated on the dielectric surface of a HR mirror and then baked. The thickness of the SWCNT absorber layer was measured to be few hundred of nm.

The sample was preliminarily characterized with time-resolved pump-probe and nonlinear reflection measurement techniques as described by I. H. Baek et al. [10], whereas the linear reflection was measured with a spectrophotometer showing qualitatively the similar behavior as observed earlier. The SACNT-SAM exhibited a fast decay of <150 fs with a slow nonlinear response of <1.5 ps. The saturation fluence was estimated to be <29 μJ/cm^2, the modulation depth of about 0.15% and the nonsaturable loss of 1%.

Here, the SWCNT-SAM was employed as an end-mirror and the radius of the cavity mode over it (≈30–60 μm) was controlled by varying the distance between the SWCNT-SAM and R2 mirror (Fig. 1). We observed that the spot size on the SWCNT is not a critical parameter for obtaining cw mode-locking. The shortest pulses, reported in Fig. 3, were measured to be 122 fs with a bandwidth of 7.5 nm (FWHM), resulting in a time-bandwidth product of 0.36, close to the Fourier limit. The spectrum was centered at 875 nm and the average output power was 16 mW. Larger bandwidths have been obtained with even larger time-bandwidth products, turning out in slightly longer pulses.

Mode-locking threshold was relatively low, occurring at an absorbed pump power of about 73 mW and at an output power of 4.7 mW. A very low fluence of about 40 μJ/cm^2 on the SWCNT-SAM was measured at mode-locking threshold.

The central output wavelength tunability, in the mode-locked operation, was relatively narrow. Continuous central wavelength tuning was possible only from 868 nm up to 882 nm (see Fig. 4), even though femtosecond mode-locking at 820 nm was occasionally observed.

As expected by the theory of soliton mode-locking, the higher intracavity average power with double-diode pumping was beneficial in mode-locked operation since it allowed higher pulse energies and hence shorter pulses duration. The best result we obtained was a 106-fs pulse train, with a time-bandwidth product of 0.43. Both the pulse autocorrelation trace and the corresponding optical spectrum are shown in Fig. 5.

Output powers in mode-locked regime were dependent on the position of the incident beam in which the SWCNT was hit, and varied

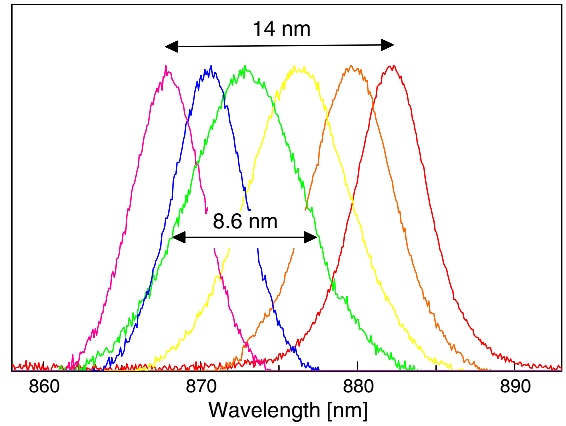

Fig. 4. Central output wavelength tunability in mode-locked regime.

between 16 and 24 mW because of the inhomogeneous absorber layer thickness. We noticed that more stable pulse trains were observed when average output power was ≈16 mW, probably due to a higher local modulation depth of the saturable absorber mirror. Either with the single- or double-diode pumping, mode-locking was not self-starting and laser pulse trains were stable for only few minutes. Usually a cw component started growing and eventually collapsed the spectrum, but the original cw mode-locking performance was completely recovered simply stopping the pump beam or translating the SWCNT-SAM. As we observed with other bulk solid-state lasers such as Ti:sapphire and Cr:forsterite, these phenomena are suppressed by purging the absorber layer with a steady flux of dry nitrogen, suggesting some reversible photo- or thermal-reaction occurring in presence of the dielectric mirror coated substrate and currently subject of further investigation. Instead, the transmitting SWCNT-SAs deposited on a transparent substrate do not show such behavior with all other lasers investigated previously and allow definitely more stable operation without nitrogen purging, but unfortunately no suitable SWCNT-SA were available for this Cr:LiSAF experiment.

4. Conclusions

In this work we report, to the best of our knowledge, the first demonstration of SWCNT-SAM passive cw mode-locked operation of a Cr:LiSAF laser. 122-fs pulses with an average output power of 16 mW and a central output wavelength tuning range of 14 nm

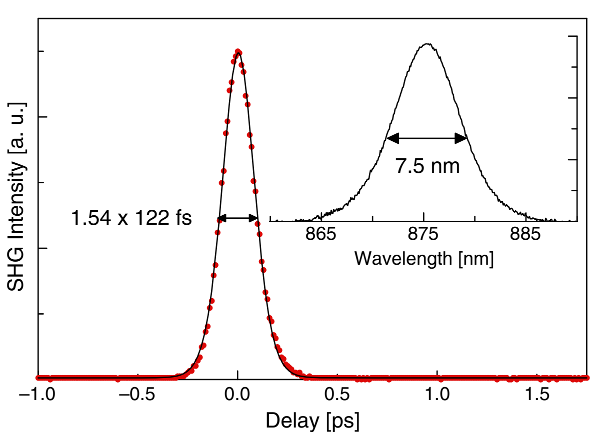

Fig. 3. Autocorrelation trace and correspondent optical spectrum of the mode-locked pulse train obtained with single diode-pumping.

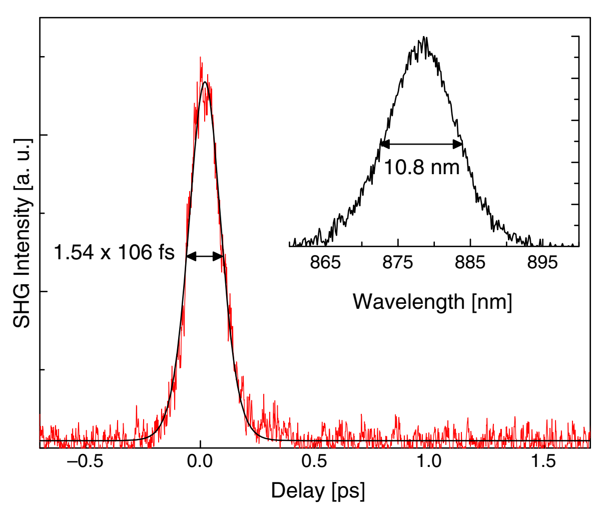

Fig. 5. Shortest pulse autocorrelation obtained with double diode pumping. In the inset is shown the correspondent optical spectrum.

were obtained with a single diode pumping system. Increasing the pump power thanks to a second laser diode coupled in polarization allowed us to reduce the pulse duration to 106 fs and increase the output power to 24 mW. Some destabilizing effect occurring with the SWCNT-SAM at this particular wavelength has been identified and is being currently investigated. Interestingly, this appears to be closely related to the presence of the multilayer mirror substrate and is a completely reversible effect. However, SWCNT films deposited on transparent glass substrates do not show such issues and should be more promising as already demonstrated with $Ti:Al_2O_3$ laser experiments.

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