High-energy, high-efficiency harmonic generation from a Cr:LiSrAlF$_6$ laser system

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We have developed a near-diffraction-limited Q-switched Cr:LiSrAlF$_6$ (Cr:LiSAF) laser system capable of producing 860-nm, 400-mJ pulses at a repetition rate of 2 Hz. Our harmonic-generation experiments with β-BaB$_2$O$_4$ nonlinear crystals have yielded 200 mJ of energy at 430 nm and 40 mJ at 215 nm. To the best of our knowledge, the 50% second-harmonic conversion efficiency and UV energy demonstrated in this research are the highest yet obtained from a Cr:LiSAF laser.

Cr:LiSrAlF$_6$ (Cr:LiSAF) is a relatively new tunable laser operating on the near-infrared vibronic transition of the Cr^{3+} ion.[1] Compared to the Ti:sapphire laser system, Cr:LiSAF does not have as wide a tuning range, but the much longer upper-state lifetime (67-μs) and two broad, visible-wavelength absorption bands permit efficient pumping with standard flash lamps. Lamp-pumped Cr:LiSAF rods have been used to amplify the output of mode-locked Ti:sapphire oscillators to the multiterawatt level.[2,3] Lamp-pumped rod oscillators operating in the long-pulse or Q-switched mode have been demonstrated by a number of groups. Stalder *et al.* reported a flash-lamp-pumped long-pulse Cr:LiSAF laser with a slope efficiency of 5% and an overall efficiency of 3.1% at 845 nm.[4] The same group obtained Q-switched pulse generation with a rotating mirror and observed 150-mJ pulses with a duration of 40–50 ns. Shimada *et al.* have demonstrated an electro-optically Q-switched multimode flash-lamp-pumped Cr:LiSAF oscillator that emitted 455 mJ of energy in a 32-nsec pulse.[5] Harter *et al.* reported the development of a 200-mJ near-diffraction-limited Q-switched Cr:LiSAF laser system pumped by a long-pulse alexandrite laser.[6] Most of the flash-lamp-pumped Q-switched Cr:LiSAF lasers reported to date have been multitransverse-mode systems; the goal of the research reported here was to develop a high-energy flash-lamp-pumped Q-switched near-diffraction-limited laser system suitable for efficient driving of nonlinear processes.

By harmonic generation of the tunable, near-IR Cr:LiSAF laser output, one can reach significant portions of the UV/blue spectral regions. The nonlinear crystal β-BaB$_2$O$_4$ (BBO) can generate the second harmonic of various pulsed, near-IR lasers, with single-crystal efficiencies of up to 60% reported for Ti:sapphire and 31% for alexandrite.[7,8] Unfortunately, BBO has narrow angular acceptance and requires the use of a near-diffraction-limited pump source for efficient conversion. In this Letter we report the development of a 400-mJ Q-switched TEM$_{00}$ oscillator–amplifier Cr:LiSAF laser system and on its performance as a drive source for second- and fourth-harmonic generation in BBO.

Many different approaches have been employed in the construction of high-energy near-diffraction-limited solid-state laser systems. The oscillator design currently used in many commercial pulsed Nd:YAG lasers is the graded-reflectivity-mirror unstable resonator. With relatively low-gain materials such as Cr:LiSAF the central reflectivity of the graded mirror must be high and the resonator magnification low to yield a reasonable lasing threshold from an unstable oscillator. Increasing the reflectivity can result in optical damage, whereas reducing the magnification will increase the sensitivity of the cavity to misalignment. Because of these considerations we chose to obtain a large, TEM$_{00}$ mode volume in the Cr:LiSAF oscillator by building a conventional stable resonator with an intracavity telescope.

