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Temporal distribution of picosecond super-continuum generated in a liquid measured by a streak camera

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When a high power picosecond laser pulse propagates through a condensed medium, it can produce an output which is nearly a white light continuum.[1] This supercontinuum phenomenon was first observed and attributed to self-phase modulation (SPM) by Alfano and Shapiro[1] in 1970. The temporal and spectral properties of the supercontinuum light source are important properties to be understood to enhance the generation process and to compress laser pulses. Using the stationary phase method,[2] it was theoretically demonstrated that the temporal location of the Stokes and anti-Stokes shifts should appear in the leading and trailing edge of the pump pulse, respectively. Recently, theoretical analyses by Stolen and Lin,[3] Yang and Shen,[4] and Manassah *et al.*[5] obtained similar conclusions. The latter two groups explained the asymmetry observed in the anti-Stokes sweep.[1] Recently, propagating an 80-fs laser pulse through a 500-μm thick ethylene glycol jet stream, Fork *et al.*[6] measured the pulse duration and location of spectrum in time by the autocorrelation method. Their results supported the SPM mechanism for supercontinuum generation. In this Letter, the temporal distribution of various wavelengths for the supercontinuum generated in CCl_4 by an intense 8-ps mode-locked glass laser pulse was measured using a 2-ps resolution streak camera system. These experimental results attributed the frequency spread to the SPM model.

The experimental arrangement used to generate and measure the supercontinuum pulse is shown in Fig. 1. The setup consists of a mode-locked Nd:glass laser system with a single pulse selector, an amplifier, and a KDP second harmonic generator, supercontinuum generator, optical delay, streak camera, and an OMA2 for data processing. A single pulse at 530 nm of 8-ps duration was divided into two beams. A main beam (M-beam) was used to generate the supercontinuum pulse and a reference beam (R-beam) was used to provide a relative time scale for the continuum pulse. The M-beam was focused into a 20-cm long cell filled with CCl_4 to generate the supercontinuum pulse. Color filters F_2 and narrowband filters F_3 were inserted after the CCl_4 to remove the unwanted wavelengths and to select a particular wavelength. In the Stokes side measurement, F_2 consisted of three Corning 3(3-67) color filters. In the anti-Stokes side measurement, F_2 consisted of three Corning 5-57 filters. F_1 was a set of neutral density filters in the R-beam. After the wedge W, the M-beam (supercontinuum pulse) and the R-beam (530-nm pulse) traveled along the same path. Both beams were optically delayed and directed into a Hamamatsu model C1587 streak camera, where the time separation between the M-beam and R-beam was measured. The streak camera was set at 300-ps full sweep range. The minimum resolution of the camera is almost 2 ps.[7]

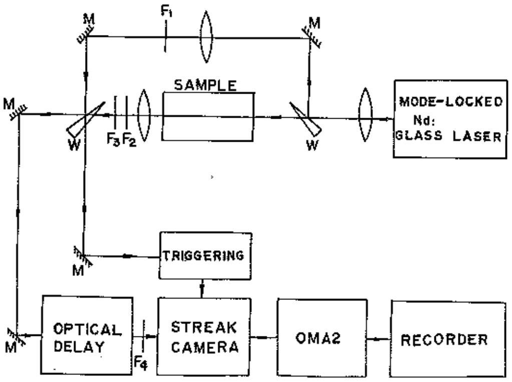

Fig. 1. Schematic diagram of the setup used to measure the supercontinuum laser pulse temporal distribution: W, wedge; M, mirror; F_1, neutral density filters; F_2, 3(3-67) Corning filters for Stokes side; F_2, 3(5-57) Corning filters for anti-Stokes side; F_3, narrowband filter set.

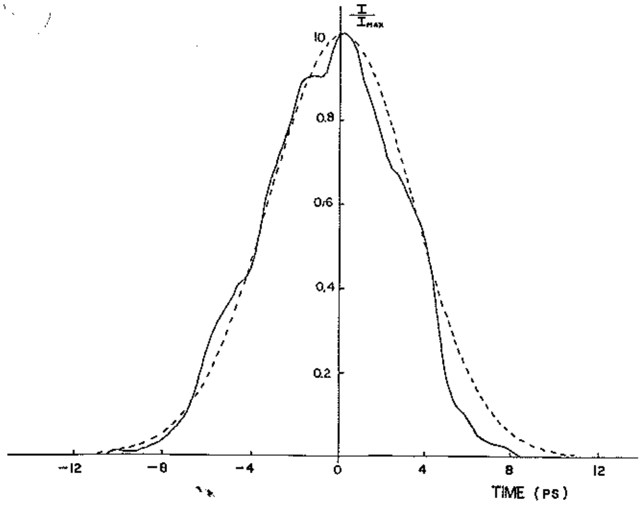

Fig. 2. Temporal profile of a 530-nm incident laser pulse measured by a 2-ps resolution streak camera. The dashed line is a theoretical fit to an 8-ps FWHM Gaussian pulse.

