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Kerr-lens mode-locked visible transitions of a Pr:YLF laser

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We report what is to our knowledge the first Kerr-lens mode locking of the visible transitions at 607 and 639 nm in Pr³⁺:YLF. Initiation of the self-mode locking took place through the use of liquid saturable absorbers to establish saturation parameters, and it was carried out in optimized all-solid-state laser configurations (but with gas-laser pumping) with thin (<100-μm) colloiddally colored CdSeS glasses. Pulse durations were in the range of 8–10 ps.

The past few years have seen considerable research interest redirected toward solid-state lasers, in particular as convenient, wavelength-versatile sources, operating cw at room temperature. Of marked importance has been the ability of these gain media to exhibit self-formation of ultrashort pulses through the mechanism of spatial intensity-dependent refractive-index Kerr-lens mode locking, generating pulse durations from the picosecond to the femtosecond regimes and approaching pulse widths determined by the gain bandwidth of the various active media.^{1–7} To date, however, operation has been essentially limited to systems that operate in the near-infrared spectral range. There are clear advantages in using systems that operate directly in the visible spectrum without recourse to nonlinear generation techniques. Recently considerable research has been undertaken with Pr³⁺-doped crystals,^{8–10} which have been shown to exhibit laser action in several bands at wavelengths in the visible.^{11,12} This resulted in the first report of cw laser action in Pr³⁺:YLF,⁸ which, with its relatively high efficiency, convenient pump bands, and moderately broad transitions, makes it attractive as a picosecond solid-state laser source of visible radiation. In this Letter we report what is to our knowledge the first Kerr-lens mode locking of Pr³⁺:YLF on two of the visible transitions, at 639 and 607 nm.

The spectroscopy,¹³ cw laser performance, and optimization of Pr³⁺:YLF has been described previously.⁸ A 9-mm-long, 6-mm-diameter Pr:YLF crystal, cut with Brewster-angled facets such that the electric-field vector was perpendicular to the crystal *c* axis, was used throughout this series of investigations. The crystal, of 0.8 at.%, which was grown at the Center for Research and Education in Optics and Lasers, was indium clad and mounted in a copper jacket that could be water cooled. Figure 1 shows a schematic of the cavity configuration primarily used throughout these investigations. Because a saturable absorber was used to initiate the mode-locking process, a ring geometry, with its associated lower loss, was used in preference to a linear cavity

configuration. However, for some initial investigations, a linear geometry was employed. An argon-ion laser that operates at 476 nm was used as the pump source. Although other argon-ion pump lines have been shown to be more efficient in the pumping of the 639-nm (³P₀–³F₂) transition in Pr³⁺:YLF,⁸ greater average power is obtainable with 476-nm excitation. A conventional astigmatically compensated cavity was employed. With a 10-cm lens, the pump radiation was focused into the Pr³⁺:YLF crystal, which was placed at the common focus of the 10-cm, nominally 100% reflecting (600–650 nm) mirrors M1 and M2. As much as 1.7 W of power at 476 nm was absorbed in the crystal.

In a conventional four-mirror astigmatically compensated z-fold linear cavity constructed with mirrors M1, M2, M5 (100% reflecting at 600 nm) and M6 (96% reflecting at 639 nm) (Fig. 1), and including a dielectric-coated tuning wedge for wavelength selection, we obtained as much as 150 mW of output power at 639 nm for a maximum pump power of 1.7 W (threshold 200 mW), which indicated a slope efficiency of 10%. This is comparable with previous results.⁸ Thresholds as low as 60 mW have been obtained with this system. On the 607-nm (³P₀–³H₆) transition the threshold was 400 mW of pump power, and 110 mW of output was obtained with a slope efficiency of 8.5%, which is considerably greater than that originally reported by Sandrock *et al.*⁸ Through

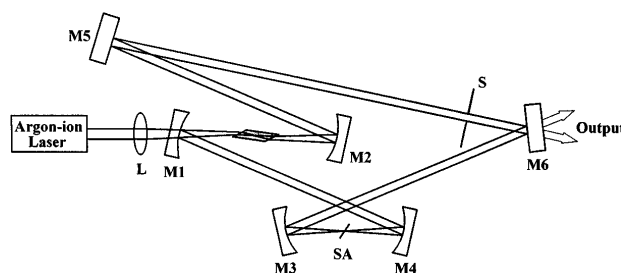


Fig. 1. Schematic of the ring cavity configuration used in the Kerr-lens mode locking of the Pr:YLF laser. L, lens; S, slit; SA, saturable absorber.

the removal of the dielectric tuning wedge and the incorporation of two intracavity prisms, simultaneous operation was possible on the 607- and 639-nm transitions with average powers of 18 and 34 mW, respectively, for 1.7-W absorbed pump power in a cavity with unoptimized output coupling.

