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Intracavity frequency-tripling of actively mode-locked diode-pumped Nd:YAG laser

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Abstract. We report the first diode-pumped solid-state laser operating in cw-mode-locked regime and simultaneously achieving intracavity frequency-tripling. This laser provide UV picosecond pulses ($\lambda = 355$ nm) of 10 ps duration with 0.5 mW average power at 150 MHz repetition rate. A different set of adjustments gave rise to a Q-switched mode-locked regime. Trains of hundred UV pulses of 60 ps duration and 4 W peak power were produced in this latter case at 50 kHz repetition rate.

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In areas such as photobiology, electronics, or real time spectroscopy, high repetition rate, ultrashort UV pulses are required for studying the dynamics of fast processes. One efficient way of generating UV pulses is to use first a diode-pumped mode-locked solid-state laser, delivering ps pulses in the near infrared, and then convert such IR radiation into visible radiation first and UV radiation in a second step by nonlinear (NL) frequency conversion with suitable nonlinear crystals. For moderate power regime, extracavity frequency conversion by single-pass into NL crystals is poorly efficient. It is possible to try frequency conversion inside an additional cavity external to the main IR oscillator [1, 2]. But this solution which requires an electronic servo to maintain the resonance locking is rather complicated. Then it is simpler and potentially very efficient to perform frequency conversion inside the IR oscillator where a high-fundamental field is available. However, intracavity frequency conversion within the IR oscillator introduces high nonlinear loss at the fundamental resulting in severe modification of laser performances [3, 4]. Indeed, with an actively mode-locked laser, it is impossible to perform efficient intracavity second-harmonic generation (SHG), even with an efficiency as moderate as 1%, without strongly increasing pulse duration from few ps to several hundreds of ps [4]. Intracavity SHG introduces inside the IR oscillator losses which increase with the laser peak power. Then it is a competing mechanism with active mode-locking which involves lower loss for pulsed than for continuous wave (cw) operation. It leads to large pulse duration, low peak intensity of visible pulses weakly suitable for efficient further NL frequency conversions into the UV spectral region.

A promising technique using passive mode-locking with an antiresonant Fabry–Pérot saturable absorber has been proposed [5]. B. Braun et al. have demonstrated that it was possible to achieve intracavity frequency-doubling without significant pulse broadening and with high conversion efficiency allowing further efficient conversion into UV.

In this paper, we solved this problem as follows: we started from a diode-pumped acousto-optically mode-locked Nd:YAG laser operating at 1064 nm. A first nonlinear crystal devoted to SHG generates inside the cavity 532 nm green radiation during a first pass. This crystal is positioned in the cavity such that the green pulse is back-converted to infrared by difference-frequency mixing during its second pass after reflection on the totally reflecting rear mirror. Thus, the second-harmonic radiation is mainly confined in one part of the cavity, between the SHG crystal and the end mirror (Fig. 1). In this way, green pulses which are not available

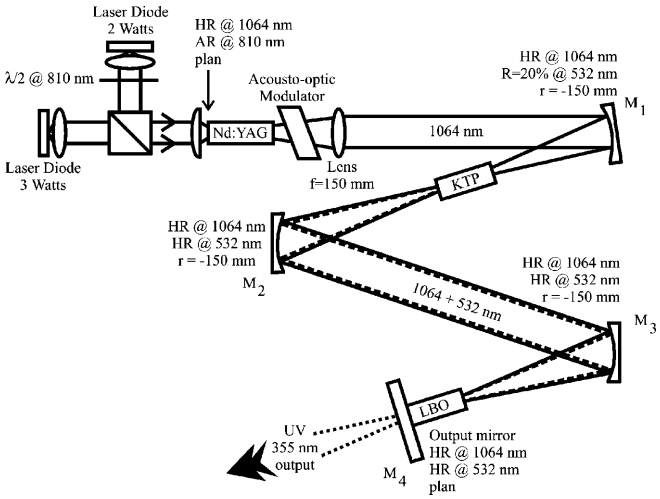

Fig. 1. Intracavity frequency-tripled actively mode-locked diode-pumped Nd:YAG laser. Second-harmonic radiation is mainly confined between the SHG KTP crystal and the output mirror

outside the cavity do not contribute to the nonlinear fundamental loss any more. Stable mode-locking provided by the acousto-optic modulator is preserved. Moreover this set-up is closed to the scheme of parametric nonlinear mirrors [6, 7] (due to K.A. Stankov) that was used for passive mode-locking of lasers. So it reinforces the acousto-optic modulator action. In doing so, green and infrared, which are confined in the cavity, can be converted into 355 nm ultraviolet by a second nonlinear crystal via sum-frequency generation (SFG). This second NL crystal is inserted in the resonator branch where the 532 nm power is the highest. The ultraviolet ps pulses exit the cavity through the end mirror which is dichroic (AR@355 nm; HR@1064 + 532 nm).

