

Generation of 150-fs tunable pulses in Cr:LiSrAlF₆

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We report ultrashort-pulse generation in a Cr:LiSrAlF₆ solid-state laser. The 15-mm crystal was pumped by 1 W of red light from a cw krypton laser and actively mode locked at an 82-MHz repetition frequency using an acousto-optic modulator. Wavelength tuning was demonstrated over the range 800–880 nm, limited by the bandwidth of the optical components. By including intracavity dispersion compensation, pulse widths of 150 fs were obtained with evidence that self-phase modulation contributes to spectral broadening and pulse shortening.

In the past few years we have witnessed rapid developments in ultrashort-pulse generation in solid-state lasers. In particular, tunable subpicosecond pulses have been achieved in Ti:sapphire lasers by a variety of mode-locking techniques including acousto-optic modulation,¹ saturable absorption,² coupled cavity,³ synchronous pumping,⁴ and self-phase modulation.⁵ Cr³⁺-doped LiSrAlF₆ (Cr:LiSAF) is a promising alternative solid-state laser material for the near-IR spectral region, with the advantage over Ti:sapphire of being readily pumpable with either flash lamps or diode lasers. The emission bandwidth of Cr:LiSAF extends from 750 nm to beyond 1 μ m. In this Letter, we report our first ultrashort-pulse generation in Cr:LiSAF.

Cr:LiSAF, a uniaxial crystal with trigonal symmetry, was first shown to be a useful tunable laser source by Payne *et al.*^{6,7} A strong absorption band between 550 and 750 nm produced lasing by pumping with a cw krypton laser operating at 647 nm. Tuning was demonstrated from 780 to 980 nm with the free-running laser output occurring at 825 nm. An upper-state lifetime for this four-level, phonon-terminated laser was measured at 67 μ s (compared with 3 μ s for Ti:sapphire), and a maximum efficiency of 53% was deduced from the results.⁶ In these early experiments, the pump laser was chopped to reduce excess heating of the Cr:LiSAF crystal. More recently, improvements in crystal growth have produced laser-quality single-crystal boules up to 120 mm long and 25 mm in diameter.⁸ The ability to grow Cr:LiSAF with a wide range of doping allows optimization for different applications. Flash-lamp pumping of 100-mm-long, 4-at. %-doped Cr:LiSAF rods by Stalder *et al.*,⁹ who used standard Nd:YAG laser flash lamps, produced long-pulse and Q-switched operation tunable between 780 and 1010 nm. Availability of high-power diode lasers within the absorption band of Cr:LiSAF has allowed diode pumping under widely varying conditions. Scheps *et al.*¹⁰ used laser diodes emitting approximately 670 nm to produce up to 19.9-mW cw and 78.0-mW (peak) pulses with 2% Cr:LiSAF. Dixon *et al.*¹¹ and Zhang *et al.*¹² used 10% and 15%

Cr:LiSAF to demonstrate thresholds of less than 10 mW with 670-nm diodes, while Krupke *et al.*¹³ employed 38% Cr:LiSAF to demonstrate lasing with a 752-nm laser diode.

Early investigations of Cr:LiSAF were based on crystals grown by the horizontal zone-melting technique.^{6,7} Because the crystals adhere to the Pt boat and are intrinsically fragile, as-grown crystals are always badly cracked. Our investigations have shown that Cr:LiSAF melts nearly congruently at about 750°C and that single crystals can be grown by the Czochralski pulling technique. Great care is needed to control the thermal profile of the furnace as well as the cooling procedure in order to prevent cracking. We have optimized both the furnace design and the growth process to allow routine production of large, good-quality crystals.

An advantage of Cr:LiSAF over many other host materials is that there is almost no concentration quenching for the chromium doping. The doping concentration can thus be optimized for a specific pumping scheme. The nominal chromium concentration for flash-lamp pumping is 4 at. % in the melt (giving 2% in the crystal). In this Letter, we have used a lower chromium concentration for pumping at 647 nm, which is close to the peak of the absorption band. The Cr:LiSAF melt contained 2% chromium. Because of the decomposition of CrF₃ during both initial heating and subsequent growth, the actual doping concentration is ~0.8%. The crystal was grown along the [10 $\bar{1}$ 0] direction, and a rectangular block of size 6 mm \times 6 mm \times 20 mm was cut with *a* and *c* axes in the plane of the end faces. Both ends were then cut and polished at Brewster's angle for light polarized parallel to *c*, which gave a physical path length of 15 mm in the crystal.

