Passively mode-locked diode-pumped Nd:YVO₄ oscillator operating at an ultralow repetition rate

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Received April 18, 2003

We demonstrate the operation of an ultralow-repetition-rate, high-peak-power, picosecond diode-pumped Nd:YVO₄ passively mode-locked laser oscillator. Repetition rates lower than 1 MHz were achieved with the use of a new design for a multiple-pass cavity and a semiconductor saturable absorber. Long-term stable operation at 1.2 MHz with a pulse duration of 16.3 ps and an average output power of 470 mW, corresponding to 24-kW peak-power pulses, is reported. These are to our knowledge the lowest-repetition-rate high-peak-power pulses ever generated directly from a picosecond laser resonator without cavity dumping.

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OCIS codes: 140.3580, 140.4050.

Many applications in biology, such as fluorescence lifetime measurements, require pulsed laser sources emitting visible radiation. Because the fluorescence lifetime of numerous molecules is in the hundreds of picoseconds range, short pulses of several tenths of picoseconds are ideally needed from reliable laser sources. The repetition rate of the laser source used is also an important parameter in experiments with photon-counting detection chains: High repetition rates ensure fast acquisition of the fluorescence decay signal and allow the dynamic processes to be studied. However, if the repetition rate exceeds several megahertz, problems appear in the complex signal-processing devices during the acquisition of the data. Consequently, a repetition rate of approximately 1 or 2 MHz seems to be a good requirement for this kind of application. A classical approach to generating short visible pulses is the use of an oscillator operating at a fixed wavelength in the IR region (1064 nm) and the use of nonlinear processes to reach the visible range. But producing picosecond pulses in the IR region at a repetition rate of 1 MHz is not straightforward: The first alternative is the development of a *Q*-switched microchip laser.¹ However, in such systems pulse durations shorter than 100 ps with peak power that is high enough cannot be directly achieved without the use of amplification stages. The second alternative is the development of a mode-locked laser oscillator. However, those sources typically operate at several tenths of megahertz if the cavity size does not exceed several meters: For a mode-locked laser the repetition rate, f , of the pulses is fixed by the length of the cavity, L , according to the relation $f = c/2L$, where c is the speed of light. Therefore, to achieve a repetition rate as low as 1 MHz, the cavity length should be increased to 150 m. To achieve such long cavities, one has to introduce a multiple-pass cavity (MPC). In this Letter we report the operation of a passively mode-locked diode-pumped Nd:YVO₄ oscillator at a repetition rate lower than 1 MHz, emitting pulses of 16.3-ps duration with 470 mW of average power.

The classical Herriott-style MPC with two concave mirrors already widely used in a number of applica-

tions, especially as a delay line and for gas cells,² was recently used successfully to decrease the frequency of a Ti:Al₂O₃ mode-locked laser to 4 MHz.³ In such a MPC, however, the number of passes through the cavity is strictly defined by the distance between the two concave mirrors, whereas the beam entrance conditions, i.e., the initial distance r of the beam from the axis of the MPC and the angle θ , affect only the shape and the size of the spot pattern on the concave mirrors.²

The configuration of the MPC used in our system is in fact a version of the Herriott-style MPC folded in two with the help of two plane mirrors (Fig. 1). In our configuration the distance of both concave mirrors ($R = 2$ m) from the two plane mirrors is fixed at approximately the focal length of the concave mirrors, specifically at 105 cm. The beam is periodically focused and defocused after each reflection on the concave and plane mirrors, respectively. The overall ABCD matrix of the MPC in this case is of course no longer a unity transformation matrix.³ Thus the design of the cavity as a whole, already containing the MPC, is necessary. With the use of an ABCD matrix simulation program (simulating the MPC as a repeated series of 2-m concave mirrors separated by a distance of approximately 2 m) we observed no significant change in the intracavity beam properties

Fig. 1. Schematic of the laser cavity with the MPC consisting of four low-loss $R > 99.9$ mirrors (two folding plane mirrors and two concave mirrors with $R = 2$ m). OC, output coupler.

