High-repetition-rate femtosecond pulse generation in the blue

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Received November 20, 1991

We report the generation of high-repetition-rate femtosecond pulses in the blue by intracavity doubling of a mode-locked Ti:sapphire laser using β-BaB_2O_4. To reduce the pulse-broadening effect of group-velocity mismatch, an extremely thin β-BaB_2O_4 crystal is used. By pumping the Ti:sapphire laser with 4.4 W of power from an Ar^+ laser, as much as 230 mW of 430-nm light is produced at a 72-MHz repetition rate and a 89-fs pulse width. This represents an effective conversion efficiency of ~75% from the typical infrared output to the second harmonic. Pulse widths as short as 54 fs are achieved for the blue output.

Extension of the wavelength range accessible to femtosecond pulses has been a topic of much interest. The two techniques used most frequently to generate <100-fs pulses at otherwise unattainable wavelengths are continuum generation and frequency conversion with the use of crystals. Femtosecond pulse generation techniques based on amplification followed by continuum generation permit tunability from the UV into the IR.[1] However, amplification reduces the pulse repetition rate to the order of a kilohertz, and there is often a loss of time resolution in the final pulse. In contrast, frequency conversion in crystals can maintain the high repetition rate of the femtosecond megahertz-rate laser and requires only a single cw pump laser. The higher repetition rate results in much smaller pulse fluctuation and excellent experimental signal-to-noise ratios.

In recent years, much progress has been made in extending the spectral range of high-repetition-rate femtosecond pulses throughout the visible and IR by using frequency conversion in crystals. The 80-MHz femtosecond optical parametric oscillator permits broad tunability throughout the near IR and mid-IR.[2,3] High-repetition-rate femtosecond pulse generation in the UV and blue–green has been somewhat more limited. Colliding-pulse mode-locked (CPM) lasers have directly generated <100-fs pulses in the range of 493 to 554 nm at milliwatt outputs,[4,5] and intracavity doubling of the Rhodamine 6G/diethyloxadiacarbocyanine iodide (Rh6G/DODCI) CPM dye laser has resulted in a 100-MHz source of femtosecond pulses with milliwatt outputs in the 310–315-nm range. The Rh6G/DODCI CPM laser was first intracavity doubled by using KDP.[6] Soon thereafter, β-BaB_2O_4 (BBO) was used to intracavity double the CPM laser with a per-pass conversion efficiency as high as 5.5%, which generated 20 mW of UV output per arm with <100-fs pulse widths, and pulse widths as short as 43 fs.[7] This gives an effective conversion efficiency of nearly 100% from the typical CPM output in the red to the UV.

While the standard Rh6G/DODCI CPM dye laser operates at a wavelength slightly shorter than the tuning range of the Ti:sapphire laser, the broad tunability, the high average output power, and the obvious advantages of a solid-state laser have made the dispersion-compensated mode-locked Ti:sapphire laser[8] an extremely attractive replacement for the CPM dye laser. At present, the mode-locked Ti:sapphire laser can potentially operate with <200-fs pulse widths and >100-mW average power over the range of 700 to 1053 nm.[9] Frequency doubling over this spectral range provides femtosecond pulses from 350 to 525 nm. Doubling of the Ti:sapphire laser outside the cavity has been reported.[10] The best conversion efficiency of 25% was achieved at 750 nm, although no second-harmonic pulse widths were reported and the length of the doubling crystal was not given. The group-velocity mismatch for type I second-harmonic generation (SHG) in BBO at 750 nm is 225 fs/mm, and in order to maintain the narrowest temporal pulse width a thin doubling crystal is required. Use of a thin crystal therefore necessitates a high peak power to achieve high conversion efficiency, and thus intracavity doubling is required to achieve simultaneously the shortest pulses and the highest power in the second harmonic. As discussed further below, extremely high intracavity conversion efficiency is possible, which would result in UV, blue, or green outputs of hundreds of milliwatts average power. Using an extremely thin (55 μm) crystal of BBO, we demonstrate a 72-MHz repetition-rate source of blue pulses of 89-fs duration (FWHM) and 115 mW of power per arm (two arms of BBO; see Fig. 1). Reducing the pulse width for the blue to 54 fs, we measure 45 mW of power per arm.

Figure 1 shows a schematic of the dispersion-compensated intracavity-doubled Ti:sapphire laser. The SF-10 prisms are spaced 50 cm tip to tip. The

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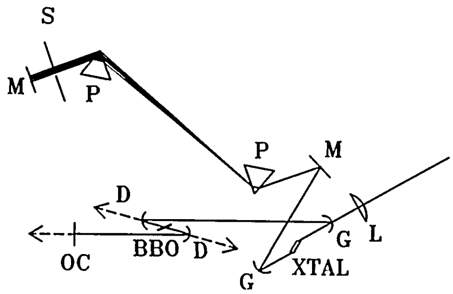


Fig. 1. Schematic of the intracavity doubled Ti:sapphire laser. XTAL, Ti:sapphire crystal; G's, gain mirrors; L, focusing lens; P's, SF-10 prisms; M's, flat mirrors; D, dichroic mirror; BBO, doubling crystal; S, tuning slit; OC, output coupler.

