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An all-solid-state laser system for generation of 100 μJ femtosecond pulses near 625 nm at 1 kHz

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Abstract. Frequency doubling the output of a high-power femtosecond Cr:forsterite regenerative amplifier with > 50% conversion efficiency in a temperature-tuned noncritically phase-matched LBO crystal produces femtosecond pulses of > 100 μJ energy in the visible range near 625 nm at a pulse duration of about 200 fs or > 65 μJ at < 170 fs.

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Recently there has been considerable interest in the realization of all-solid-state short-pulse laser systems operating in the visible near 600 nm to fill in the gap between the fundamental and the second harmonic of Ti:sapphire laser systems and as an alternative to the previously developed multipass dye amplifiers. Several concepts to produce high energy (> 1 μJ) femtosecond pulses in this spectral range exist, starting with a Ti:sapphire regenerative amplifier. A visible optical parametric generator (OPG) can be pumped by frequency-doubled femtosecond pulses near 400 nm [1] and the use of noncollinear group-velocity matching in a multistage OPG resulted in pulse energies as high as 8 μJ [2]. A near-infrared kilohertz OPG pumped at the fundamental [3] can, however, produce much higher energies. By frequency doubling the signal output or mixing the idler with the pump, energies up to 90 μJ were demonstrated near 600 nm [4]. Although the combined down- and up-conversion in a single crystal has also been demonstrated [5] the two frequency conversion steps and especially the optical parametric amplification considerably complicate all these schemes and make them sensitive to pump parameter fluctuations.

Regenerative amplifiers based on Cr:forsterite can provide compressed pulse energies on the order of 100 μJ [6] to 200 μJ [7] near 1.25 μm at 1 kHz and seem very promising for covering the 600-nm spectral range by a single frequency conversion step – second harmonic generation (SHG). We note that previously, instead of amplifying in Cr:forsterite, the output of a cavity-dumped Cr:forsterite oscillator was first frequency-doubled and then amplified in a dye amplifier, pumped at 523 nm, to 0.4 μJ at 4 kHz [8]. First amplifying in Cr:forsterite and then performing the SHG is, however, not only an all-solid state system, but much simpler. It also has higher conversion potential since the fundamental of the Nd:YAG laser can be used as the pump source for the amplifier. Unfortunately, the low overall extraction and conversion efficiencies recently found with this method [9] resulted in pulse energies of only 7 μJ near 625 nm at 1 kHz. In this article we demonstrate a significant increase in both the conversion efficiency (to > 50%) and the output energy achieved by using temperature tuning in a noncritically phase-matched LBO crystal pumped by 200 μJ from our regenerative amplifier [7]. Specifically we produced > 100 μJ, 200-fs pulses or > 65 μJ, < 170-fs pulses in the visible at a 1-kHz repetition rate.

The Cr:forsterite femtosecond oscillator and regenerative amplifier used in the present work are described in detail elsewhere [7, 10]. The shortest compressed pulses from the regenerative amplifier had a duration of about 155 fs and a spectral width corresponding to 1.5 times the Fourier limit, assuming Gaussian pulse shapes. The maximum fundamental pulse energy was 200 μJ at a 1-kHz repetition rate.

Frequency doubling of femtosecond Cr:forsterite lasers in LBO was demonstrated previously by using both critical [11] and noncritical [10] phase-matching. For SHG with amplified pulses we employed noncritical temperature-tuned phase-matching in LBO (propagation along the *x* axis, type-I oo-e interaction). Although as compared to other nonlinear crystals applicable in this spectral range LBO has a relatively small nonlinearity, the lack of spatial walk-off and the large angular acceptance provide an almost circular beam profile at the second harmonic, which is a unique feature. The internal angular acceptance (FWHM) of LBO calculated using the Sellmeier data from [12] is $\Delta\varphi l^{1/2} = 5^\circ\,\mathrm{cm}^{1/2}$ (l, crystal length). In fact the low nonlinearity of LBO can be compensated by using longer crystals, because the dispersion of LBO is also lower, and/or by choosing appropriate focusing conditions. As well as the low dispersion, which means a small group delay or temporal walk-off between the fundamental and the second harmonic, another important advantage of LBO, especially when dealing with high-power femtosecond pulses, is the relatively small value, $1.7 \times 10^{16}\,\mathrm{cm}^2/\mathrm{W}$, for the nonlinear refractive index (measured by self-phase-

modulation spectral broadening at 616 nm [13]). Although absolute measurements at longer wavelengths have not been published yet, from a comparison of the available data [13] it can be expected for our SHG case that the Kerr-type nonlinearity for LBO is 0.47 of that for KDP and 0.32 of that for BBO.

The 5-mm-long LBO crystal used in the present experiment was antireflection-coated at 800 nm and was provided by CASIX. It was mounted in a metal holder and attached to a copper block equipped with a thermoelectric cooler. The fundamental beam was focussed by a 20-cm best-shape lens and the nonlinear crystal was positioned before the focal point. At a fundamental wavelength of 1.252 μm we measured a phase-matching temperature of 3.5 °C (lower than the calculated value of 11 °C) and a FWHM for the temperature bandwidth of 9 °C (smaller than the calculated value of 13.5 °C).

