HIGH POWER ND: YVO₄ LASER MODE-LOCKED BY CASCADING SECOND ORDER NONLINEARITY

HRISTO ILIEV, IVAN BUCHVAROV

Department of Quantum Electronics, Faculty of Physics, St. Kliment Ohridski University of Sofia

Христо Илиев, Иван Бъчваров. СИНХРОНИЗАЦИЯ НА МОДОВЕТЕ НА МОЩЕН ND:YVO₄ ЛАЗЕР ПОСРЕДСТВОМ КАСКАДНА НЕЛИНЕЙНОСТ ОТ ВТОРИ ПОРЯДЪК

В статията са представени експериментални резултати от пасивна синхронизация на модовете на Nd:YVO₄ лазер посредством първи тип генерация на втора хармонична в периодично поляризирани нелинейни кристали. Описани са два нелинейни ефекта, които допринасят за синхронизацията на модовете: формиране на нелинейна $\chi^{(2)}$ -леща в резонатора и отражение от нелинейно огледало с удвояване на честотата (HOVЧ). HOVЧ, чието отражение зависи от интензитета на лъчението в резонатора, осигурява самостартиране и устойчиво поддържане на режима на синхронизация на модовете, докато формирането на $\chi^{(2)}$ -лещата допринася за многократно скъсяване продължителността на импулсите. Тази хибридна техника позволява генерирането на стабилна последователност от импулси с висока средна мощност и продължителност на отделния импулс до няколко рs. Получената средна мощност с нелинейното огледало, базирано на нелинеен кристал (литиев триборат), е в интервала 3,2–4,1 W, а с периодично поляризиран стехиометричен литиев танталат е 0,7–2,7 W Продължителността на импулсите е съответно 8,3 ps и 6,3 ps, а честотата на следване ~110 MHz.

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Experimental results on passive mode-locking of an Nd: YVO_4 laser using type-I intracavity frequency doubling in bulk and periodically poled nonlinear crystals are reported. Both, negative $\chi^{(2)}$ lensing and frequency doubling nonlinear mirror (FDNLM) are exploited for the laser mode-locking. The FDNLM based on intensity dependent reflection in the laser cavity ensures self-starting and self-sustaining mode-locking while the $\chi^{(2)}$ lens process contributes to substantial pulse shortening. This hybrid technique enables generation of stable trains of pulses at high-average output power with several picoseconds pulse width. The pulse repetition rate of the laser is in order of 110 MHz with average output power ranging from 3.2- 4.1 W in the case of using bulk lithium triborate (LBO) nonlinear crystal and 0.7 W to 2.7 W in the case of using periodically-poled stoichiometric lithium tantalate. The measured pulse durations were 8.3 ps and 6.3 ps respectively.

Keywords: Diode pumped lasers, passive mode-locking, quasi-phase matching, periodically-poled crystals

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For contacts: Ivan Buchvarov, Department of Quantum Electronics, Faculty of Physics, St. Kliment Ohridski University of Sofia, 5 James Bourchier Blvd., BG-1164 Sofia, Bulgaria, tel. 00359-28161708, fax. 00359 2 962 5276, E-mail: *ivan.buchvarov@phys.uni-sofia.bg*

1. INTRODUCTION

Nowadays, diode-pumped mode-locked neodymium (Nd) lasers operating at \sim 100 MHz repetition rate, are the main source of high average power picosecond pulses with duration ranging from a few up to more than hundred picoseconds [1–4]. Such laser sources are particularly attractive for nonlinear optics and spectroscopy as well as for applications in medical diagnostics and micro-machining. In general, much shorter pulses can be generated with passive mode-locking which, in contrast to active mode-locking techniques, does not require external driving source and the repetition rate is determined solely by the round trip time of the cavity [1].

