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Generation of 3-ps pulses by spectral selection of the continuum generated by a 25-ps second harmonic Nd:YAG laser pulse in a liquid

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High-power ultrashort light pulses propagating in a condensed medium experience self-phase modulation (SPM) leading to the generation of a broadband light continuum.¹ Recent theoretical analyses have concluded that the Stokes and anti-Stokes shifts should appear in the leading and trailing edge of the pump pulse, respectively.^{2–4} This result was supported by experimental studies where the temporal location of various parts of the continuum was determined using the autocorrelation⁵ and streak camera^{6,7} methods in both the femtosecond and picosecond regimes. Gomes *et al.*⁸ demonstrated a pulse compression technique based on the spectral temporal distribution of an SPM spectrally

16 APPLIED OPTICS / Vol. 27, No. 1 / 1 January 1988



Fig. 1. Schematic diagram of the experimental setup: SHG, second harmonic generator; F1, color filter to remove the fundamental laser wavelength; BS, beam splitter; L1, focusing lens; L2, collimating lens; NB, narrowband filter used to select particular wavelengths within the continuum; M, mirror; D, delay line.



Fig. 2. Streak camera temporal profile of the 530-nm incident laser pulse and 10-nm bandwidth pulse at 580 nm.



WAVELENGTH (nm)

Fig. 3. Pulse duration vs peak transmission wavelength of several narrowband filters. Horizontal error bars correspond to 10-min bandwidths of the filters.

broadened pulse in a fiber. According to their analysis, since the Stokes and anti-Stokes shifts are proportional to the intensity gradients on the sides of the pulse, a window within the broadened spectral profile should eliminate the wings of the generated pulse where the high- and low-frequency components are located. They obtained a threefold shortening of 80-ps pulses to 30 ps from a Nd:YAG laser broadened from 0.3 to 4 Å after propagation through 125 m of optical fiber with a monochromator as a spectral window. Recently, the continuum generated in CCl₄ by a weakly focused 8-ps laser pulse was measured by a 2-ps resolution streak camera at several wavelengths.⁹ The continuum was found to have a shorter duration than the pump (~6 ps) and to be generated over a local spatial domain in the liquid cell.

In this Letter, we demonstrate a method to generate pulses down to 3 ps using a continuum spanning 1400 Å generated in D_2O by an intense 25-ps Nd:YAG second harmonic laser pulse. This achieves both pulse shortening and wide tunability. The duration of the compressed pulse was measured using a 2-ps resolution streak camera system.

The experimental arrangement used to generate and measure the continuum pulse is shown in Fig. 1. The setup consists of a passively and actively mode-locked Quantel Nd:YAG laser system with a single-pulse selector, a doublepass ring amplifier, a KDP second harmonic generator, the continuum generator, an optical delay, a streak camera, and a temporal analyzer. The second harmonic 25-ps 5-mJ pulse from the laser system was divided by a 5% reflecting beam splitter into two beams. The stronger beam was used to generate the continuum pulses. The weaker one was used as a prepulse to provide a relative time scale for the continuum pulse and to determine the pulse duration of the second harmonic pulse. The continuum was generated by focusing the laser beam with a 20-cm focal length lens into a 5-cm cell filled with D_2O . The continuum spanned from 450 to over 600 nm with a minimum average energy of at least 20 nJ/nm across the spectrum. Narrowband filters were inserted after the D₂O cell to select a particular spectral window. Measurements were made with 10-nm bandwidth filters at several wavelengths. The pump and prepulse beams were then combined, optically delayed, and directed into a Hamamatsu model C1587 streak camera, where the separation between the two beams and pulse duration was measured. The streak camera was set at a 300-ps full sweep range. The minimum resolution of the camera was $\sim 2 \text{ ps}^{10}$

Figure 2 gives a temporal display of both the second harmonic laser pulse and the part of the continuum pulse selected by after a 10-nm bandwidth narrowband filter centered at 580 nm. The pulse duration at 530 nm is 24 ps, while the pulse derived from the continuum is clearly much shorter. The measured pulse duration (FWHM) is $\sim 3 \pm 1$ ps and is almost limited by the resolution of the streak camera; thus a minimum compression ratio of 8 is achieved.

In Fig. 3 the pulse duration is plotted as a function of the peak transmission wavelength of different narrowband filters. Each point on the curve is the average of at least six separate measurements. No deconvolution of data for the prompt 2-ps response was performed. The pulse durations were <4 ps except close to the pump laser wavelength (530 nm) where the signal pulse duration was equal to the input pulse duration (>25 ps). This is expected since a very small portion ($\approx 1\%$) of the laser energy is converted into continuum with such long input pulses,¹ and the signal at 530 nm consisted mainly of the laser pulse.

To identify the generation mechanism the temporal distribution of the continuum spectrum was determined by measuring the time delay between the continuum and a reference





beam at different wavelengths. The results are displayed in Fig. 4. The time delay was ~ 22 ps for a 140-nm change in wavelength as predicted by the SPM mechanism the Stokes wavelengths led the anti-Stokes wavelengths.⁶ The delay due to group velocity over a 5-cm D₂O cell for the 140-nm wavelength change is <4 ps. The remaining 18 ps is well accounted for by the SPM mechanism using a 25-ps (FWHM) pulse and the stationary phase method.⁶ Furthermore, a 10-nm change in the temporal distribution curve corresponds to an ~2.6-ps width matching the measured pulse duration. This suggest that by using narrower bandwidth filters the pulse duration can be shortened nearly to the uncertainty limit.

In conclusion, a method to generate short pulses using a 5cm D_2O cell for SPM continuum generation and narrowband filters for spectral selection was demonstrated. The pulse duration was measured by a 2-ps time resolution streak camera. Eightfold pulse shortening was obtained resulting in the generation of 3-ps laser pulses between 460 and 600 nm for 25-ps Nd:YAG second harmonic laser pump pulses. Further amplification of the short pulses using dyes is possible.

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