Generation of Synchronized Ultraviolet and Red Femtosecond Pulses by Intracavity Frequency Doubling

GLENN FOCHT and M. C. DOWNER

Abstract—**We efficiently extract an ultraviolet femtosecond pulse train of milliwatt average power and 100 MHz repetition rate from a colliding pulse mode-locked dye laser by intracavity frequency doubling in KDP. The ultraviolet and visible outputs, which are comparable in power and pulse duration, are perfect synchronized with each other.**

FOR a number of years, the colliding pulse mode-locked dye laser operating at 620 nm[1], with subsequent modifications [2], was the only available source of optical pulses less than 100 fs in duration. Recently several investigators [3]–[8] have developed new femtosecond dye lasers which operate at wavelengths ranging from green to near infrared. However, femtosecond source lasers in the blue and ultraviolet have not yet been developed; furthermore, schemes for *synchronized* generation of unamplified femtosecond pulses at differenct wavelengths have not been realized. Consequently, generation of femtosecond pulses at wavelengths beyond the currently available range of sources, and/or synchronized generation of pulses at different wavelengths, must be achieved through a cumbersome amplification process at a much lower repetition rate than the source laser, followed by wavelength shifting through nonlinear wave mixing or white light continuum generation [9].

We have developed a unamplified source of synchronized red and ultraviolet pulse trains of milliwatt average powers and 100 MHz repetition rate by intracavity frequency doubling in a passively mode-locked femtosecond dye laser. While our results have been obtained with a colliding pulse mode-locked ring laser, the same technique can extend the operation to other types of femtosecond dye lasers to ultraviolet wavelengths. We obtain efficient ultraviolet generation by utilizing the high intracavity power of the fundamental red beam appropriately focused into a KDP doubling crystal, which is thin enough (1 mm) to minimize broadening of the second harmonic pulses caused by group velocity walk-off and phase-matching brandwidth limitation. Insertion of the doubling crystal leaves the operating characteristics of the dye laser virtually unperturbed, apart from minor effects detailed below. The power of the extracted ultraviolet beam ($\sim$ 1 mW) is sufficient to be easily measured with a calibrated power meter, and is brightly visible on a surface such as an ordinary business card with fluoresces in response to ultraviolet illumination. This ease of observation and measurement greatly facilitates experimental alignment of the extracted ultraviolet beam. Intracavity frequency doubling has been applied previously to synchronously mode-locked picosecond dye lasers [10]–[12]. Our results, however, represent the first application in the femtosecond doman.

Our basic dye laser cavity follows the design of Valdmanis *et al.* [2], which includes an intracavity configuration of four prisms [13] for control of group velocity dispersion. We introduce the doubling crystal near the focus of an additional intracavity subresonator formed by the two slightly off-axis 5 cm focal length spherical mirrors, as depicted in Fig. 1. These mirrors focus the intracavity red beam to a measured diameter of approximately 60 μm in the plane of incidence at the KDP crystal. We observe a nearly circular generated ultraviolet beam profile (minor axis/major axis $\simeq 2/3$), indicating a nearly circular fundamental beam profile at the focus. The modest asymmetry arises from astigmatism introduced by the use of off-axis spherical mirrors throughout the laser cavity. The beam waist dimensions also vary slightly with particular cavity alignment. The doubling crystal was cut for Type I phase-matched second harmonic generation at Brewster angle incidence in order to minimize reflective insertion losses. By using doubling crystals which are compatible with antireflection coatings, normal incidence insertion of the crystal would also be possible. This would allow easy insertion and removal of the crystal without deflecting the intracavity beam path. We extract the second harmonic pulses, after a single pass of the intracavity red pulses, through one of the subresonator mirrors, which has been coated for 99.9 percent reflectivity at 620 nm and 65 percent transmission at 310 nm. The beam is then recollimated by an extracavity fused silica lens. By using a similar dichroic mirror for the opposing subresonator mirror, an equivalent ultraviolet beam could be extracted in the opposite direction.

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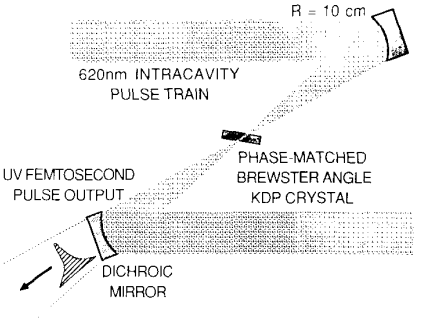

Fig. 1. Schematic of the subresonator for intracavity frequency doubling, showing the bidirectional red pulse train, Brewster angle cut KDP crystal, and the dichroic output coupling mirror for the generated ultraviolet beam. The remainder of the complete cavity configuration follows the design depicted by Valdmanis *et al.* [2].