With few exceptions, e.g. the additive mode-locking [5], multi-Watt operation of picosecond diode-pumped Nd oscillators has been demonstrated mainly by two passive mode-locking methods, one based on semiconductor saturable absorber mirrors (SESAMs) [6-8] and the other—on intracavity frequency doubling. The highest average powers demonstrated so far with SESAM technology and Nd:YVO₄ or Nd:YAG are in excess of 20 W [7,8,9]. Although SESAMs are well established devices for lasers emitting around 1 μ m, their residual absorption, leading to heating, is the major intrinsic drawback which determines their damage threshold (current state of the art is ~0.5 GW/cm² at 20 ps pulse duration) [7], and limits their power scaling capabilities. Intracavity frequency doubling is a promising mode-locking approach for up-scaling the power of such picosecond solid-state lasers because the damage threshold of nonlinear crystals is an order of magnitude higher and the low residual absorption at the fundamental wave enables operation at high average power. Furthermore, this approach is easily extendable to any laser spectral region.

In the case of intracavity type-I second-harmonic generation (SHG), two different passive mode-locking mechanisms can be utilized. The first one is based on the intensity dependent reflectivity of the frequency-doubling nonlinear mirror (FDNLM), consisting of a crystal for SHG, a dielectric mirror, and dispersive medium between them [10-13]. In this scheme, the second harmonic generated upon passing through the nonlinear crystal (NLC) is completely reflected by the dielectric mirror and interacts again with the fundamental on its way back through the same nonlinear crystal, thus a double-pass (cascaded) SHG process takes place. The dielectric mirror should be a dichroic mirror (DM), having comparatively low reflectivity at the fundamental and high reflectivity at the second harmonic. Hence, the reflectivity of the combination, crystal for SHG and a dielectric mirror, called FDNLM, is governed by the back conversion from second harmonic to the fundamental [12,14]. The dispersive medium determines the relative phase shift of the fundamental and second harmonic waves accumulated between the NLC and the DM. The main drawback of the FDNLM is the longer duration of the output

pulse in comparison with the pulse traveling inside the laser cavity [15]. The second mode-locking mechanism is based on cascaded $\chi^{(2)}$ lens formation [2,16]. It does not exhibit the pulse duration limitation characteristic for the FDNLM. Thus it provides shorter pulses but higher nonlinearity is required for self-starting mode-locking. Also, this technique is sensitive to external perturbations. In this respect, $\chi^{(2)}$ lens mode-locking assisted by FDNLM has a potential which has not been exploited effectively, yet [17], although this is a natural combination of two effects taking place in the same mode-locking device, in contrast to combining $\chi^{(2)}$ lens formation with a SESAM [18]. Moreover, the potential of quasi-phase-matched materials with their high effective nonlinearity and absence of spatial walk-off seems not to have been properly utilized. There are only a few reports in which SHG in periodically poled KTP (PPKTP) has been employed for passive mode-locking [10,19] although the FDNLM with PPKTP produced so far the highest average powers (5.6 W with Nd:YVO₄) for the intracavity frequency doubling approach [10].

In this work we present the results on passive mode-locking of an Nd: YVO₄ laser by cascading second order nonlinearity. The FDNLM is investigated using a commercially available LBO NLC. The hybrid technique is also utilized with periodically polled stoichiometric lithium tantalate. In this schema the FDNLM initiates the passive mode-locking process and ensures self-sustained operation while the defocusing $\chi^{(2)}$ effect leads to the shortest pulse duration of 6.3 ps. Thus, although each of the two mode-locking mechanisms in principle can be used alone for passive mode-locking of the laser, the hybrid scheme proves beneficial for the robust laser performance in this study. The output power is ranging from 0.7 W to 2.7 W.

2. EXPERIMENTAL SET-UP

The design of the mode-locked laser is based on a 1.4 m long linear cavity (Fig. 1). The active element (AE) is a 5-mm long, 0.5 at. % doped Nd: YVO_4 crystal with an aperture of 2.5×2.5 mm². The *a*-cut cut sample is 1.5° -wedged. It is pumped by a 50-W 808-nm laser diode bar coupled into a 400 µm optical fiber (NA = 0.22). The AE is mounted in a metal holder maintained at a temperature of 25° C by running water. The resonator end mirrors are the high-reflective rear face of the AE and the flat output coupler (OC). The other face of the AE is antireflection coated. The radius of curvature of the folding mirror M1 and the focal length of the lens F3 (80 mm) are chosen to ensure beam radius of 80 µm in the nonlinear crystal (NLC) and 200 µm in the AE. Single transverse mode operation is achieved by overlapping the resonator eigenmode with the pump waist formed by the objective lenses F1 and F2.



