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Visible continuous-wave laser transitions in Pr^{3+}:YLF and femtosecond pulse generation

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Fourteen new cw, visible laser transitions have been observed in Pr^{3+}:YLF, several of which have exhibited modest tunability. The $^3P_0 - {}^3H_6$ transition near 613 nm has been shown to have a tunable linewidth of ~1 nm, which, for the first time to our knowledge, has permitted the direct production of femtosecond pulses in the visible from a cw solid-state laser by Kerr-lens mode locking. © 1996 Optical Society of America

Praseodymium (Pr^{3+}) as an active ion dopant for solid-state laser crystals and glasses provides a wide choice of visible laser sources. To date, 16 principal laser transitions have been identified in Pr^{3+}-activated materials ranging from the blue[1] to the mid infrared.[2] Until quite recently the main excitation mechanisms were pulsed by either flash lamps or dye laser pumps. As a result of the relatively narrow yet strong absorption channels in Pr^{3+}, flash-lamp pumping has tended to be inefficient, and despite sensitization methods[3] and improved pumping configurations[4] the efficiency has remained low.[5] The narrow absorption features of Pr^{3+}-doped crystals are particularly amenable to Ar^{3+}-ion laser pumping, and recently efficient room-temperature cw laser action was reported with Pr^{3+}-doped oxide[6] and fluoride[7] crystals. Efficient laser operation at six wavelengths was demonstrated in Pr^{3+}:YLF,[7] and Kerr-lens mode locking (KLM) of two of these lines at 607 and 639 nm was achieved, initiated through the use of liquid and solid-state saturable absorbers.[8,9] The potential of Pr:YLF to support picosecond pulses employing the 639-nm transition has also been demonstrated in a flash-lamp-pumped system, which was Q switched and mode locked by an acousto-optic modulator.[10] Although in that rather inefficient scheme[10] no picosecond pulse measurements were made, the potential of picosecond operation was inferred from the measured spectral linewidths. Pr^{3+}:YLF clearly provides an attractive, room-temperature solid-state cw source of picosecond pulses at diverse wavelengths throughout the visible. To date the spectral width of the reported lasing transitions has limited mode-locked operation to the picosecond regime. However, as a result of Stark splitting within the energy levels there are many more allowed transitions in Pr^{3+}:YLF. We have identified 14 new room-temperature cw lasing transitions in this crystal, several of which permitted modest tunability, with adequate bandwidth to support subpicosecond pulses. Consequently we have demonstrated KLM and femtosecond pulse generation by using one of these transitions, which we also report here.

The Pr^{3+}:YLF crystal was grown at the Center for Research and Education in Optics and Lasers, University of Central Florida. It was doped with 0.8 at.% Pr and was 9.8 mm long, 6 mm in diameter, cut with Brewster-angled facets, clad with indium, and mounted in a copper jacket, which could be water cooled. Throughout, the crystal was maintained at a temperature of 15 °C. For the majority of the results presented here the crystal employed was cut such that the pump and the lasing electric field vectors were parallel to the crystal c axis. However, an additional, similarly doped crystal was also cut, with the electric field vector perpendicular to the c axis for access to other transitions. Pumping was achieved with an argon-ion laser operating at 476 nm, with a maximum of 1.5 W of power absorbed by the crystal. To investigate the laser action of the various allowable transitions in Pr^{3+}:YLF, we used a conventional three-mirror astigmatically compensated cavity. Pumping was undertaken with a 100-mm convex lens via the 100-mm radius-of-curvature folding mirror of the cavity. A 75-mm broadband retroreflecting mirror was employed in the folding section to return transmitted pump radiation to the crystal. The cavity was completed with a plane, nominally 100% broadband reflector. Tuning was achieved with a Brewster-angled intracavity SF10 prism, and, to enhance wavelength discrimination, we introduced a variable-width vertical slit into the cavity. No attempt was made to optimize the output coupling or the lasing efficiency of the system in these initial investigations of the allowable lasing transitions. The lasing spectra were recorded on a scanning grating spectrum analyzer with a spectral resolution of 0.1 nm, which in most circumstances was insufficient to resolve the transition linewidths.

