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# Self-mode-locked $\text{Cr}^{3+}:\text{LiCaAlF}_6$ laser

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A cw-pumped  $\text{Cr}^{3+}:\text{LiCaAlF}_6$  laser has been configured to produce chirp-free, self-mode-locked pulses of 170-fs duration. The prism-dispersion-compensated cavity contained no active or passive mode-locking device. Mode locking was accomplished through self-focusing in the  $\text{LiCaAlF}_6$  crystal with 2.5 W of red light from a cw krypton laser. Rocking of the output coupler initiated the ultrashort-pulse formation.

Since the first report of sustained self-mode-locking of a Ti:sapphire laser in 1990,<sup>1</sup> remarkable progress has been made in the development of this solid-state laser as the primary source of widely tunable, ultrashort pulses in the near infrared and other spectral regions through nonlinear optical interactions.

A number of techniques that resulted in mode-locked operation of Ti:sapphire in the femtosecond regime have been reported, but it is now evident that all of these rely on self-focusing, caused by the optical Kerr effect within the laser rod itself, to induce spectral pulse broadening and short-pulse generation.<sup>2</sup> The different methods amount to variations in starting mechanisms.<sup>3</sup> Common to all subpicosecond Ti:sapphire lasers are elements that compensate for intracavity group-velocity dispersion. When due account is paid to minimizing higher-order dispersion terms, pulses as short as 20 fs have been produced directly from the laser.<sup>4</sup> The simplest of these mode-locking schemes has no active or passive nonlinear elements within the cavity to induce modulation. In this case the mode locking is typically initiated by a transient perturbation on the system such as moving or vibrating a mirror.

The generic nature of the mode-locking mechanism in Ti:sapphire makes it attractive to investigate other tunable solid-state lasers. The new class of chromium-doped, colquiriite-structured solid-state lasers<sup>5,6</sup> are promising candidates as femtosecond laser sources and amplifiers because of their broad fluorescence bandwidths in the near infrared, long spontaneous emission lifetimes, and absorption bands in the red spectral region. We have recently reported the production of 150-fs mode-locked pulses in the 800–880-nm range from both  $\text{Cr}^{3+}$ -doped  $\text{LiSrAlF}_6$  (Cr:LiSAF) and  $\text{Cr}^{3+}$ -doped  $\text{LiSr}_{0.8}\text{Ca}_{0.2}\text{AlF}_6$  (Cr:LiSCAF) lasers<sup>7,8</sup> using a cavity that incorporated a prism pair arrangement to compensate for group-velocity dispersion. In these cases an acousto-optic modulator was employed to initiate mode-locked operation, but because the pulses were of ultrashort duration, the mode locking was attributed to self-focusing in the laser rod analogous to that in Ti:sapphire.<sup>2,3,9</sup> This occurs despite the fact that the nonlinear refractive index  $n_2$  of these fluoride crystals is a factor of 4 smaller than the  $n_2$  of

Ti:sapphire. Recently sub-100-fs pulses have been reported<sup>10</sup> from a forsterite ( $\text{Cr}^{4+}:\text{BeAl}_2\text{O}_4$ ) laser at 1260 nm, similarly operated with an acousto-optic modulator and incorporating prism compensation. In this Letter we report what is to our knowledge the first cw-pumped, self-mode-locked operation of a  $\text{Cr}^{3+}:\text{LiCaAlF}_6$  (Cr:LiCAF) laser. No intracavity mode-locking element was needed to sustain pulses in the femtosecond range when the laser was pumped by 2.5 W of red light from a cw krypton laser.

The fluorescence emission peak of Cr:LiCAF is centered at 763 nm with a peak cross section of  $1.2 \times 10^{-20} \text{ cm}^2$  and a spontaneous emission lifetime of 175  $\mu\text{s}$ .<sup>6</sup> A Cr:LiCAF laser was first reported by Payne *et al.*<sup>11</sup> with the use of cw krypton pumping. A strong absorption band peaking at  $\sim 630 \text{ nm}$  is well matched to the 647-nm output of the krypton laser. Tuning was demonstrated over the range 720–840 nm, and a 61% slope efficiency was measured.<sup>12</sup> Diode-laser-pumped operation has been achieved by Schepps *et al.*<sup>13–15</sup> with a pump threshold at approximately 13 mW of absorbed power from red diodes. Flash-lamp pumping of Cr:LiCAF has been reported by Payne *et al.*<sup>16</sup>