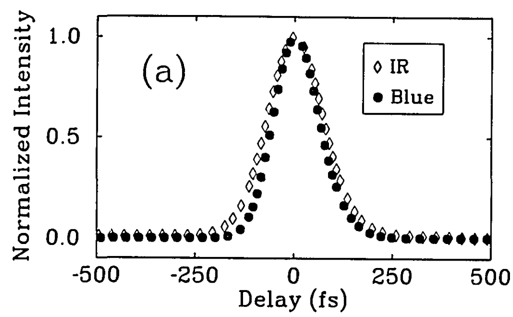
argon pump laser is focused by a 10-cm focal-length lens through one of the $r = 10$ cm gain mirrors onto the 18-mm-long titanium-doped (0.1%) sapphire crystal. The additional intracavity focus at the BBO crystal consists of $r = 5$ cm dichroic mirrors (fused-silica substrates, $R = 100\%$ at 860 nm, $T = 70\%$ at 430 nm). The outcoupler has $T = 1\%$ for the IR and was replaced by a high reflector when the highest power in the blue was generated. Before insertion into the laser cavity, the crystal is aligned for maximum SHG conversion efficiency in the extracavity beam of the mode-locked Ti:sapphire laser operating at the intended doubling wavelength of ~860 nm. The proper alignment of the BBO can be preserved on insertion into the laser cavity.

Pulse-width measurements for both the fundamental (IR) and the second-harmonic light are made by autocorrelation with collinear type I SHG in BBO. The BBO crystal used to measure the IR autocorrelation has a thickness of 0.8 mm and is cut for a phase-matching angle of $\theta = 27.5°$. The BBO crystal used to measure the blue pulse widths has a thickness of 0.67 mm and is cut at $\theta = 69°$. The second harmonic of the blue (215 nm, the fourth harmonic of the Ti:sapphire) is passed through a 0.2-m monochromator and detected by a solar-blind photomultiplier tube. The spectra for the fundamental and second-harmonic outputs from the laser are measured by using a 0.25-m monochromator to disperse the light onto an optical multichannel analyzer.

We point out that the type I SHG cutoff wavelength in the blue for BBO is 409 nm. Below this wavelength, accurate pulse-width measurement requires a more difficult technique such as cross correlating the fundamental beam with the second-harmonic beam by using phase-matched sum-frequency generation. Owing to the significant group-velocity mismatch between the fundamental and second-harmonic pulses for fundamental wavelengths below 820 nm (the group-velocity mismatch is >170 fs/mm for BBO at $\lambda_{IR} = 820$ nm and increases for shorter wavelengths), a thin cross-correlation crystal is required.[7] Thus, for the convenience of using collinear type I SHG autocorrelation to measure the pulse width of the doubled light, we operated the Ti:sapphire laser at $\lambda > 820$ nm.

The intracavity-doubled mode-locked laser is started by a slight mechanical perturbation, usually by a small-amplitude, gentle back-and-forth translation of one prism. Once well aligned, the mode-locked laser operates stably indefinitely (observed for as much as ~6 h), although significant mechanical perturbation can stop mode-locked operation. The mode locking generally is not self-starting. Variation of the intracavity dispersion compensation permits control of the pulse width. On starting, the laser is pushed to shorter pulses simply by adding prism glass and adjusting the focusing slightly to maintain high stability. While the laser stability is excellent even at the longer pulse widths, the oscilloscope trace of the IR mode-locked pulse train indicates somewhat quieter operation as the pulse width is decreased. The spatial mode of the fundamental beam is TEM_{00} with faint, simple higher-order modes superimposed. The blue beam mode is a clean TEM_{00} that shows no sign of higher-order modes, thus verifying that the power of the fundamental lies almost entirely in the TEM_{00} mode.

When the laser is run with a high reflector in place of the outcoupler, 107-fs IR pulses produce 230 mW of second harmonic. Without the intracavity doubling crystal, the maximum output of the mode-locked Ti:sapphire laser operating at 860 nm is ~300 mW for 4.4-W pump power; thus generation of 230 mW of blue power gives an effective conversion efficiency of ~75% from the IR output typical at this pump power. The dichroic mirrors transmit ~72 mW of power per arm of the blue second-



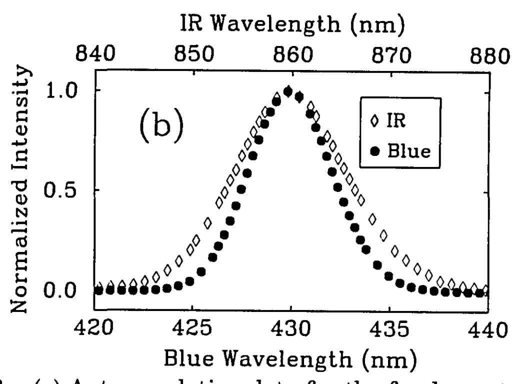


Fig. 2. (a) Autocorrelation data for the fundamental and second-harmonic pulses in the longer-pulse limit. The FWHM for the fundamental is 107 fs, and for the second harmonic it is 89 fs. (b) Spectra for the fundamental and second-harmonic beams. The FWHM for the fundamental is 12.7 nm, which gives $\Delta\nu\Delta t = 0.55$, and for the second harmonic it is 4.9 nm, which gives $\Delta\nu\Delta t = 0.71$.