Table 1 shows the experimentally measured wavelengths of 18-cw laser transitions in Pr^{3+}:YLF from 600 to 722 nm (there are also a considerable number lying outside this range). The majority of the lasing transitions investigated were for the electric field parallel to crystal c axis, except for those indicated by σ,

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Table 1. Experimentally Measured CW Lasing Transitions in Pr^{3+}:YLF[a]

Transition	Frequency (cm^{-1})[b]	Wavelength (nm)
$^3P_0 - {}^3H_6$	16 546	604.4
σ	*16 466*	*607.3*
σ	16 406	609.2
	16 303	613.0*
$^3P_1 - {}^3F_2$	16 221	615.8
	16 204	618.0
	16 071	620.1*
$^3P_0 - {}^3F_2$	15 659	638.8
σ	*15 637*	*639.5*
	15 518	644.4
$^3P_1 - {}^3F_3$	14 905	670.3
$^3P_0 - {}^3F_3$	*14 339*	*697.7*
	14 174	705.5
$^3P_1 - {}^3F_4$	14 280	699.4
$^1I_6 - {}^3F_4$	14 105	708.2
$^3P_0 - {}^3F_4$	13 928	719.5
	13 877	*720.9**
	13 723	722.2

[a]The designated transitions were determined from Refs. 11 and 12. All transitions are for the electric field vector parallel to the crystal c axis, except those indicated by σ, for which it is perpendicular. The wavelengths in italics were reported previously.[7] Tunability recorded within the transition is indicated by an asterisk.

[b]From Refs. 11 and 12.

for which the orthogonal polarization was employed. The recorded wavelengths were in good agreement with those expected from the data of previously measured energy levels of Pr^{3+}:YLF.[11,12] Four of the transitions listed in Table 1 were reported previously[7]; however, of these, one at 720 nm was shown to be tunable from 720.5 to 721.0 nm, which had not been previously reported. Two additional transitions were shown to be tunable; they are indicated by asterisks on the transition wavelengths in Table 1. These nominal transitions at 613 and 620.1 nm were tunable from 612.4 to 613.5 nm and from 620.1 to 620.5 nm, respectively. Vibronic structure in the emission spectrum of the $^3P_0 - {}^3H_6$ transition near 613 nm had been reported by Esterowitz *et al.*[11] Figure 1 shows the spectral location of the lasing transitions relative to the amplified spontaneous emission spectrum obtained from the sample under 476-nm excitation. No attempt was made to optimize the output coupling or the slope efficiency of these various lasing transitions, which for typical operation with ~1% output coupling were in the range 5–10%. The limited availability of the output couplers limited the optimization of output power. Over the spectral range examined the output mirrors employed had a transmission that varied from 1% to 3%. For transmission of <0.5%, threshold pump powers of <100 mW were obtained, for example, on the 613- and 639-nm transitions. Within a multiplet, various efficiencies related to oscillator strength were observed. For example, with a 1% output coupler the 604.4- and 607.3-nm lines had slope efficiencies of 5%, whereas the 613-nm line exhibited an output efficiency of greater than 10%. Typical thresholds for these lines were 200, 450, and 500 mW, respectively. However, it should be noted that these measurements were not undertaken for identical configurations, with the 607-nm system employing a rod cut with the c axis perpendicular to the electric field vector. With optimization on the 607.3-nm line we have obtained slope efficiencies of 9%. Simultaneous operation at 522 and 604 nm (and at other wavelength combinations) was possible with a combined average power of 40 mW, although with reduced overall efficiency in the transitions compared with that for single-line operation, as would be expected as a result of the upper states' being common to the lasing transitions.

The relatively high efficiency of the 613-nm transition (>10%) together with its vibrationally broadened spectral width of 1 nm makes it a potentially attractive source of femtosecond pulses. We examined KLM of the Pr^{3+}:YLF laser at this wavelength, using the experimental setup shown in Fig. 2. A four-mirror, astigmatically compensated symmetrical cavity was used, which incorporated the Pr^{3+}:YLF rod placed between 100% reflecting, 100-mm radius-of-curvature mirrors. A symmetrical pumping scheme, employing 100-mm focal-length lenses pumping through the folded section mirrors, was used, and at maximum 1.5 W of 476-nm pump radiation was absorbed. The cavity was completed by a 100% reflecting and a 3% transmitting output coupler at 613 nm. A vertical slit was incorporated into the cavity to enhance the intensity discriminating nature of the KLM process. Dispersion compensation was achieved through the introduction of a pair of F2 prisms. Self-mode locking was initiated by a mechanical perturbation of the end mirror, although stable laser operation was achievable only for several minutes as a result of the critical requirement of the cavity alignment at the relatively low pump power available. Alternatively and more reliably we initiated KLM by placing the