Initially, mode locking of the linear laser cavity operating at 639 nm was undertaken with a conventional prism acousto-optic modulator in front of the 100%-reflecting plane mirror. Pulse durations in the 100-ps regime were obtained. Passive mode locking also was demonstrated by the moving-mirror technique.¹⁴ In this case, the 100% plane reflector M5 was vibrated at 12.5 Hz, generating contiguous trains of pulses, with minimum recorded pulse widths of 55 ps.

In general, the Kerr-lens mode locking of solid-state lasers requires a start-up mechanism.¹⁵ Most commonly, this can be undertaken by rapidly varying the Q of the cavity by vibrating a cavity element, by using an active modulator, or through passive techniques such as the application of a fast saturable absorber in liquid- or solid-state format. Clearly, passive techniques are simpler to implement. In the research reported here we employed saturable absorbers to initiate the pulse formation process, which was further enhanced by the Kerr-lens mechanism to produce pulse widths substantially shorter than would be expected from the action of the fast saturable absorber alone. Without Kerr-lens mode locking, the pulse durations in the absence of any additional pulse compression mechanism (i.e., gain saturation) are of the order of the recovery time of the fast saturable absorber, which was ~ 100 ps in the research described here.

The saturable absorbers were placed at the common focus of the 50-mm radius-of-curvature, broadband (500–700 nm), nominally 100%-reflecting mirrors M3 and M4 (see Fig. 1). Initially, liquid saturable absorbers were used to establish the low-level loss, absorption cross section, and recovery time requirements of the appropriate absorber. These dyes were dissolved in ethylene glycol and flowed in a 200- μ m-thick jet stream at the common focus of the mirrors. The saturable absorber used, 1,3'-diethyl-4,2'-quinolyloxacarbocyanine iodide, has an absorption cross section of 0.16×10^{-16} cm² at 639 nm and a recovery time of 120 ps in ethylene glycol.¹⁶ For an absorber concentration of 1.7×10^{-5} M, stable cw mode-locked operation was observed with pulse durations near 10–15 ps. The stability and pulse selection of the mode-locked operation were enhanced by the presence of an intracavity slit, which suggested that Kerr-lens mode locking was playing a role in the pulse formation. This view also is reinforced by consideration of the concentration and recovery time of the absorber with a low-level absorption of 0.4%, which, in the absence of the Kerr-lens mechanism, would be unable to generate pulses of the minimum durations recorded. Intermittent dropout of the mode locking was observed in the absence of the slit. In optimized mode-locked operation, typical average output powers were in the range of 25 mW at pulse repetition

rates in the 100-MHz regime and pulse durations near 10 ps.

For operation at 607 nm, the concentration of the absorber was reduced such that the low-level absorption was near 0.1%, i.e., fairly insignificant for conventional passive mode-locking purposes. In addition, mirrors M3 and M4 were replaced by ones of similar curvature but with a 100%-reflective coating that covered 500 to 620 nm, to suppress the 639-nm line. The mode-locking behavior was similar to that for the 639-nm operation, with an average mode-locked output power level of 24 mW obtained for an absorbed pump power of 1.5 W and minimum recorded pulse widths near 10 ps (Fig. 2). The threshold pump power for mode-locked operation was 1.1 W, and cw mode-locked trains were obtained up to the maximum absorbed pump power. The spectral width of the mode-locked output for both transitions was limited by the 0.1-nm resolution bandwidth of the spectrum analyzer.

Having established the saturable absorber requirements (i.e., low-level absorbance and recovery time of less than ~ 150 ps), we replaced the liquid absorber with a solid. Cadmium selenide sulfide (CdSe_{1-x}S_x) colloidal colored glasses previously have been shown to operate as ultrafast saturable absorbers¹⁷ and to initiate Kerr-lens mode locking in picosecond solid-state lasers.¹⁸ The transmission characteristic of a Schott Glass RG 610 filter, opti-

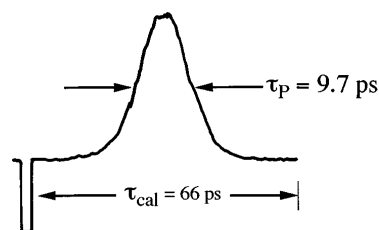


Fig. 2. Intensity autocorrelation of a representative pulse width recorded at 607 nm from the Kerr-lens mode-locked Pr:YLF laser, initiated by means of a liquid saturable absorber. The deconvolved pulse width is indicated.