The four-mirror, X-fold laser cavity arrangement, shown in Fig. 1, was based on a modified Schwartz Titan (Ti:sapphire) laser extended to incorporate an acousto-optic mode locker and two prisms. The Cr:LiSAF crystal was mounted on a water-cooled Invar block and pumped by a 3-W Laser Ionics krypton laser operating on both the 647- and 676-nm red lines (power ratio 4:1). A 10-cm focusing lens

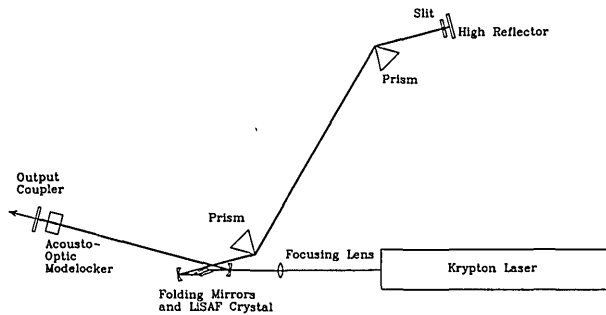


Fig. 1. Schematic of the mode-locked Cr:LiSrAlF₆ laser cavity configuration.

gave a spot size on the order of 25 μm . A Newport Electro-Optic Systems high-Q, 41-MHz acousto-optic (AO) modulator was placed close to the output coupler to modulate the cavity loss at 82 MHz (loss modulation 50–60%). Antireflection coatings on the AO modulator crystal were centered on 800 nm. Two different output couplers were employed, averaging 1% and 4% transmission over the 800- to 900-nm region. The prism pair comprised 60° prisms made from SF10 glass with the light incident close to Brewster's angle. Wavelength tuning was accomplished by a variable-aperture slit on a translation stage placed between the back mirror and the prism closest to it. The pulse durations were measured by a real-time autocorrelator, which could be adjusted to provide either zero-background or collinear interferometric autocorrelations.¹⁴ The pulse train was monitored using a fast silicon photodiode, and the spectral content of the pulses was determined by a 0.25-m monochromator with a silicon array-optical multichannel analyzer combination.

Using the 4% output coupler, we found that the threshold pump power level for cw lasing was 210 mW. Stable mode-locked operation of the laser was achieved at approximately 1 W input with an average output power of 200 mW when the cavity length was matched to the AO frequency. It was found unnecessary to chop the pump beam at this excitation level. Without prism compensation, pulse durations of 6 ps were obtained with an output spectral bandwidth of less than 0.5 nm.

Incorporation and careful adjustment of the prisms to compensate for group-velocity dispersion allowed a reduction of the output pulse width. A background-free autocorrelation trace recorded at the minimum pulse width is shown in Fig. 2(a) for an optimized prism separation of 48 cm. A sech^2 -fitted pulse shape with 1 ps FWHM implies a laser pulse duration of 650 fs. The spectral content of the pulses is plotted in Fig. 2(b). The 2.4-nm linewidth gives a time-bandwidth product $\Delta\tau_p\Delta\nu = 0.64$ (compared to the Fourier-transform limit of 0.32). An interferometric autocorrelation trace is shown in Fig. 2(c). At the minimum pulse width, a small degree of frequency chirp is evident by the loss of coherence in the wings. Attempts to achieve chirp-free pulses always resulted in longer pulse widths. In principle, the prisms should allow compensation for the chirp, indicating that there may be some interplay between group-velocity dispersion

and self-phase modulation. The output could be tuned from 800 to 880 nm by translating the aperture across the beam.

In light of recent advances in mode-locked Ti:sapphire lasers that achieve sub-100-fs pulses by using self-phase modulation in the laser crystal, we may consider Cr:LiSAF to be another good candidate for such a mode-locking scheme. This process, referred to as Kerr lens mode locking, is based on the bound-electron nonlinear refraction n_2 of the laser rod, which provides lensing that favors the higher mode-locked intensities through gain overlap and aperturing of the beam. We have demonstrated 100-fs-duration pulses from Ti:sapphire in the same cavity as that used for these Cr:LiSAF studies. Furthermore, the argon and krypton pump lasers employed in each case were matched Laser Ionics models 1400-15 and 1400-3K, respectively, and the same crystal length of 15 mm was chosen deliberately. The optimum prism separation was found to be similar for both the Ti:sapphire and the Cr:LiSAF lasers. (Preliminary Z-scan measurements indicate a value for n_2 approximately a factor of 4 smaller in Cr:LiSAF than that in Ti:sapphire.¹⁵) The principal difference in the operating conditions was a higher IR output power of approximately 500 mW (for an argon input power of ~ 6 W) in Ti:sapphire. Increasing the input power of the krypton pump to the Cr:LiSAF above 1 W caused a reduction in the output power of the laser. Re-adjusting the cavity configuration for higher powers did not remedy this, indicating that the problem is not thermal lensing but rather is due to the reduction in upper-state lifetime that occurs in Cr:LiSAF above room temperature.¹⁶ However, we have observed evidence of self-mode locking with the acousto-optic modulator removed from the cavity. By vibrating the rear mirror, broad output pulses were obtained at the cavity round-trip time riding on a cw background.