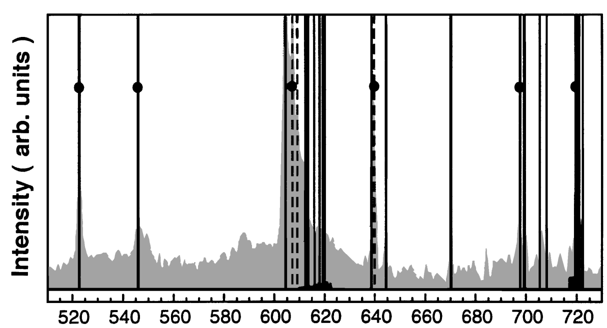

Fig. 1. Lasing transitions and amplified spontaneous emission in Pr^{3+}:YLF under 476-nm excitation. Previously observed[7] laser transitions are indicated by filled circles. All transitions are for E parallel to the crystal c axis, except those indicated by the dashed lines.

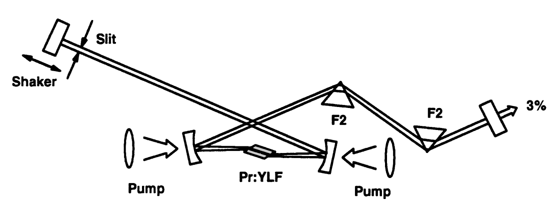

Fig. 2. Schematic of the cavity configuration of the femtosecond KLM Pr^{3+}:YLF laser.

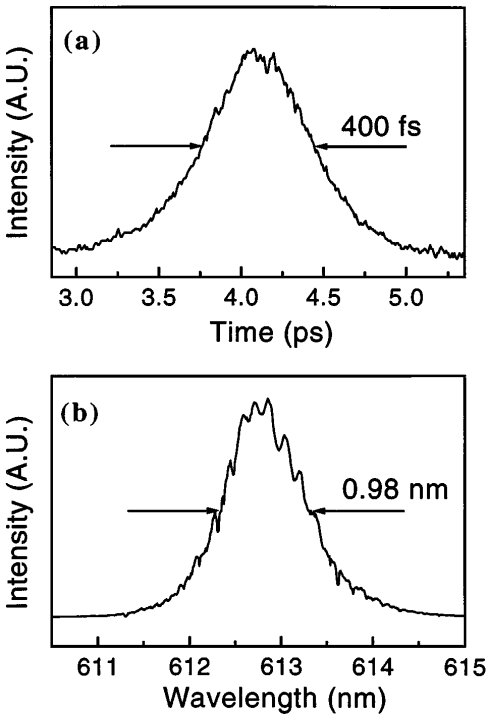

Fig. 3. (a) Autocorrelation trace (deconvolved pulse duration indicated) and (b) corresponding spectrum of 400-fs pulses generated by KLM of the 613-nm transition in Pr^{3+}:YLF.

high-reflecting cavity end mirror on a continuously vibrating assembly. Figure 3 shows a representative autocorrelation trace and corresponding spectrum of 400-fs pulses generated by this laser. The 0.98-nm bandwidth of the mode-locked pulses was limited by the effective bandwidth of the transition as measured above. Employing a 3% output coupler and for an absorbed pump power of 1.5 W, we obtained 45-mW average power in the mode-locked output at 613 nm.

In conclusion, we have reported relatively efficient cw laser action in 14 transitions in Pr^{3+}:YLF, for the first time to our knowledge. Several of these transitions exhibited modest tunability, and in particular a ${}^3P_0-{}^3H_6$ transition near 613 nm was shown to have a bandwidth of ~ 1 nm, which was capable of supporting femtosecond pulses. This transition was mode locked by the KLM technique, generating pulses of 400 fs. We believe that this is the first demonstration of femtosecond pulse generation in a visible solid-state laser. The generated pulses were effectively limited by the gain bandwidth of the transition, and to obtain shorter pulses it may be necessary to investigate other transitions in Pr^{3+}:YLF or to examine Pr^{3+} transitions in other host materials.

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