The telescopic resonator, which was first described by Sarkies, can be used to reduce the resonator length required for production of a given mode volume by a factor of M^2, where M is the telescope magnification.[9] In our system layout, which is shown in Fig. 1, we included a 2.5× telescope in the oscillator to produce a mode diameter ($1/e^2$) at the output coupler of 2.6 mm. To avoid optical damage we placed

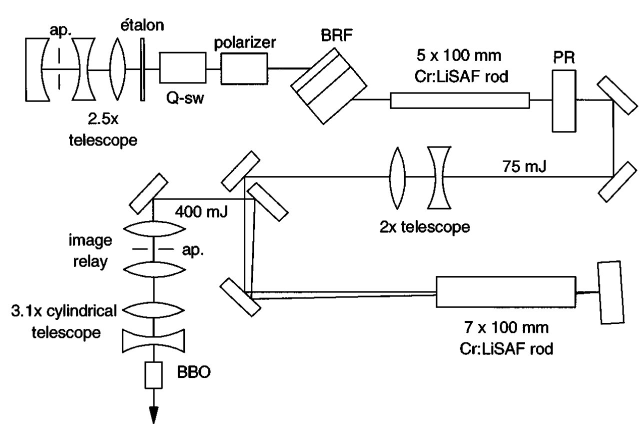

Fig. 1. Frequency-doubled Cr:LiSAF laser system. The design consisted of a telescopic oscillator followed by a double-pass amplifier and a BBO second-harmonic generator. All mirrors are highly reflecting from 800 to 900 nm, unless otherwise noted in text. Q-sw, Q switch; ap.'s, apertures.

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all the resonator components except for the 3-m radius-of-curvature high reflector and the 1.4-mm-diameter aperture to the right of the telescope. The 1.5%-doped antireflection-coated 5 mm × 100 mm Cr:LiSAF laser rod, oriented for π polarization, was mounted in a diffuse-reflecting water-cooled pump cavity along with a 450-Torr Xe flash lamp, which was driven by a 75-μs (FWHM) pulse produced by a transistor-switched power supply. The distance between resonator mirrors was 128 cm, and the flat output mirror (PR) had 36% transmission at 860 nm. We Q-switched the oscillator with a fluid-filled antireflection-coated KD*P Pockels cell driven by a thyratron-switched high-voltage power supply. A three-plate, crystal-quartz birefringent filter (BRF, 0.5:1:7.5 mm) allowed us to tune the oscillator output and narrow the linewidth. In all the results discussed below we tuned the laser to a 860-nm wavelength. We inserted a Brewster-angle calcite polarizer into the oscillator cavity to increase the selectivity of the birefringent tuner. We also added an intracavity, 200-μm-thick fused-silica étalon with a 30%-reflecting coating on each face to narrow the linewidth further. At 31 J of lamp input and a 2-Hz pulse rate the oscillator produced 75 mJ of near-diffraction-limited ($M^2 = 1.5$) output in a 126-ns ($1/e^2$) pulse. At this operating point we measured the bandwidth with a Burleigh pulsed wavemeter to be 0.13 nm (FWHM). Intrinsic phase distortions and the presence of scattering sites in the rod may have been factors in reducing the beam quality.

We increased the diameter of the output beam to 5.2 mm with a 2× telescope and folded the beam into an amplifier stage. The 7 mm × 100 mm 0.8%-doped antireflection-coated amplifier rod was contained in a water-cooled, diffuse-reflecting cavity and was pumped by two 450-Torr Xe flash lamps, which were driven by 75-μs-duration (FWHM) current pulses. To increase the extraction efficiency from the amplifier we used a flat high reflector to double-pass the rod; the mirror was tilted at a small angle to separate the input and output beams. With each flash lamp pumped at 45 J the amplifier output energy was 400 mJ for 75 mJ of input. We employed two 60-cm focal-length lenses spaced by 120 cm to image relay the profile present at the end of the amplifier rod into the second-harmonic crystal. We placed a 1.1-mm-diameter aperture at the focus of the first lens to provide spatial filtering of the beam. Over time, both the oscillator and the amplifier rods developed internal damage sites, probably originating at inclusions or impurities present in the laser material. These sites appeared to stabilize and did not lead to internal fracture in the rods, but they did affect the near-field spatial beam profile. After the sites developed, we measured the profile of the amplifier output after the spatial filter (Fig. 2) with a Big Sky Laser Technologies beam analyzer. The horizontal and vertical slices plotted were taken through the peak of the profile. We measured the beam quality of the system operating at the 400 mJ level to be $M^2 = 1.5$.