In the KDP crystal, the intracavity red pulses ($\sim$ 5 nJ in energy) reach a peak intensity of nearly 10^9 W/cm^2. For a crystal thickness of 1 mm at the phase-matching angle, a second harmonic conversion efficiency of approximately 0.3 percent, or an ultraviolet pulse energy of about 15 pJ, is readily computed using standard formulas [14]. This computation assumes Gaussian spatial and temporal profiles for the incident and second harmonic pulses, and takes into account the partial phase mismatch in the wings of the pulse spectrum. The result shows that the intracavity pulse parameters are almost ideally suited for achieving the maximum second harmonic conversion efficiency in a 1 mm crystal consistent with the continuing operation of the dye laser. Our somewhat lower measured ultraviolet pulse energy of 7 ± 2 pJ, or about 10^7 photons/pulse, results primarily from ultraviolet reflective losses at the dichroic extraction mirror, the fused silica recollimation lens, and the KDP exit face. With more efficient doubling crystals, conversion efficiencies as high as 1–2 percent should be readily achievable.

With appropriate adjustments of the intracavity prisms after insertion of the doubling crystal, we preserve a typical red pulsewidth of 60–70 fs, as shown by the background-free autocorrelation traces with and without the doubling crystal in Fig. 2(a). Similar compensation for the broadening effect of dispersive intracavity elements has been observed previously with acoustooptic cavity dumpers [15]. Furthermore, insertion of the doubling crystal does not limit the frequency bandwidth of the fundamental red pulses, as shown by the comparison of spectra with and without the doubling crystal in Fig. 2(b). The small spectral shift evident in Fig. 2(b) results from slight intracavity mirror adjustments required upon insertion of the doubling crystal, rather than from an inherent effect of the doubling crystal itself. We have observed slight shifts in both directions or no shift at all, depending on the particular cavity alignment. Both the pulse duration and bandwidth measurements were performed with the intracavity doubling crystal tuned for maximum ultraviolet output power. Detuning from the optimum phase-matching angle had no observable effect on either quantity.

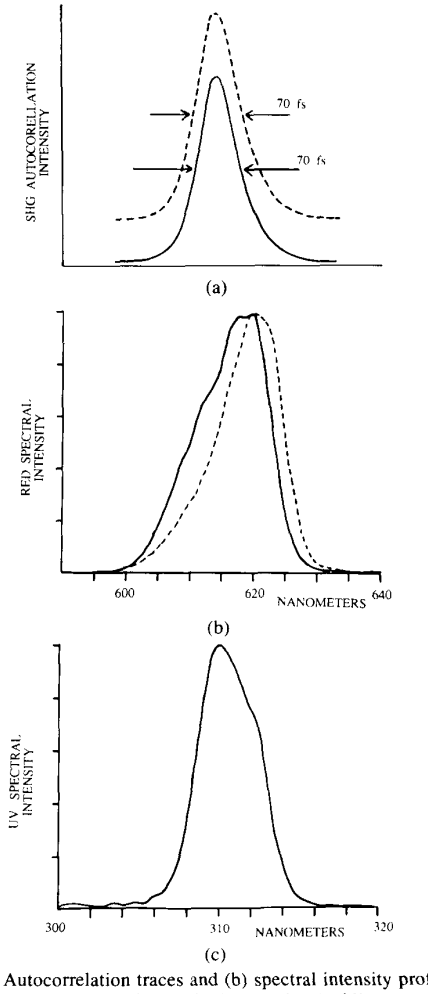

Fig. 2. (a) Autocorrelation traces and (b) spectral intensity profiles of the fundamental red pulses without the intracavity doubling crystal (dashed curves) and with the intracavity doubling crystal tuned for maximum output power (solid curves). A sech^2 temporal pulse profile has been assumed in arriving at the pulse duration shown in (a). (c) Spectral intensity profile of the generated ultraviolet pulse.

These observations contrast sharply with previous observations of significant pulse broadening and bandwidth limitation caused by intracavity doubling crystals in synchronously mode-locked picosecond dye lasers [10]–[12]. We attribute the more favorable performance of our frequency-doubled laser to the presence of a saturable absorber, which compensates the pulse broadening effect of power-dependent loss in the doubling crystal. Quantitative analysis of this compensating effect is in progress.

