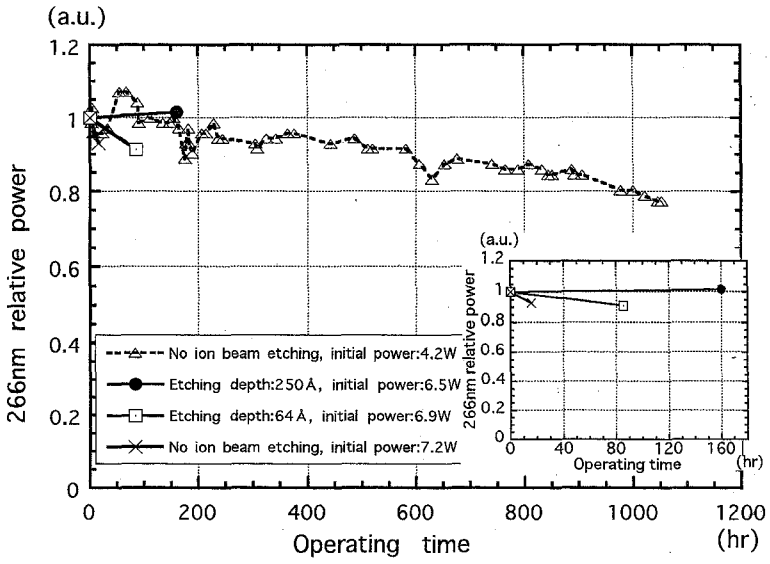
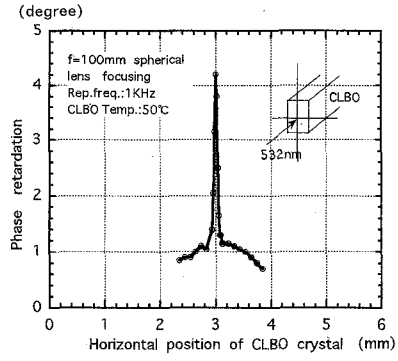
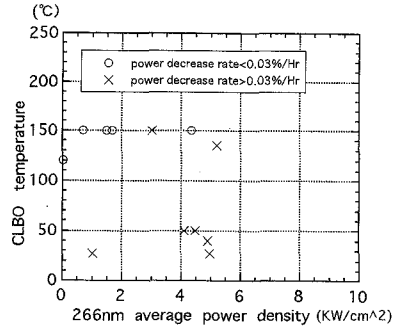


CFG3 Fig. 1. 266-nm average output power vs. operating time. CLBO temperature was elevated to 120°C, and high-purity nitrogen gas was flowed around the crystal at the rate of 0.7 L/min to prevent CLBO surface from moisture.



CFG3 Fig. 2. Phase retardation at 633 nm in the CLBO vs. horizontal position.



CFG3 Fig. 3. Relation of 266-nm power density and temperature at CLBO with respect to above and below 0.03%/h of decrease rate.

output power decreased drastically to <50% of the initial power during 15 h of irradiation at room temperature. Although no surface damage was observed in this case, the output beam profile was distorted as the output power decreased. These results lead to speculation that the refractive-index change occurred in the focused area of the CLBO. As Fig. 2 shows, we observed the abrupt refractive-index change using an optical heterodyne method with a frequency-stabilized transverse Zeeman laser as a probe beam. Because the optical heterodyne signal is proportional to the difference of the refractive index between the ordinary and extraordinary waves at the phase-matching direction, the refractive-index change can be the phase-retardation change in the focused region. Therefore, this is considered to be direct evidence of refractive-index change. When the angle of incidence was detuned from the phase-matching angle to eliminate the fourth-harmonic generation, such retardation change was not observed even though the same amount of focused beam at 532 nm was input to the crystal more than 15 h. Therefore, we can conclude that this refractive-index change was caused by self-heating of CLBO by absorbing 266-nm light.