when the number of passes through the MPC was arbitrarily increased, if the design starts with a small cavity (without the MPC) providing a fairly collimated beam. The main advantage of this MPC configuration is that simply changing the alignment of the plane or the concave mirrors allows almost continual control of the number of passes through the MPC. The remarkable freedom of the easy control of the beam path in the MPC is the result of the use of the two plane mirrors. Each reflection on them, when the proper alignment is achieved, actually resets the beam into the MPC in such a way that it is forced to stay in the MPC for an arbitrarily large number of round trips. It is possible that the use of three or even four plane mirrors instead of two could supply better and easier control of the number of reflections, complicating of course the final setup. Additionally, in our case it was not necessary to drill or cut the concave mirrors for the entrance and exit of the beam.³

In this system a 5-mm-long, 0.1%-doped Nd:YVO₄ crystal is pumped by a fiber-coupled laser diode at 808 nm of 15-W maximum power. Passive mode locking is achieved with the use of a semiconductor saturable-absorber mirror^{4,5} (SESAM) with a 6.1% modulation depth. During the alignment of the MPC inside the laser cavity, various repeated beam spot numbers and patterns were observed, resulting each time in the operation of the system on a number of different frequencies. Specifically, we achieved stable operation at 20, 12, 5, 3, 2.3, and 1.2 MHz by simply adjusting the alignment of the MPC. For the case of 1.2 MHz an asymmetric repeated elliptical beam spot pattern on the two concave mirrors (Fig. 2) and a corresponding pattern on the two plane mirrors provided a cavity length of approximately 121 m after 56 round trips inside the MPC. Stable, single-mode operation at a repetition rate as low as 1.2 MHz with a pulse duration of 16.3 ps and average power of 470 mW, corresponding to 392-nJ pulse energy and 24-kW peak power, is reported. The beam quality is close to the diffraction limit, with M^2 slightly greater than 1.1. Figure 3 shows a photograph of the pulse train measured with the help of a fast photodiode. Small peaks appearing just after the pulses correspond to relaxation peaks of the photodiode. Figure 4 shows the autocorrelation trace of the pulses and the respective Gaussian fit curve. The FWHM of the Gaussian curve is 23 ps, corresponding to a 16.3-ps pulse duration. The stability of the system for continuous operation during a period of several weeks was satisfactory.

The output power was seriously confined by diffraction losses at the edges of the MPC mirrors. When the SESAM was replaced with a second output mirror of the same transmission as the first one and by comparison of the power of the two outputs, the overall losses in the MPC (including the diffraction losses) were measured to be 30%. At higher frequencies, i.e., when fewer passes through the MPC and simpler beam spot patterns on the mirrors were chosen, higher output powers of several watts were possible (2.5 W for $f = 2.3$ MHz). On the other hand, operation lower than 1 MHz was also observed at 884 and 650 kHz.

However, in those frequencies severe multiple pulsing instabilities did not allow long-term operation. Nevertheless, we strongly believe that reduction of the frequencies far lower than 1 MHz is manageable with the use of this MPC configuration, if the cavity is designed from the beginning for such a purpose.

At such low frequencies and high intracavity energies and peak powers careful design is crucial to avoid malfunction of the SESAM. The value of the modulation depth of the SESAM, ΔR , the tendency for Q-switching mode locking (QML) and multiple pulsing instabilities that the SESAM introduces, and of course its damage threshold are the key points to consider in the correct design of the cavity. Generally, in

Fig. 2. Photograph of the MPC concave mirrors with the beam spot pattern on them for the 1.2-MHz repetition-rate mode-locked Nd:YVO₄ laser.

Fig. 3. Oscilloscope trace of a fast photodiode, showing the mode-locked laser pulse train of 1.2 MHz of 810-ns separation.

Fig. 4. Intensity autocorrelation trace showing a pulse duration of 16.3 ps assuming a Gaussian intensity profile.