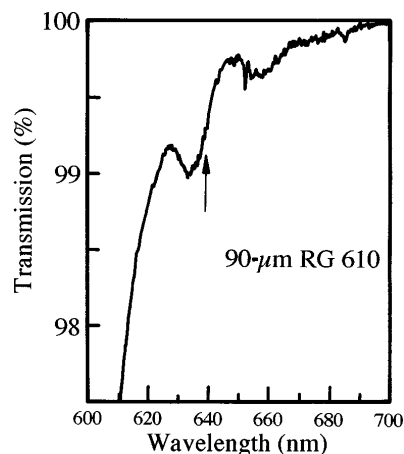


Fig. 3. Transmission characteristic over the spectral region of interest of a 90- μ m-thick Schott Glass RG 610 filter used as a saturable absorber for the Pr:YLF laser at 639 nm.

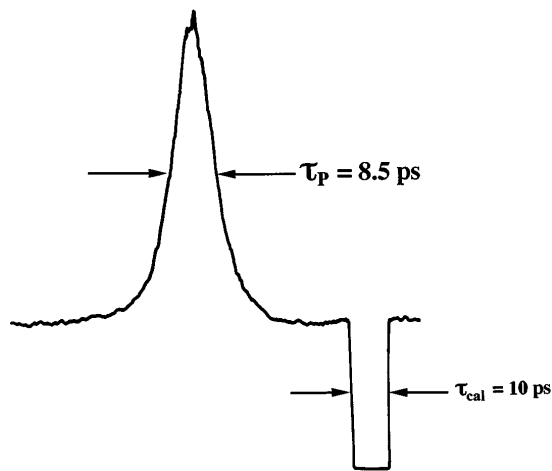


Fig. 4. Background-free autocorrelation of 8.5-ps pulses generated through the mode locking of the 639-nm Pr:YLF transition, initiated by the incorporation of an intracavity solid-state colloiddally colored glass saturable absorber.

cally polished to a thickness of $\sim 90 \mu\text{m}$, is shown in Fig. 3. At 639 nm this filter had a measured absorption of $\sim 0.7\%$. In the region of the 639-nm Pr^{3+} transition, as shown in Fig. 3, the transmission exhibited a resonant absorption. This phenomenon most likely is due to the quantum confinement nature of the semiconductor microcrystallites in the glass host.¹⁹ For the cavity configuration in the absence of the solid-state saturable absorber, the threshold pump power was 390 mW; on the insertion of the sample, the threshold pump power rose to 480 mW.

For pump powers of 1.4 W and above, continuous stable self-mode-locked trains of pulses were obtained. Below this threshold, simultaneous Q switching and mode locking were observed, with a characteristic period of the damped Q -switched trains near $20 \mu\text{s}$. In true cw mode-locked operation, an average output power of 30 mW and minimum pulse widths (at $\sim 125 \text{ MHz}$) near 8 ps were generated (Fig. 4). When cadmium selenide sulfide, colloiddally colored, polished (~ 50 – $100 \mu\text{m}$ thick) glasses with equivalent low-level absorptions were used, which did not exhibit the excitonic feature, mode locking was unstable, and the outputs tended to be in the simultaneously Q -switched and mode-locked regime.

In conclusion, we have reported what is to our knowledge the first Kerr-lens mode locking of a solid-state laser that operates on two transitions in the visible at 639 and 607 nm, initiated through the use of a solid-state saturable absorber. Successful mode locking was also demonstrated with a liquid absorber that was used initially to determine the required saturation parameters of an intracavity absorber for the laser system. The generated pulse widths tended to be in the region of 8–10 ps, which are considerably longer than the predicted minimum of $\sim 1 \text{ ps}$ supportable by the gain bandwidth of the active transitions. The introduction of dispersion correction may be necessary to yield re-

duced pulse widths. Through the appropriate cavity configuration,²⁰ it also should be possible to obtain self-starting, self-maintaining Kerr lens mode locking of the visible transitions of Pr:YLF. In addition, the use of a diode-pumped upconversion pumping scheme should permit the realization of an all-solid-state mode-locked visible laser system.

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Note added in proof: Using optimized cavity configurations, we have observed self-starting, self-mode-locked operation of the Pr:YLF laser, with the generation of picosecond pulses.

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