The Cr:LiCAF crystal used in this research was grown by the Czochralski pulling technique under conditions similar to those of Cr:LiSAF.<sup>7</sup> The crystal was doped during the growth process with 2 at. % of  $\text{Cr}^{3+}$ . Because of the decomposition of  $\text{CrF}_3$  during both initial heating and subsequent growth, the actual doping concentration is  $\sim 0.8 \text{ at. \%}$ . The crystal was cut at Brewster's angle for the light polarized parallel to the  $c$  axis, and the facets were polished by hand on a polymer sheet coated with  $0.3\text{-}\mu\text{m Al}_2\text{O}_3$  grit. The physical path length for the laser beam in the crystal was 15 mm.

The laser was configured in an X-fold cavity that contained the LiCAF crystal between two fold mirrors each with a radius of curvature of 10 cm. A standard, midband mirror set (nominally 800 to 900 nm) from a Schwartz Electro-Optics CW-Titan laser was utilized. The crystal was heat sunk by using a small thermoelectric cooler mounted on a water-cooled aluminum block. The thermoelectric cooler maintained the temperature of the crystal near 22°C. The laser was pumped by a Laser Ionics Model 1400-3K cw

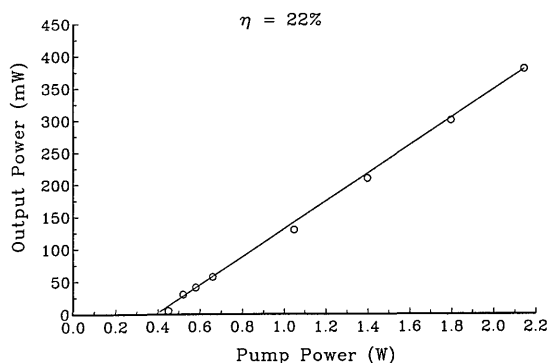


Fig. 1. Cw output power of the Cr:LiCAF laser versus the krypton laser pump power.

krypton laser operating on both the 647- and 676-nm red lines (power ratio 4:1). For the length of crystal and doping concentration employed the absorption of the pump beam was 98.5%.

Figure 1 shows the cw LiCAF laser output power as a function of the pump power. The threshold input power for cw lasing was found to be 450 mW, and the slope efficiency was measured to be  $\sim 22\%$  with a 4% transmission output coupler. No significant thermal dependence of the lasing efficiency was encountered during the cw measurements up to a pump power of 2.5 W, which gave an output power of 400 mW. This was in marked contrast with previous measurements<sup>7,8</sup> on LiSAF and LiSCAF crystals under similar heat-sinking conditions whereby the output power was found to drop at pump powers above 1.5 W. This is attributed to the fact that the reduction in fluorescence lifetime of the LiCAF crystal occurs at a much higher temperature<sup>17</sup> than the latter two crystals. However, differences in background absorption between the different crystals could also be significant. Wavelength tuning from 790 to 830 nm was achieved by using a birefringent filter in the cavity with a single midband mirror set. The tuning range was restricted by the bandwidth of the mirror coatings used in our laser, which shifts the lasing frequency to the long-wavelength side of the fluorescence peak.

In order to achieve mode-locked operation, a dispersion-compensating prism pair was placed between the crystal and the high-reflecting (HR) back mirror. Figure 2 is a schematic representation of the X-fold laser cavity configuration. The output coupler used had a transmission of 1% in the 800–900-nm range, and 60° prisms made from SF10 glass were used in the dispersion-compensating arrangement. The ultrashort-pulse formation was initiated by rocking the output coupler toward the LiCAF crystal, and when the output coupler was stationary the mode-locked pulses remained stable until a perturbation caused the laser to revert back to the cw state. Stable mode-locked operation was obtained when the crystal was pumped with 2.5 W of power from the krypton laser, with gave an average output power of 100 mW.

The mode-locked pulses were monitored by a real-time collinear autocorrelator that could be set up to either show a 1:3 intensity autocorrelation or give

interferometric data. The pulse was monitored by using a fast photodiode and a 0.25-m monochromator with a silicon array, and an optical multichannel analyzer was used to observe the spectral content of the pulses.