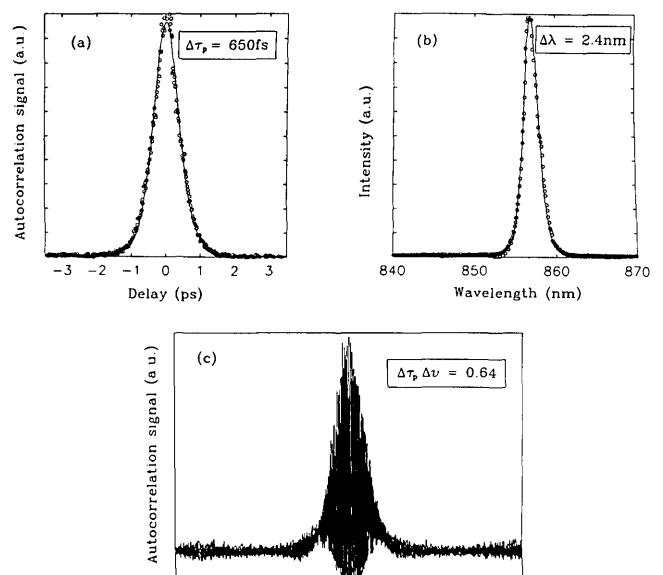


Fig. 2. (a) Intensity autocorrelation, (b) spectrum, and (c) interferometric autocorrelation of the output pulses of the prism-compensated laser using a 4% output coupler.

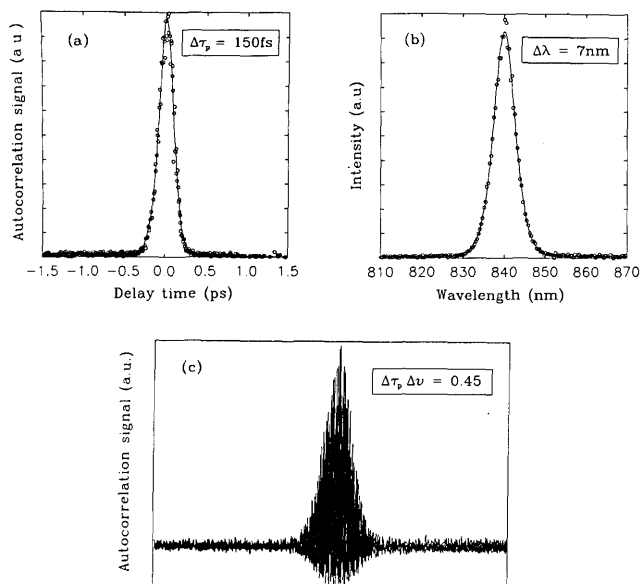


Fig. 3. (a) Intensity autocorrelation, (b) spectrum, and (c) interferometric autocorrelation of the output pulses of the prism-compensated laser using a 1% output coupler.

In order to increase the cavity intensity with the same pump power, a higher-reflectivity 1% transmission output coupler was employed. This gave a further reduction in pulse width when optimized. Figure 3(a) shows a 225-fs-width autocorrelation trace, which indicates 150-fs laser pulse durations assuming a sech^2 pulse shape. The corresponding spectral linewidth shown in Fig. 3(b) was 7 nm, giving $\Delta\tau_p\Delta\nu = 0.45$. Interferometric autocorrelations shown in Fig. 3(c) do not show evidence of chirp. The most likely cause of the pulse narrowing at higher cavity intensities is frequency spreading and pulse shortening by the Kerr lens mode-locking mechanism. Under these conditions, mode locking was less stable, indicating some thermal instabilities. Stable operation was only obtained by matching the cavity length to within a few micrometers, and mode locking ceased when the AO modulator was switched off. Amplitude stability was determined by pump laser noise at a few percent. The output retained a good spatial profile with approximately 50 mW average power when pumped with 1 W from the krypton laser. No self-Q-switching or driven relaxation oscillations were observed unless the cavity was far off cavity-length matching conditions.

In summary, a 15-mm-long, 2% Cr^{3+} -doped crystal of LiSrAlF_6 has been acousto-optically mode locked. Incorporation of an intracavity prism pair to compensate for group-velocity dispersion gave a continuous train of 150-fs pulses at 82 MHz. It would seem likely that with suitable cavity and crystal optimization and improved heat sinking, Cr:LiSAF will provide an ultrashort pulse source with operational characteristics similar to those of self-mode-locked Ti:sapphire lasers. The low threshold and lack of concentration quenching for the chromium doping

make Cr:LiSAF a versatile solid-state laser material with good prospects for use as a miniature, diode-pumped ultrafast source and for large-scale femtosecond amplifiers in the near IR.

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