Figure 3 shows the methodology of CLBO for fourth-harmonic generation. Although reported earlier about the necessity of using CLBO at an elevated temperature (>130°C), it is also important to use CLBO at reduced 266-nm average power density below 2 KW/cm^2 in order to avoid drastic decrease of output power. Below this density, no refractive-index change was observed.

Department of Electrical Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565, Japan

**Department of Electronics, Osaka Institute of Technology, 5-16-1 Ohmiya, Asahiku, Osaka 535, Japan*

CFG4 11:15 am

Second-harmonic generation in doped YCOB

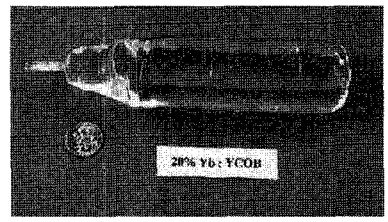
Won K. Jang, Qing Ye, Jason Eichenholz, Bruce H.T. Chai, Martin Richardson, *CREOL, University of Central Florida, 4000 Central Florida Blvd., Orlando, Florida 32816; E-mail: jwk1@lorien.creol.ucf.edu*

Frequency upconversion in nonlinear crystals is the most common approach to the generation of visible and UV laser light with solid-state lasers, most of which operate in the near infrared. Although KDP is most widely used for second-harmonic generation (SHG), in the last few years, several new nonlinear optical crystals such as β barium borate (BBO) and LBO have been developed that have high nonlinear coefficients, high damage thresholds, and a broad transparency range. However, all these crystals are soluble in water, and the flux-grown borcate crystals are limited to small sizes. We report encouraging results with a new class of nonlinear optical crystals, doped versions of rare-earth calcium oxoborate crystals,[1] that have a high d_{eff}, a high damage threshold, are nonhygroscopic, and can be grown fast in large sizes with good mechanical properties, which allow easy optical polishing.

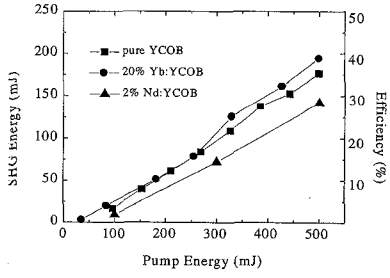
Recently, SHG has been measured in two oxoborate crystals YCOB and GdCOB.[2,3] We are extending the development of the oxoborate crystal for nonlinear frequency conversion by investigating the effects of dopant ions on the crystalline structure and, as a consequence, improving the optical nonlinearity. In addition, the use of specific dopants opens a path for these crystals to be used in self-frequency-doubled lasers.

We have measured nonlinear harmonic generation in a wide range of oxoborate crystals that include YCOB [$YCa_4(BO_3)_3O$], Nd:YCOB, and Yb:YCOB. These crystals were fabricated by the Czochralski growth technique. Large boules of length ~150 mm and diameter ~30 mm were grown with good optical quality, which is shown in Fig. 1.

SHG has so far been investigated in four of these crystals, pure YCOB, 2% Nd-doped YCOB, and 20% and 44% doped Yb:YCOB. The output of a high-repetition-rate, 100-Hz, Q-switched Nd:YAG laser collimated in a narrow beam was used to generate SHG in uncoated, 25-mm-long crystals having a clear aperture of 4 mm × 4 mm. In Fig. 2, the measured SHG energy output as a function of



CFG4 Fig. 1. Photograph of 20% Yb:YCOB boule.



CFG4 Fig. 2. Second-harmonic energy in new nonlinear crystals vs. pump energy at the fundamental of Nd:YAG laser.

fundamental laser pulse energy is plotted for these crystals and shows some changes in conversion efficiency between pure YCOB and doped YCOB. Conversion efficiencies approaching 40% were measured with 20% Yb-doped YCOB for a pump power of ~11 MW.

Investigations are under way to get a better understanding of the effects of dopant ions in the crystal structure and on optimizing these materials for harmonic generation and self-frequency-doubled lasers.