The overall nonlinear losses, i.e. non-reconverted second-harmonic and third-harmonic powers, experienced by the fundamental radiation must be maintained at a sufficiently low value in order to avoid pulse-duration lengthening as previously noted [4]. Indeed the SFG NL process can still counteract acousto-optic mode-locking. But the problem has been shifted into the UV region.

Additionally we tried Q-switched mode-locked operation. This kind of temporal behavior can be obtained when the acousto-optic modulator is detuned from the cavity length [8]. It must be noted that this self Q-switched mode-locked regime leads to pulse-duration increase by a factor 6. Despite this fact, this configuration led to more than twice UV average power in comparison with continuous-wave mode-locked regime.

1 Experimental set-up and results

For this experiment, we have used a W-shaped cavity with three curved folding mirrors (M_1, M_2, M_3). This geometry involves three beam waists (Fig. 1), the first one corresponds to the position of the laser crystal and the two others to the positions of the nonlinear crystals. The amplifier medium of the cavity laser was a Nd:YAG crystal (1 cm length). It was longitudinally pumped by two laser diodes emitting at 808 nm 2 W (SDL 2372 P1) and 3 W (SDL 2282 P1). The pump power incident on the laser crystal was 3.9 W. For the first conversion step (SHG), we used a KTP type II crystal ($\theta = 90°, \phi = 23.3°$ critically angle-tuned) with a length of 7 mm. A SFG LBO type I crystal of 8 mm in length was placed near the rear mirror of the cavity. An acousto-optic modulator was used to obtain mode-locked operation.

At first sight we had two alternatives to place the two nonlinear crystals. We first placed the KTP crystal at the end of the cavity, and the LBO crystal in the middle of the cavity. So we took advantage of the double-pass in the KTP crystal to make a stronger conversion to the visible. We obtained more that 100 mW of average power for the 532 nm wavelength. But, as previously said, the SHG introduces intensity-dependent losses on the laser wavelength that counteracts the mode-locking process which reinforces on the contrary high intensities. The resulting effect we observed was an increase of the pulse duration from about 30 ps to several hundreds of ps, and the laser stability became critical.

After such rather disappointing but foreseeable results we decide to move to a second configuration where the KTP crystal is placed in the middle of the cavity, and the LBO crystal at the end of the cavity. Our goal here was to confined the second-harmonic radiation between the KTP crystal and the end mirror. The KTP crystal produced SH of infrared radiation coming from the laser medium; it then performed reconversion of green into infrared by difference-frequency mixing with unconverted infrared. The second step of difference-frequency mixing only occurs for a proper phase relationship between green and infrared [9] which was achieved in practice by use of the dispersion of air paths, by use of the phase-shifts introduced by the mirror coatings, and by a fine tuning of the crystal orientation and position.

With this scheme, we first observed cw mode-locked operation. The second-harmonic average power that exited through mirror M_1 was 3.9 mW which corresponds to 5.4 mW of intracavity SH power. By measuring the amount of SH average power that exited through mirror M_3 we were able to deduce that the intracavity SH power at this place was 40 mW. It means that the SH power stored between the KTP crystal and M_4 is 7.4 times larger than between the KTP and YAG crystals. This measurement confirmed the existence of a nonlinear mirror effect that reinforces the mode-locked operation. Fundamental pulse duration of 18 ps has been measured from background-free IR autocorrelation. Green and UV pulse duration of 12.7 ps and 10.4 ps, respectively, were deduced from this IR autocorrelation. The pulse repetition rate was 150 MHz. For 3.9 W of 808 nm pump power incident to the laser crystal we obtained more than 0.5 mW of available UV cw power. Such a value is somewhat moderate but it must be noted that our system was far from being optimized. Indeed, the beam waist imposed within the KTP NL crystal was 70 μm. It is too large in comparison with the optimum value [10] which is close to 30 μm. So we could expect to increase the fundamental intensity incident to the KTP crystal by a factor 5.4 and then 30 times more SHG by changing the radius of curvature of our cavity mirrors in order to impose the suitable IR beam size. With such optimization the threshold of nonlinear mirror operation would decrease leading to shorter pulse duration. Moreover, the SFG LBO crystal has been manufactured for type I phase-matching condition i.e. second-harmonic and pump polarizations are expected to differ by 90°. Unfortunately this is not the case in our present experiment. Indeed, the KTP crystal being a type II phase-matching crystal, the polarization of the second harmonic is 45° with respect to the IR polarization. The SFG was consequently not optimized. We should change our SHG and SFG crystals for two crystals of, respectively, type I (LBO or $KNBO_3$ for instance) for SHG, and type II (BBO) for SFG. Another solution is to keep the KTP II SHG and LBO I SFG crystals but insert between the two NL crystals an additional dichroic wave-plate to restore the suitable polarization states. With this new configuration we could expect to double the SFG power amount. Finally, with proper coatings on the mirrors (end mirror HR@1064 + 532 + 355 nm, folding mirror HR@1064 + 532 and AR@355 nm) we could take advantage of the double-pass into the tripling crystal leading to quadrupling the UV output power. After all optimizations, we could expect theoretically more than 100 mW of UV cw power. It must be pointed out that at such NL conversion level the reappearance of large pulse broadening due to high NL (SHG + SFG) losses seems to be unavoidable exactly in the same manner as in the SHG configuration [4]. So if we want to preserve the short pulse duration, 10 mW of UV power after optimizations is a more realistic estimation.