Fig. 1. Schematic of the laser cavity: F1, F2—pump objective; AE—Nd: YVO_4 active element; M, M1, M2—highly reflecting mirrors at the fundamental wavelength, F3—focusing lens, NLC nonlinear crystal, OC—output coupler

After initial cavity alignment, the distance between lens F3 and output coupler OC is optimized for maximum output power in the fundamental transverse mode - TEM_{00} . The laser operates in σ -polarization. The distance between the NLC and OC is set around 25 mm. This distance is optimized for each mode-locking experiment by translating the NLC in order to achieve self-starting and continuous mode-locked operation.

3. MODE-LOCKING TECHNIQUE

The intracavity type-I SHG provides two different types of passive modelocking mechanisms: The first one is amplitude shaping and based on the intensity dependent reflectivity of the FDNLM and the second one is phase shaping and based on cascaded $\chi^{(2)}$ nonlinear phase shift of the fundamental wave, i.e. $\chi^{(2)}$ lens mode-locking.

A schematic of the FDNLM is shown on Fig. 2. The FDNLM consists of the frequency doubling NLC and the dichroic mirror DM, high reflecting at the second harmonic (2 ω) and partially reflecting at the fundamental (ω) wavelength. In general, the reflection by the FDNLM includes a two-way pass of the beam through the NLC. At perfect phase-matching of the SHG, the FDNLM reflection is governed by the SHG conversion efficiency which is a function of the input beam intensity and the phase difference $\Delta \varphi_{in} = 2\varphi_{1,in} - \varphi_{2,in}$ between the fundamental and second harmonic wave in the beginning of the second pass through the NLC. If $\Delta \varphi_{in} = \Delta \varphi_{out} \pm (2m+1)\pi$, m = 0, 1, 2..., see Fig. 2, the second harmonic is partially reconverted into the fundamental on the way back, leading to increase of the FDNLM reflectivity at the fundamental wavelength. The higher the intracavity intensity, the higher will be the FDNLM reflectivity with respect to the fundamental wave.



Fig. 2. Schematic of the FDNLM: NLC, nonlinear SHG crystal; DM, dichroic mirror

If the DM reflects partially the fundamental wave, the value of the FDNLM reflectivity can be higher than the value of the linear reflectivity of this mirror. Thus the FDNLM introduces a positive nonlinear feedback into the laser resonator. The nonlinear reflectivity $R_{\rm NL}$ normalized to the reflectivity R_{ω} of the dichroic mirror is shown on Fig. 3a as a function of the single pass SHG efficiency. In the case of $\Delta \varphi_{in} = \Delta \varphi_{out} \pm 2m\pi$, m = 0, 1, 2..., in the second pass through the nonlinear crystal, the phase difference $\Delta \varphi_{in}$ between the fundamental and the second harmonic waves enables further conversion of the fundamental into the second harmonic wave leading to further decrease of the fundamental beam intensity. Thus the FDNLM introduces a negative nonlinear feedback and the FDNLM reflectivity decreases with increasing intensity of the fundamental beam, i.e. with increasing single pass SHG efficiency, see Fig. 3b. The proper adjustment of the value of $\Delta \varphi_{in}$ for specific FDNLM operation is realized by changing the distance between NLC and DM or by slightly rotating a glass plate placed between the NLC and DM. In both cases the dispersion in air and glass causes change in $\Delta \varphi_{in}$. Originally the FDNLM was proposed first by Stankov [19] as a mode-locking technique based on perfectly phase-matched SHG.