The separation between the prisms was set at 45 cm, which we previously found to be the optimum in compensating for self-phase-modulation in a Ti:sapphire crystal of similar length and group-velocity dispersion in the laser cavity. The autocorrelation trace recorded under these conditions is shown in Fig. 3(a). A fitted  $\text{sech}^2$  autocorrelation pulse shape with a 255-fs FWHM implies a laser pulse duration of 170 fs. The spectral content of the pulses shown in Fig. 3(b) has a bandwidth of 4.5 nm (FWHM). The time-bandwidth product  $\delta\tau_p\delta\nu$  of 0.36 (compared with the Fourier transform limit of 0.32) indicates that the pulses are almost chirp free as is evidenced in the interferometric autocorrelation traces of Fig. 3(c). In the absence of a tuning element such as a birefringent filter, the laser operated at a wavelength of  $\sim 800$  nm, which was imposed by the overlap of the fluorescence bandwidth of the LiCAF crystal and the reflecting bandwidth of the mirror set used. It is therefore likely that the bandwidth of the ultrashort pulses was partly constrained by the mirror set employed in this research. Optimization of the separation between the prisms to compensate for group-velocity

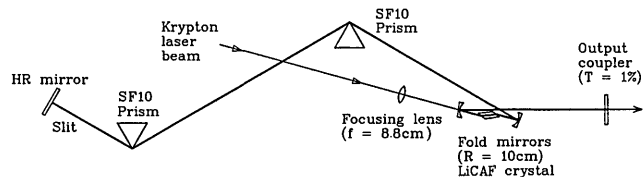


Fig. 2. Schematic layout of the self-mode-locked Cr:LiCAF laser.

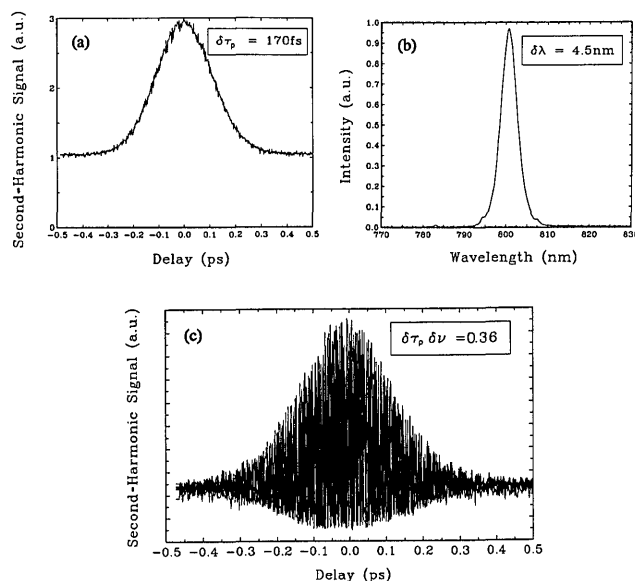


Fig. 3. Mode-locked characteristics of the Cr:LiCAF laser: (a) autocorrelation trace, (b) spectral bandwidth, and (c) interferometric autocorrelation trace of dispersion-compensated pulses.

dispersion should also improve the pulse width of the laser.

In conclusion, we have demonstrated the self-mode-locked operation of  $\text{Cr}^{3+}:\text{LiCAF}$ , a colquiriite laser crystal. A continuous train of 170-fs pulses at a repetition frequency of 90 MHz was achieved with no extra mode-locking elements other than the nonlinear refraction in the laser crystal. It is anticipated that with better crystal facet polishing and mirror coatings with bandwidth matching more closely the fluorescence bandwidth of the LiCAF crystal, as well as better optimization of the group-velocity dispersion in the cavity, the performance of the laser would be greatly improved for higher power and shorter pulse width. In comparing  $\text{Cr}:\text{LiCAF}$  with  $\text{Cr}:\text{LiSAF}$ , the principal difference is the shorter emission wavelength for  $\text{Cr}:\text{LiCAF}$ .<sup>12</sup> The fluorescence bandwidth is narrower, and the peak emission cross section is nearly a factor of 4 smaller. However, the spontaneous emission lifetime is longer (170  $\mu\text{s}$  compared with 67  $\mu\text{s}$  at room temperature) and does not decrease until a much higher temperature. This has allowed pumping of the LiCAF with 2.5 W from a krypton laser, which results in self-mode-locked operation. The good lasing efficiency of the crystal and its absorption band in the red wavelength give this laser potential advantages over  $\text{Ti}:\text{sapphire}$  as a diode-pumped ultrashort-pulse source.

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