The measured frequency spectrum of the ultraviolet beam is shown in Fig. 2(c). The full width at half maximum of 5 nm is consistent with a transform-limited pulse duration of 40 fs. Group velocity walk-off and phase-matching bandwidth limitation in the doubling crystal, however, broaden the ultraviolet pulse from this ideal transform-limited duration. We have quantitatively accounted for these effects by calculating the freqency-dependent amplitude and phase of the second harmonic pulse

for each spectral component of a 70 fs, Gaussian fundamental input pulse using standard formulas [14]. The Fourier transform of the resulting second harmonic pulse spectrum yields the temporal ultraviolet pulse envelope. We find that the ultraviolet pulse is broadened to approximately 170 fs, for a 1 mm KDP crystal, in agreement with similar calculations by others [16]. The same calculation shows, however, that a 0.1 mm $LiIO_3$ crystal would yield 90 fs ultraviolet pulses, and that a 0.22 mm β-BaB_2O_4 crystal would yield 95 fs pulses with the same conversion efficiency as a 1 mm KDP crystal.

Insertion of the doubling crystal has no discernible effect on the power of the red beam nor on the mode-locking stability of the dye laser. We observe a 50 percent increase in the practical argon laser pump power to between 3.0 and 4.0 W, an increase caused by residual intracavity reflection losses from the crystal, minor diffraction, absorption, and scattering losses caused by optical imperfections in the crystal, and losses due to second harmonic generation. In addition, we observe a broadening of the hysteresis which normally characterizes the dye laser threshold as the pump power is ramped upwards and downwards. Normally, the "turn-on" threshold, which is observed as the pump power increases from zero, is approximately 0.3 W higher than the "turn-off" threshold, observed as the pump power decreases with the dye laser opening. With the doubling crystal in place, this hysteresis widens to approximately 1–1.5 W.

The dye laser can "blink" off unpredictably when operated at a pump power within this hysteresis window. We attribute this behavior to a thermooptic lensing effect, normally caused by the heating of the gain jet by the pump laser, and augmented here by the heating of the KDP crystal by the intracavity beam. A more pronounced hysteretic effect of similar origin has been observed and analyzed previously with intracavity frequency doubling crystals in solid-state lasers [17], [18] with higher intracavity power. As the pump power and intracavity beam power change, the effective focal length of the thermally induced index lenses change, thus changing the optical cavity alignment and the lasing threshold. Although the light power absorbed in the KDP crystal is three orders of magnitude smaller than the gain jet, the thermooptic lensing effect is larger in magnitude because of its tenfold smaller thermal conductivity ($0.021\ W \cdot cm^{-1} \cdot K^{-1}$), its ten-fold larger thermal index gradient dn/dT ($3.34 \times 10^{-5}\ K^{-1}$), and its threefold greater thickness. Furthermore, the jet is flowing at approximately $7\ m \cdot s^{-1}$, which removes the heat 10^4 times faster than by heat conduction. The major practical effects of the increased thermooptic effects are a higher required pump power and a temporary increase in the tendency of the dye laser to "blink" off when first brought into operation. However, once a thermal steady state is reached within several minutes, this latter tendency typically disappears, and any residual thermooptic effect can be compensated by adjustment of the subresonator mirror separation. These minor effects can be alleviated by using a doubling crystal with smaller residual absorption, higher thermal conductivity, or smaller dn/dT.

In summary, intracavity frequency doubling provides a simple, inexpensive, and virtually non-perturbative method of extending the operation of femotosecond lasers to ultraviolet wavelengths. Such wavelength-extended femtosecond lasers should be applicable to time-resolved photoionization and photoemission experiments. In addition, the tighter focusability of ultraviolet beams can enhance the spatial precision of electrooptic sampling and optoelectronic switching in the micron and submicron gaps between microelectronic circuit elements. Type I phase-matched sum frequency mixing of 620 nm fundamental pulses with 310 nm second harmonic pulses to generate 205 nm pulses has become possible for the first time with the recent development of the β-BaB_2O_4 crystal [19], [20]. The intracavity frequency-doubled output of the colliding pulse mode-locked laser may serve as a seed pulse for injection into the recently developed XeCl subpicosecond pulse amplifer [21]. To date, such amplifiers have required injection pulses of nanojoule to microjoule energies, formed by first amplifying the fundamental dye laser output before frequency doubling [22], in order to compete successfully with amplified spontaneous emission (ASE) within the XeCl gain cell. Development of improved methods for suppression of ASE, such as spatial filtering, and use of lower gain amplifier cells, however, may permit injection with the lower energy pulses from the source described here, thus circumventing the need for a visible wavelength dye amplifier.

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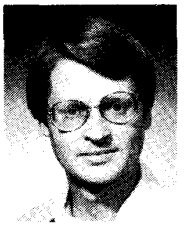

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