In the presence of phase-mismatch, the back conversion to the fundamental upon the second pass through the NLC depends on the phase shift $q = \Delta q_{in} + \Delta kL$, where $\Delta k = k_2 - 2k_1$ the wave vector mismatch, and L is is the length of the SHG crystal. Hence, the intensity dependent reflectivity of the FDNLM is a function of the additional parameter ΔkL which leads to new requirements for $\Delta \varphi_{in}$ in comparison to the perfect phase matching case. Figure 4 shows the normalized nonlinear reflectivity $R_{\rm NL}/R_{\omega}$ of the FDNLM, the pulse shortening ratio $\tau_{\rm out}/\tau_{\rm in}$, equal to the reflected pulse duration $\tau_{\rm out}$ divided by the pulse duration of the input pulse $\tau_{\rm in}$, and the nonlinear phase shift φ_1 of the fundamental wave reflected by the FDNLM.



Fig. 3. Analytically calculated normalized FDNLM reflection as a function of the SHG efficiency for perfect phase matching and different R_{ω} , (a) $\Delta \varphi_{in} = \Delta \varphi_{out} \pm (2m+1)\pi$; (b) $\Delta \varphi_{in} = \Delta \varphi_{out} \pm 2m\pi$, m = 0,1,2...

The numerical analysis in Fig. 4 is performed within the plane-wave approximation assuming vanishing residual absorption. The corresponding coupled wave equations can be found elsewhere [15]. The parameters chosen are $R_{\omega} = 80\%$ and $u_{10} = 1$ where the normalized input amplitude u_{10} is defined by $u_{01} = a_{01}L$, with —the nonlinear coupling coefficient, and a_{01} —the input amplitude [20]. The physical meaning of the normalized input amplitude u_{10} can be understood from the expression for the stationary conversion efficiency $= \tanh^2(u_{01})$ in the case of single pass SHG of a plane wave with input amplitude a_{01} and perfect phase matching. The magnitude of the pulse shortening as well as the actual existence of such pulse compression capability, depend primarily on $\Delta\varphi_{in}$. The pulse shortening is maximized when $\Delta\varphi_{in}$ is in the 1.5π – 1.75π range and this range remains in the vicinity of 1.5π when the phase mismatch ΔkL is varied between 0.3 and 1.5 rad. For phase-mismatch exceeding 5 rad, the pulse shortening effect of the FDNLM becomes negligible.



Fig. 4. Calculated pulse shortening ratio τ_{out}/τ_{in} (solid line), normalized nonlinear reflectivity R_{NL}/R_{ω} (dotted line), and nonlinear phase shift φ_1 of the fundamental wave (dashed line), as functions of $\Delta \varphi_{in}$

The second mode-locking mechanism is based on self-phase modulation of the fundamental wave, due to cascaded $\chi^{(2)}$ nonlinearity, where the cavity losses are modified by the $\chi^{(2)}$ lens formation. This leads to fundamental wavefront defocusing/focusing with intensity dependent focal length. The resulting lens in combination with appropriate hard or soft aperture causes strongly nonlinear intracavity loss modulation and eventually leads to mode-locking. In contrast to the third order Kerr effect, where the resulting lens effect is only positive (i.e. focusing lens), in cascaded second order nonlinearity the lens type depends on the sign of the phase-mismatch ΔkL and it can be "+" which means a negative or diverging lens and "–" which means a positive or converging lens. Also, the nonlinear phase shift can be saturated. The saturation intensity depends on the phase mismatch parameter ΔkL and increases when moving away from the perfect phase matching point. In contrast to the FDNLM technique, the cascaded $\chi^{(2)}$ lens mode-locking exists only for SHG at non-phase-matched condition, i.e. at $\Delta kL \neq 0$.

For better understanding of the $\chi^{(2)}$ lens formation and the way it leads to the phase shift, let us consider the case when a fundamental wave ω is incident on a type-I nonlinear crystal. In non-phase-matched condition the power is continuously converted from the fundamental to the second-harmonic (up-converted) and from second-harmonic to the fundamental (down-converted). A part of the initial fundamental wave is up-converted into second harmonic according to $2\omega = \omega + \omega$ and the two waves travel with different phase velocity. Due to the coupling of the two nonlinear processes a part of the second harmonic is back-converted to the fundamental according to $\omega_{bc} = 2\omega - \omega$ and this regenerated fundamental wave has different phase from the initial fundamental wave that has not been converted to

second harmonic. The phase difference depends on the conversion efficiency and consequently on the intensity of the incident beam. If the radial distribution of the beam is not uniform, this will introduce curvature to the fundamental wavefront and the cascaded $\chi^{(2)}$ process will act as a focusing or defocusing lens with intensity dependant focal length.