passively mode-locked solid-state lasers with weak gain saturation, such as a Nd:YVO₄ laser, an initial fluctuation of the intracavity power into the cw regime monotonically increases until steady-state mode locking is reached if and only if⁶

$$\kappa P \ln(m_i) > T_r/T_c, \quad (1)$$

where m_i represents the number of the initially oscillating modes, P can be approximated by the steady-state intracavity power, T_r is the round-trip time, T_c is the effective correlation time defined by the inverse 3-dB full width $\Delta\nu_{3\text{dB}}$ of the first beat note of the free-running laser, and κ is a characteristic of the nonlinear device (SESAM) that gives the change in round-trip power gain per unit intracavity power.⁶ Because the values of P , m_i , and T_c should be considered fixed for a specific system, for very long cavities, i.e., large T_r values, the previous condition becomes demanding. Since the left-hand factor in condition (1) approximates the effective modulation depth, ΔR ,⁶ of the SESAM, choosing a sufficiently large value for ΔR is crucial. For $T_r \approx 1 \mu\text{s}$ and typical values of $\Delta\nu_{3\text{dB}}$ of approximately 1–10 KHz,⁶ a secure choice of ΔR is greater than 3%. Our system employs a SESAM with $\Delta R = 6.1\%$. On the other hand, a SESAM introduces a Q -switching tendency that can drive the laser into the unwanted QML regime. To obtain cw mode locking free of QML instabilities, one should ensure that the intracavity pulse energy E_p satisfies the condition

$$E_p > (F_{\text{sat},L} A_{\text{eff},L} F_{\text{sat},A} A_{\text{eff},A} \Delta R)^{1/2}, \quad (2)$$

where $F_{\text{sat},L}$ and $F_{\text{sat},A}$ are the saturation fluence of the gain medium and of the SESAM, respectively, and $A_{\text{eff},L}$ and $A_{\text{eff},A}$ are the effective laser mode in the gain medium and on the SESAM, respectively. Thus for a choice of large ΔR and for the fixed values of $F_{\text{sat},L}$ and $F_{\text{sat},A}$ the design of the cavity should provide values of $A_{\text{eff},L}$ and $A_{\text{eff},A}$ small enough and enough E_p that condition (2) is satisfied too. However, the saturation of the SESAM, i.e., the value of the factor $S = E_p/F_{\text{sat},A} A_{\text{eff},A}$ should always be kept below 20 to avoid multiple pulsing instabilities and damage to the SESAM.^{8,9} By properly choosing the pumping focusing optics and the cavity mirrors we were able to some extent to achieve the proper values of $A_{\text{eff},L}$ and $A_{\text{eff},A}$ so that all the previous conditions were simultaneously met. Thus, for the case of a 1.2-MHz system, i.e., for $P = 470 \text{ mW}$ with a 10% output coupler corresponding to $E_p = 3.5 \mu\text{J}$ for a double pass (under strong overlapping of the beams) through the Nd:YVO₄ crystal, i.e., $F_{\text{sat},L} = h\nu/4\sigma_L = 30 \text{ mJ/cm}^2$ and for a SESAM with $\Delta R = 6.1\%$ and $F_{\text{sat},A} = 70 \text{ mJ/cm}^2$

we chose a 1:1 two-lens focusing of the pump beam, providing a mode area in the crystal estimated to be approximately $A_{\text{eff},L} = 1.3 \times 10^{-3} \text{ cm}^2$ (pump fiber core diameter, 400 μm) and a 2-m focusing mirror on the SESAM providing a 770- μm mode diameter on the SESAM, corresponding to $A_{\text{eff},A} = 4.7 \times 10^{-3} \text{ cm}^2$. For the above values condition (2) is well satisfied, since $3.5 > 0.88$ and the saturation of the SESAM is $S = 10$.

In conclusion, operation of an ultralow-repetition-rate (lower than 1 MHz), mode-locked Nd:YVO₄ oscillator has been demonstrated with a simple and low-cost design. Stable long-term operation at 1.2 MHz with a pulse duration of 16.3 ps and 24-kW peak power was achieved. To the best of our knowledge this is the lowest repetition rate ever reported for a completely passively mode-locked laser with no use of complicated and expensive techniques such as cavity dumping. In preliminary experiments on frequency doubling, conversion efficiency into 532 nm as high as 50% was achieved with the use of 10-mm-long periodically poled KTP crystal. These results are promising for the further use of this source for the generation of short visible pulses for fluorescence lifetime measurements.

This research was partially supported by the research program POLA from the Contrat Plan Etat Région (2000–2006) (French State and Conseil Général de l'Essonne). D. N. Papadopoulos worked on this project under a Marie Curie fellowship. S. Forget's e-mail address is sebastien.forget@iota.u-psud.fr.

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