At a peak on-axis pump intensity of about 10 GW/cm^2 we obtained conversion efficiencies as high as 33%. Deconvolution of the corresponding autocorrelation trace in Fig. 1 yields a pulse duration of 169 fs at the second harmonic. This represents only a slight broadening, as compared to the fundamental pulse, which can be attributed to the group velocity mismatch (about 9 fs/mm). The corresponding spectral width (also only slightly increased as compared to the fundamental) leads to a pulselength–bandwidth product about 1.8 times the Fourier limit. A conversion efficiency as high as 52% could be achieved at peak on-axis intensities of 50 GW/cm^2 at the fundamental. In this case the pulse width broadened to 200 fs but the pulse was still single peaked with a pulselength–bandwidth product 2.2 times the Fourier limit. As can be seen from Figs. 1, 2 in both cases the autocorrelation curves and the spectra can be well fitted by assuming Gaussian pulse shapes. Spectral broadening as a result of the fivefold increase of the fundamental intensity could not be observed, and comparing our results to observations in LBO based OPGs [13] we calculated an on-axial B integral at the fundamental of $< 0.07\pi$ and conclude that higher-order (Kerr-type) nonlinear effects play a negligible role in our experiment. Consequently, it is possible to achieve comparable and even higher conversion efficiencies for SHG than with mode-locked Cr:forsterite lasers [10, 11] without distortions originating from higher-order nonlinearities. We are not aware of SHG studies in LBO at such high intensities, but it is possible to compare the result obtained here with the frequency doubling in KDP theoretically investigated in [14, 15] for amplified femtosecond pulses near 800 nm. Taking into account the different wavelength and the lower Kerr-type nonlinearity of LBO we estimated from the results in [14, 15] that the influence of the cubic nonlinearity for our experimental conditions should not appear up to on-axis intensities as high as 80 GW/cm^2 in accordance with our experimental result. We attribute, therefore, the additional temporal broadening and increase of the Fourier product at 50 GW/cm^2 to temporal and spectral reshaping as a result of the high-energy exchange between the fundamental and the second harmonic (saturation of the SHG). Note that the quoted conversion efficiencies are with respect to the energy and much higher conversion leading also to reconversion can be expected on the axis (according to [16] of the order of 87% for Gaussian pulses). At such high peak conversion the phase-matching bandwidth would be modified (according to Fig. 2 in [16] by about 30%). The resulting spectral shaping is, however, too difficult to distinguish in the present experiment to be numerically simulated since all spectral measurements are averaged over the spatial beam profile. The Fourier product, though increased, is still < 1 at 50 GW/cm^2. The main factor contributing to this value remains the fact that the fundamental pulses are not Fourier limited, whereas, as just discussed above, the contributions of temporal and spectral reshaping due to group mismatch (seen at lower power) and the high conversion are almost equal.

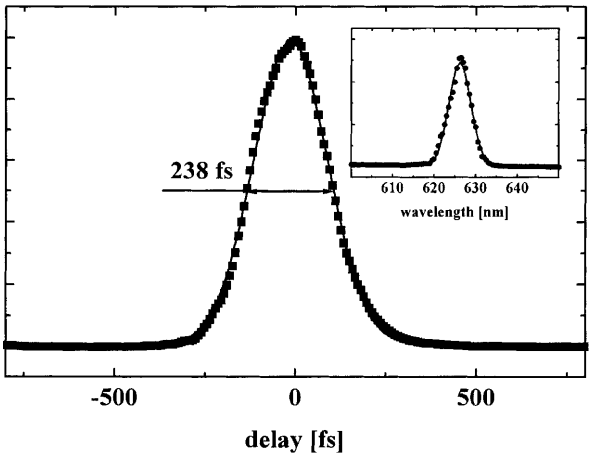

Fig. 1. Autocorrelation trace and corresponding spectrum (*inset*) at the second harmonic for an on-axis peak pump intensity of 10 GW/cm^2. The deconvolved pulse duration is 169 fs and the pulselength–bandwidth product amounts to 0.8. The *lines* are Gaussian fits to the experimental points

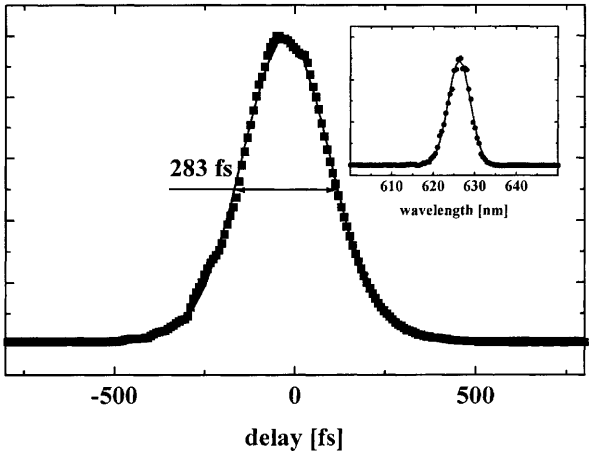

Fig. 2. Autocorrelation trace and corresponding spectrum (*inset*) at the second harmonic for an on-axis peak pump intensity of 50 GW/cm^2. The deconvolved pulse duration is 200 fs and the pulselength–bandwidth product amounts to 0.97. The *lines* are Gaussian fits to the experimental points

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