1. R. Norrestam, M. Nygren, J.O. Bovin, Chem. Mater. **4**, 737 (1992).
2. Makoto Iwai, Taisuke Kobayashi, Hiroyuki Furuya, Yusuke Mori, Tokatomo Sasaki, Jpn. J. Appl. Phys. **36**, L276 (1997).
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CFG5 11:30 am

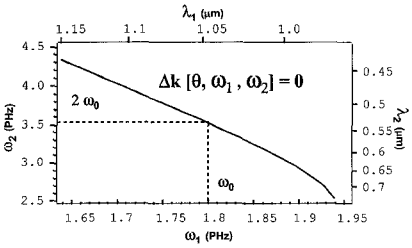
Efficient frequency tripling of 1.06-μm, 300-fs chirped pulses

F. Raoult, A.C.L. Boscheron, D. Husson, C. Rouyer, C. Sauteret, A. Migus,*
Commissariat à l'énergie atomique, Centre de Limeil-Valenton, 94195 Villeneuve St. Georges Cedex, France; E-mail: boschero@limeil.cea.fr

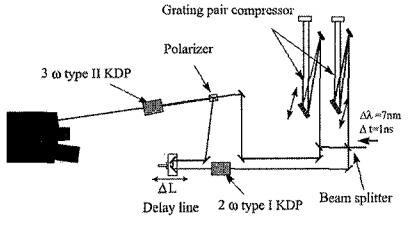
Picosecond and femtosecond energetic pulses are difficult to produce in the UV domain. One approach consists in excimer lasers but with limited performances, the other one in frequency doubling or tripling solid-state laser outputs. The difficulty in the latter case lies in the phase-matching condition, which limits the emitted bandwidth and therefore the pulse shortness with long crystals or has a low efficiency in case of thin crystals.

We report here an experimental verification of a previously published concept,[1] which allows us to efficiently frequency-triple broadband pulses using a chirped-pulse amplification (CPA) configuration. The idea is based on the manipulation of pulses with appropriate chirps for maintaining phase matching over the whole bandwidth. It either starts from two differently chirped pulses in a CPA system or from a beam-splitted stretched output of a CPA amplifier sent onto two different grating pair stages. Although not optimal, this is the configuration used here. As a consequence, two different chirps can be created, described as frequency laws versus time: $\omega_1(t) = \omega_0 + b_1 t$ and $\omega_2(t) = \omega_0 + b_2 t$. In our case of tripling, one of these pulses is frequency doubled so that the pulse chirp becomes $\omega'_2(t) = 2\omega_0 + b'_2 t$. In order to maintain phase matching for third-harmonic generation, and therefore get a large bandwidth with an efficiency as large as with the monochromatic case, the sum $\omega_1(t) + \omega'_2(t)$ has to follow the phase-matching law plotted on Fig. 1. For KDP and 1.06-μm type II tripling, this curve is linear in a large spectral range corresponding to a slope of $b'_2/b_1 = -5.17$.

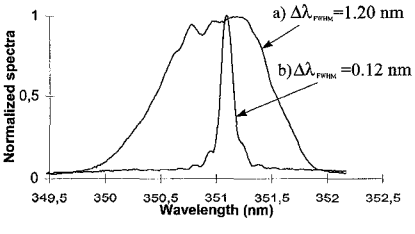
The experiments were performed using a Ti:sapphire femtosecond laser and a CPA amplifier configuration based on a Ti:sapphire regenerative amplifier and Nd:phosphate glass head delivering 1-ns chirped pulses with 100-mJ energy, which can be compressed to duration of the order of 300 fs. We use a KDP type I–type II collinear configuration as shown in Fig. 2. In order to estimate the performance improvements, we use as a reference a configuration with identical chirped pulses at 1.06 μm (55 ps duration FWHM). We verified that the third-harmonic spectral shape (FWHM: 0.12 nm) corresponds to the expected sinc^2 wavelength tuning curve of a 3-cm type II 3ω-KDP crystal [Fig. 3(a)]. In this case, the conversion efficiency is 4%. In a second experiment, one of the compressor grating is translated in order to introduce the chirp needed to get the slope of −5.17. This corresponded to mixing a 133-ps to 1.06-μm pulse and a 38-ps to 0.53-μm pulse obtained by frequency doubling the initial 55-ps to 1.06-μm pulse in a 2-cm type I KDP. As shown on Fig. 3(b), we obtain a third-harmonic spectral bandwidth of 1.2 nm, 10 times the reference bandwidth, limited only by the initial pulse spectrum. The incident energies on the 3ω crystal at 1.06 μm and 0.53 μm were 21 mJ and 7.5 mJ, the output at 351 nm 5.3 mJ, respectively. Scaling to the 2ω pulse duration, the overall efficiency is 40%. Experiments show that the bandwidth produced allows 351-nm pulses of 300-fs duration using a grating pair compressor, this technique being scalable to the petawatt level.