The characteristics of BBO for frequency doubling at 860 nm are shown in Table 1.[10] The threshold power is the most important parameter in determining the second-harmonic-generation (SHG) efficiency achievable at a fixed laser brightness, as it incorporates both the angular sensitivity and the nonlinear coefficient in a single figure of merit.[11] Because the 3.2-MW peak power of our laser was low relative to the 16.7-MW BBO threshold power, we assembled a 3.1× cylindrical telescope to reduce the beam size in the transverse direction with the largest angular acceptance. Using a Spiricon LBA-100 beam profiler, we measured the spot diameter ($1/e^2$ points) incident upon the BBO crystal to be 0.67 cm × 0.19 cm. At this point the beam divergence was 0.254 mrad × 0.895 mrad. The second-harmonic crystal was 8 mm × 8 mm × 10 mm, broadband antireflection coated, and cut for Type I phase matching at a nominal angle of 27.5° to the c axis. The squares in Fig. 3 show the conversion efficiency as a function of the input energy; the last data point represents 200 mJ of blue energy at 50% efficiency. The dashed curve is a theoretical prediction based on the model described in Ref. 12. The model was derived from Ref. 11 and can be applied to SHG experiments in which the conversion efficiency is limited primarily by angular dephasing. The model assumes a spatially and temporally uniform beam, but, as suggested in Ref. 12, we have found good agreement between theory and experiment by measuring both the beam width and the temporal profile at the $1/e^2$ points. BBO threshold power (Table 1), beam quality (M^2), spot size, crystal drive, and fundamental input power were used as input parameters to the model. The agreement is good, except at the last point, where the

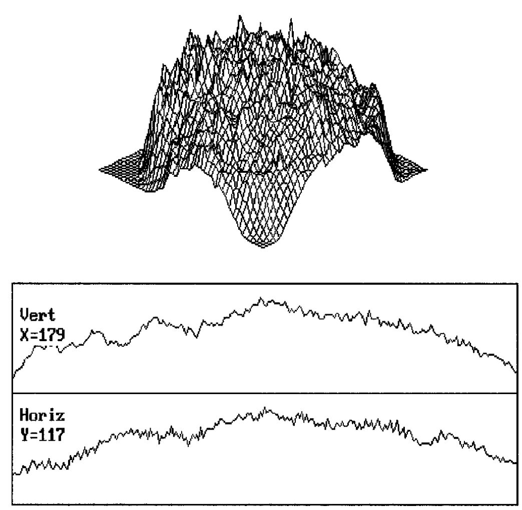

Fig. 2. Spatial profile of the 400-mJ, 860-nm beam taken after the spatial filter at the image plane of the amplifier rod. The vertical and horizontal profiles were taken through the peak.

Table 1. Parameters for BBO Frequency Doubling at 860 nm (Bandwidths HWHM)

Phase-matching angle at 860 nm (deg)	27.2
Angular bandwidth (cm mrad)	0.30
Temperature bandwidth (cm °C)	8.50
Spectral bandwidth (cm nm)	0.34
Threshold power (MW)	16.7