4. EXPERIMENTAL RESULTS AND DISCUSSION

4.1. Mode locking using SHG in LBO crystal

For the FDNLM we used a LBO crystal with dimensions $2x2x20 \text{ mm}^3$ cut for noncritical phasematching. Both $2x2 \text{ mm}^2$ faces are antireflection coated for fundamental and second harmonic wavelengths. The crystal is placed into an oven which controls the temperature to an accuracy of 0.5 °C. The NLC is slightly tilted in respect to the optical axis in order to avoid etalon effects.

When the resonator with the NLC inside (see Fig. 1) is optimized for maximal efficiency, the NLC temperature was set for optimal phase-matching conditions ($T_{\rm NLC} \sim 150$ °C), measuring second harmonic passed through mirror M1. However, the most efficient ML regime is achieved when the phase differences $\Delta \varphi_{\rm in} = \Delta \varphi_{\rm out} \pm (2m+1)\pi$. To achieve this condition, we optimized the distance between the NLC and output coupler in such a way that the SH wave, generated in the first pass through the crystal is converted back into fundamental wave during the second pass.



Fig.5. Input-output characteristics of the Nd:YVO₄ laser. Output coupler with T=20% for fundamental and HR for SH used. The mode-locking zone is marked by ovals

The dependence of the output versus the incident pump power is shown in Fig.5. We observed stable mode locking operation in the pump power range

between 12 and 15 W where the output is between 3.3 to 4.1 W, respectively. In this case, the measured FWHM of the autocorrelation trace is 12.8 ps, Fig. 6, which corresponds to pulse duration of 8.3 ps, assuming sech² pulse shape.



Fig. 6. Autocorrelation function measured by rotating-mirrors (continuously scanning) autocorrelator using non-collinear SHG

4.2. $\chi^{(2)}$ lens mode locking assisted by FDNLM

Mode-locked operation is this case was studied with four different output couplers, three DMs with partial reflectance (OC1 with 95%, OC2 with 80% and OC3 with 70%) at the fundamental wavelength and highly reflective for the second harmonic, and one DM, OC4, with partial reflectance (95%) at the fundamental and highly transmitting the second harmonic. The phase matching of the intracavity PPMgSLT crystal is controlled by changing the crystal operating temperature. The measured second-harmonic power versus temperature shows maximum conversion efficiency at 29°C (holder temperature) and ~4°C FWHM of the temperature phasematching curve (Fig. 7). Stable passive mode-locking operation is observed, however, at higher temperatures, around 34.5 °C. The optimum temperature varies in an interval of ± 0.5 °C for each experiment.



Fig. 7. Normalized single-pass second harmonic (SH) intensity as a function of the crystal holder temperature (T) measured in the continuous-wave regime with OC4 but similar dependence is observed for all DMs

A temperature induced phase-mismatch of ~4.8 rad at 34.5°C is calculated using the temperature dependent Sellmeier equations for stoichiometric lithium tantalate (SLT) [21]. Similar value of ~ 4.5 rad is calculated using the value θ = 0.81 rad/°C for the phase-mismatch coefficient, obtained by fitting the curve in Fig. 7 with theoretically calculated SHG signal for the resonator Gaussian beam as a function of mismatch ($\Delta kL/2$) = $\theta \Delta T$ [23].

The highest output power and efficiency in mode-locked operation are achieved with the 30%-transmittance output coupler OC3. Figure 8 shows the dependence of the output versus the incident pump power. The laser threshold is 3.6 W. Passive mode-locking is observed in two distinct pump power ranges, from 10 to 10.5 W and from 13.1 to 14.6 W. The modeling of the field distribution in the resonator shows two stability zones depending on the thermal lens in the Nd:YVO₄ AE. In the vicinity of the zone limits the mode size in the AE is changing. Therefore the efficiency is dropping due to non-optimum overlapping with the pump beam.