CFG5 Fig. 1. Phase-matching curve for mixing ω_1 and ω_2 in a tripling type II KDP ($\theta = 59°$). This curve is linear in a large spectral range for the frequency ω_0 we consider.



CFG5 Fig. 2. Experimental setup of broadband third-harmonic generation. Different chirps can be created by using two compressors.



CFG5 Fig. 3. Third-harmonic relative phase-matching efficiency vs. wavelength using a 2-cm type I 2ω KDP and a 3-cm type II 3ω KDP. Curve (a) shows the third-harmonic spectrum obtained with a classical configuration. Curve (b) shows the third-harmonic spectrum obtained with appropriate chirped pulses for maintaining phase matching over the whole bandwidth.

Laboratoire pour l'Utilisation des Lasers Intenses, CNRS UMR 100, Ecole Polytechnique, 91128 Palaiseau Cedex, France

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CFG6 11:45 am

Tunable visible solid-state lasers based on second-harmonic generation of LiF: F_2^- in KTP

Shirin M. Giffin, Iain T. McKinnie, Valerii V. Ter-Mikirtychev,* *Department of Physics, University of Otago, PO Box 56, Dunedin, New Zealand;*
E-mail: shirin@physics.otago.ac.nz

Tunable green-yellow lasers are required for a wide range of applications including dermatology, turbid surf-zone imaging, and isotopic enrichment. However, the current solid-state sources in this region, optical parametric oscillators (OPOs) and solid-state dye lasers, have limitations. Operating lifetimes of solid-state dye lasers remain extremely short, and OPOs place strict requirements on the spatial and spectral properties and pointing stability of the pump laser. Here we investigate a new solid-state route to a tunable visible laser, which minimizes these problems. Efficient operation with broadband tunability through the green-orange region is demonstrated using intracavity second-harmonic generation of room-temperature LiF:F_2^- lasers in KTP. The unique 1070-nn to 1310-nm operating wavelength range of LiF:F_2^-[1] makes it the only solid-state laser with the potential for tunable second-harmonic generation across the entire 550-nm to 650-nm region. However, to our knowledge, second-harmonic generation of LiF:F_2^- lasers has so far received little attention,[2,3] and characterization of efficient intracavity frequency doubling has not been carried out.

LiF:F_2^- lasers are limited by thermo-optical effects, passive loss, and bleaching. These problems are alleviated with lower repetition rates and shorter pulses, and recent improvements in LiF:F_2^- crystals have led to stabilization of the F_2^- centers[4] and significant improvements in laser performance.[5] In our experiments, we have observed no degradation in laser performance during 7 mo of pulsed room-temperature operation.

Performance of the frequency-doubled LiF:F_2^- laser has been characterized in linear and Brewster-prism cavities, pumped at 1064 nm by a seeded Q-switched Nd:YAG laser. The KTP crystal was cut at $\phi = 0°$, $\theta = 65°$ for efficient type II critical phase matching at 1235 nm and coated for dual-band