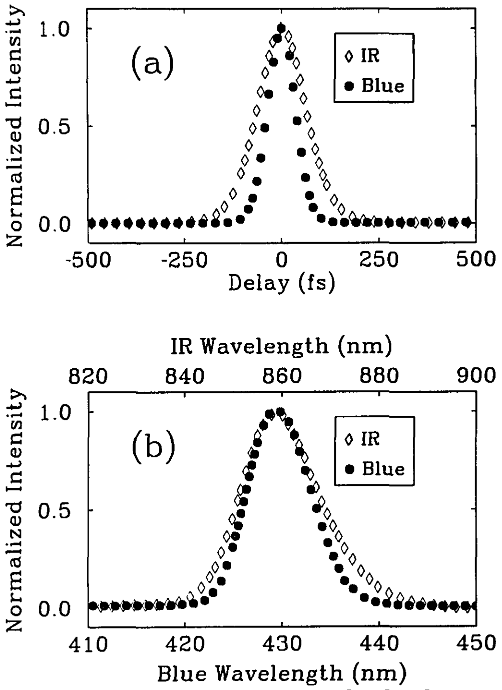


Fig. 3. (a) Autocorrelation data for the fundamental and second-harmonic pulses for the shortest second-harmonic pulses. The FWHM for the fundamental is 93 fs, and for the second harmonic it is 54 fs. (b) Spectra for the fundamental and second-harmonic beams. The FWHM for the fundamental is 18.6 nm, and for the second harmonic it is 7.7 nm. This gives $\Delta\nu\Delta t = 0.70$ for the fundamental and $\Delta\nu\Delta t = 0.67$ for the blue second-harmonic pulses.

harmonic light. On compression of the blue pulses by a dispersion-compensating prism pair, a pulse width of 89 fs is measured (see Fig. 2). The prism pair allows compensation for the dispersion of the dichroic mirror substrate and of other intracavity optics as well as for any upchirp that the pulses may have on generation in the intracavity BBO crystal. The IR pulses are not extracavity dispersion compensated. The spectral FWHM's of the IR and blue are 12.7 and 4.9 nm, respectively, which give $\Delta\nu\Delta t = 0.55$ for the IR and $\Delta\nu\Delta t = 0.71$ for the blue pulses. Pulse widths and time–bandwidth products are determined assuming a $\mathrm{sech}^2(t)$ intensity envelope.

We achieved the shortest blue pulses when running the laser with a 1% outcoupler in place of the high reflector and operating closer to net zero intracavity group-velocity dispersion (see Fig. 3). The power of the IR coupled out is 27 mW, whereas the blue power transmitted by the dichroic mirrors is ~31 mW per arm. The extracavity dispersion-compensated blue pulses have a FWHM of 54 fs and a spectral FWHM of 7.7 nm, which gives $\Delta\nu\Delta t = 0.67$. The IR pulses (which again are not extracavity dispersion compensated) have a pulse width of 93 fs and a spectral FWHM of 18.6 nm, which yields $\Delta\nu\Delta t = 0.70$. It is believed that the IR pulses may be compressed by an extracavity two-prism sequence, and we hope to verify this in the near future. Again, a $\mathrm{sech}^2(t)$ intensity envelope is assumed.

The observed intracavity SHG conversion efficiency of 3.2% per pass for the shortest blue pulses agrees well with the theory (3.5%) for conversion by a nondepleted pump wave.[11] Without the intracavity BBO crystal, we have observed stable mode-locked operation for <100-fs pulses at intracavity powers as high as 8 W. For the same focusing and BBO crystal length presented here, 8 W of intracavity power at a 110-fs pulse width would yield a more than fourfold increase in the output of the second harmonic, or ~500 mW of blue light. For this case, the peak intracavity intensity at the focus would approach the reported single-shot damage threshold for BBO of 50 $\mathrm{GW/cm^2}$.[12] However, this threshold pertains to pulses of 8-ns duration, and we expect the threshold to increase by orders of magnitude for the 100-fs pulse-width regime. The average intensity is orders of magnitude below the long-term damage threshold for BBO.[12]

In conclusion, we have demonstrated highly efficient intracavity doubling of a mode-locked Ti:sapphire laser that yields a source of femtosecond pulses in the blue with the same high repetition rate of 72 MHz, short pulse width, excellent beam quality, and power in the blue representing appreciable recovery of the typical IR output at this 4.4-W pump level. This research represents an extension of intracavity doubling to solid-state mode-locked lasers and results in a source of femtosecond pulses potentially tunable from the near UV into the green, thus broadly expanding the potential spectral range for femtosecond pulses.

The authors thank W. S. Pelouch, P E. Powers, and D. C. Edelstein for helpful conversations. This research was supported by the Joint Services Electronics Program and the National Science Foundation.

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