In a second step we observed a self Q-switched mode-locked regime (Fig. 2). This Q-switched regime is obtained by slightly detuning the frequency of the driver of the acousto-optic modulator from the laser cavity length. The Q-switched envelope consisted in 0.7-μs-duration bursts at 50 kHz repetition rate modulating the 150 MHz ps pulse-train. The pulse duration deduced from IR autocorrelation was about 60 ps at 355 nm wavelength. With this configuration we obtained 1.2 mW of average power in the UV corresponding to a maximum peak power of 4 W. With suitable optimized elements we could expect Q-switched mode-locked ps UV pulses of more than 20 mW average power and 80 W peak power.

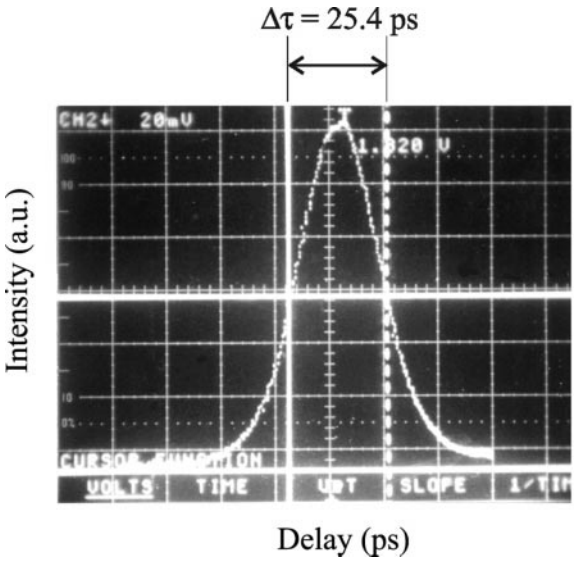

Fig. 2. Background-free autocorrelation measurements of infrared pulses by SHG. The autocorrelation full width at half maximum is 25.4 ps which corresponds to 18 ps IR pulse duration assuming Gaussian shape. Green and UV pulse duration of 12.7 ps and 10.4 ps, respectively, were deduced from this IR autocorrelation

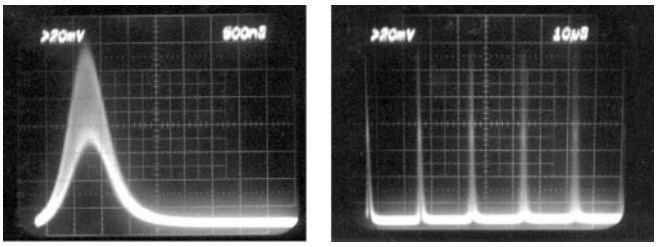

Fig. 3. Self Q-switched and mode-locked regime. The Q-switched envelope consisted of 0.7-μs-duration bursts at 50 kHz repetition rate modulating the 150 MHz ps pulse train

2 Conclusion

We have demonstrated both stable active mode-locking of a diode-pumped Nd:YAG laser and intracavity frequency-tripling. Pulses as short as 10 ps have been generated at 355 nm with 0.5 mW average power at 150 MHz repetition rate. Q-switched mode-locked regime is also shown leading to high-peak-power UV pulses. This UV light source is potentially interesting for many applications such as photobiology experiments involving time-resolved spectroscopy.

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