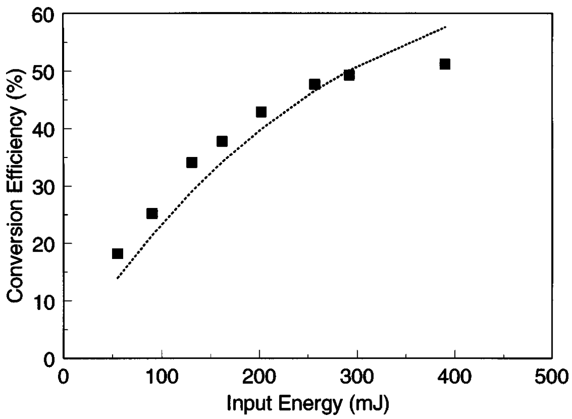

Fig. 3. Conversion efficiency as a function of input energy for SHG at 860 nm in a 10-mm-long BBO crystal. The data are represented by the solid squares; the dotted curve is a theoretical model.

theoretical curve appears to diverge from the experimental points. This effect was most likely caused by either (1) spectral dephasing that is due to the relatively broad linewidth (0.13 nm) of the Cr:LiSAF laser or (2) the Gaussian profile of the beam.

We are currently in the process of exploring efficient generation of higher-order harmonics from the Cr:LiSAF system and thus far have produced 40 mJ of energy at 215 nm by SHG of the doubled Cr:LiSAF output. In principle, BBO can be phase matched to produce wavelengths as short as 205 nm by SHG, but the effective nonlinearity falls to zero at the short-wavelength limit.

We have demonstrated a flash-lamp-pumped Cr:LiSAF laser system that is capable of producing high-energy Q-switched near-diffraction-limited pulses in the IR. Although the pulse width produced by our system was long, we have achieved 50% second-harmonic conversion efficiency in BBO through the use of cylindrical focusing. Because the fracture strength of Cr:LiSAF rods has been known to vary from one crystal to the next, possibly related to the level of defects, we have chosen to operate the system at a low pulse rate rather than to risk mechanical damage to one or both laser rods. Recent improvements in the growth of Cr:LiSAF have reduced the passive loss to <0.2%/cm,[13] and material of this quality may be able to operate, in systems similar to ours, free of internal damage, with improved beam quality and at higher pulse rates. Applications such as remote sensing of chemical pollutants require tunable UV/blue laser sources, and Cr:LiSAF may prove to be a practical alternative to current laser systems.

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References

1. S. A. Payne, L. L. Chase, L. K. Smith, W. L. Kway, and H. W. Newkirk, J. Appl. Phys. **66,** 1051 (1989).
2. P. Beaud, M. Richardson, E. J. Miesak, and B. H. T. Chai, Opt. Lett. **18,** 1550 (1993).
3. T. Ditmire, H. Nguyen, and M. D. Perry, J. Opt. Soc. Am. B **11,** 580 (1994).
4. M. Stalder, B. H. T. Chai, and M. Bass, Appl. Phys. Lett. **58,** 216 (1991).
5. T. Shimada, J. W. Early, C. S. Lester, and N. J. Cockroft, in *Advanced Solid-State Lasers,* Vol. 20 of OSA Proceedings Series (Optical Society of America, Washington, D.C., 1994), pp. 188–191.
6. D. J. Harter, J. Squier, and G. Mourou, Opt. Lett. **17,** 1512 (1992).
7. G. A. Rines, H. H. Zenzie, and P. F. Moulton, in *Digest of Topical Meeting on Advanced Solid-State Lasers* (Optical Society of America, Washington, D.C., 1991), pp. 64–65.
8. D. W. Chen and J. J. Yeh, Opt. Lett. **13,** 808 (1988).
9. P. H. Sarkies, Opt. Commun. **31,** 189 (1979).
10. G. Catella and D. Roberts, Cleveland Crystals, 19306 Redwood Avenue, Cleveland, Ohio 44110-2738 (personal communication).
11. D. Eimerl, IEEE J. Quantum Electron. **QE-23,** 575 (1987).
12. R. G. Beausoleil, Lasers Optron. **11,** 17 (1992).
13. G. Quarles, Lightning Optical, 431 E. Spruce Street, Tarpon Springs, Fla. 34689 (personal communication).