Fig. 8. Laser output power as a function of incident pump power using OC3

The highest output power (2.5 W) in stable mode-locking regime is obtained at 12.8 W of incident pump power. In this case, the measured FWHM of the autocorrelation trace is 14.4 ps, Fig. 9a, which corresponds to pulse duration of 9.4 ps, assuming sech² pulse shape. The shortest pulse duration, 6.3 ps, see Fig. 9b, is obtained using OC1 with 5% transmittance at the fundamental, highly reflecting at the second harmonic.



Fig. 9. Autocorrelation functions using two dichroic output couplers with high reflectivity at the second harmonic, OC3 (a) and OC1 (b)

Successful mode-locking is achieved with all the three dichroic OCs with different transmission at the fundamental wavelength and high reflection at the second harmonic. In this case both nonlinear reflection and $\chi^{(2)}$ lens process are simultaneously present. The nonlinear reflectivity of the FDNLM strongly depends

on the phase-mismatch and reaches a maximum level when the second harmonic is perfectly phase-matched. On the other side, $\chi^{(2)}$ lens exists only if there is a phase-mismatch. As can be seen from Fig. 10, this effect is, however, essential for obtaining short pulses, compare with Fig.7. In fact, at the experimentally observed phase-mismatch, calculations predict negligible pulse-shortening effect from the FDNLM action as described in Fig. 4.



Fig. 10. Pulse duration (symbols) of the mode-locked Nd:YVO₄ laser at maximum output power with OC1, measured in dependence on the NLC holder temperature T

In order to analyze the relative contribution of the two mechanisms to the mode-locking process we suppress the FDNLM effect by using OC4 having the same reflectivity as OC1 at the fundamental but high transmission at the second harmonic. The laser threshold is the same as in the case of using OC1. The stable output power in the mode-locking regime amount 0.7 W, at a pump power around 13 W. The measured autocorrelation trace width is 7.2 ps corresponding to pulse duration of 6.3 ps assuming sech² pulse shape. The mode-locking displays amplitude fluctuations less than 2% on millisecond time scale and long term stability with hours of continuous operation.

4. CONCLUSION

We demonstrate operation of a passively mode-locked Nd:YVO₄ laser by cascading second order nonlinearities. In the hybrid scheme, the cascaded $\chi^{(2)}$ lens mode-locking process is the dominant mechanism for generation of pulse durations below 10 ps. Combination of the FDNLM technique with the cascaded $\chi^{(2)}$ lens improves the long term stability and the self-starting capability of the system. The main result from the present study is the proof that stable mode-locking can be obtained up to the maximum output power achievable in the CW mode of operation. Thus we achieve an order of magnitude higher average powers in

comparison to the previously reported 350 mW with cascaded $\chi^{(2)}$ lens formation in a Nd:GdVO₄ laser [16] or a combination with the FDNLM in a Nd:YVO₄ laser [18]. So far we are unable to reach the highest power level achieved with the FDNLM alone, but although 5.6 W were reported in [10] with the same laser material, the pulse durations in that work (20-ps) are roughly 3 times longer. We believe, however, that further power scaling of the present system will be possible by redesigning the laser cavity with special consideration of the thermal lens formation, shifting the region of stable mode-locking to yet higher output powers, the limit again being set by the TEM₀₀ power achievable in the CW regime.

As already mentioned, the technique described is feasible in wide spectral regions, and we plan to apply it in other types of lasers operating at wavelengths where no SESAMs exist, like Tm-lasers near 2 microns. Moreover, this technique could be interesting also for Yb-lasers, which emerge as an alternative to Nd-lasers in the 1-micron spectral range, with a huge potential for power scaling due to the smaller quantum defect, especially in combination with the thin disk concept. Although such systems, based on Yb:YAG, have already produced tens of Watts of output power at sub-picosecond pulse durations [5,23], one of the disadvantages of three-level Yb-based materials related to their longer lifetimes is that in the mode-locking regime they are more prone to Q-switching instabilities.

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