The incident 530-nm laser pulse temporal profile is shown in Fig. 2. The pulse shape can be fitted with a Gaussian distribution[8] with duration τ(FWHM) = 8 ps. The spectral and temporal distributions of the supercontinuum pulse

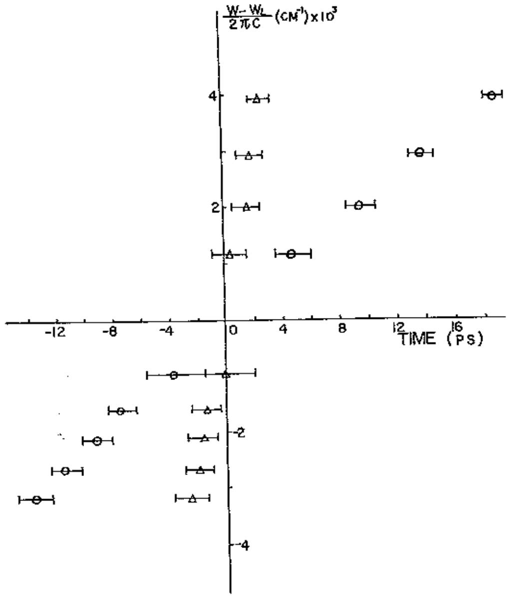

Fig. 3. Measured supercontinuum temporal distribution at different wavelengths: ○, data points with correction of the optical path in filters; △, data points with the correction of both the optical path in filters and group velocity dispersion in liquid.

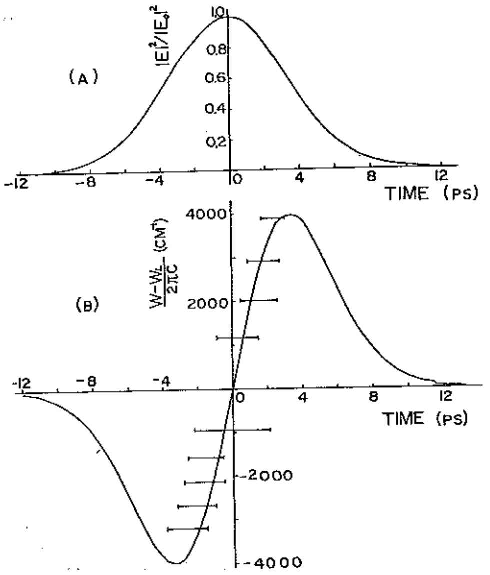

Fig. 4. Comparison of the measured temporal distribution of supercontinuum with a SPM model. The horizontal axis is the time in picoseconds. (A) A theoretical incident laser pulse with 8-ps FWHW Gaussian temporal profile. The vertical axis is an arbitrary intensity scale. The electronic nonlinear index of refraction change δn in condensed matter has the same temporal profile as the incident laser pulse. (B) Comparison of generated supercontinuum frequency change as a function of time with the SPM model. The time axis (B) matches the time axis of the corresponding incident laser pulse in (A). The vertical axis is the frequency shift of the supercontinuum. The displayed data have been corrected for both the optical path in filters and the group velocity dispersion in the CCl$_4$. The solid line is a calculated curve from Eq. (1), $\omega - \omega_L = -(\omega_L l n_2/c)\partial|E|^2/\partial t$, where ω_L is the laser frequency and ω is the supercontinuum frequency. To fit the experimental data, the following parameters have been chosen: $V = w_L n_2 l/c = 4.5 \times 10^{-7}$ and peak intensity = 3.6 $\mathrm{TW/cm}_2$.

were obtained by measuring the time difference between the M- and R-beams. The measured results are shown as circles in Fig. 3. Each data point corresponds to an average of about six laser shots. This observation is consistent with the SPM and group velocity dispersion. To determine the temporal distribution of the wavelengths generated within a supercontinuum, the group velocity dispersion effect[9] in CCl$_4$ was corrected. For example, the time delay $\Delta\tau$ due to group velocity dispersion between $\lambda = 530$ nm and $\lambda = 440$ nm is $\sim$0.87 ps/cm. Results corrected for both the optical delay in the added filters and the group velocity in CCl$_4$ are also displayed as triangles in Fig. 3. The salient feature of Fig. 3 indicates that the Stokes wavelengths of continuum lead the anti-Stokes wavelengths.

Using the stationary phase SPM method,[2,10] the generated frequency ω of the supercontinuum can be expressed by

$$w(t) - \omega_L = -(\omega_L l/c)\partial(\delta n)/\partial t, \tag{1}$$

where ω_L is the incident laser angular frequency, l is the length of the sample, and δn is the induced nonlinear refractive index n_2E^2. By choosing appropriate parameters to fit the experimental data of Fig. 3, a theoretical calculated curve for the sweep is displayed in Fig. 4. An excellent fit using a stationary phase model[1,2] up to maximum sweep demonstrates that the generation mechanism of the temporal distribution of the supercontinuum arises from the SPM. The missing portion of the curve in Fig. 4(b) from 5 to 10 ps and -5 to -10 ps can be accounted for by the lower incident intensity of the SPM signal. If there were SPM generation from the far wing of the incident laser pulse, it would be difficult to measure. For example, in the theoretical calculation of Fig. 4, $\Delta\omega = (\omega - \omega_L)/2\pi c = 2000$ cm^{-1} can be generated at +1 and +6.5 ps. However, due to the intensity difference at $|E(t = 1 \text{ ps})|^2 \approx 6|E(t = 6.5 \text{ ps})|^2$, the signal generated at the far wing of the laser pulse would be too weak to be measured. The signal would be buried under the near wing of the supercontinuum pulse at the same SPM wavelength. This would be difficult to determine. Due to the linear chirp about the peak shown in Fig. 4, a pair of gratings could be inserted into the output to compress the duration of a supercontinuum band into a pulse much shorter than the incident laser pulse.[11]

In conclusion, the temporal distribution of various wavelengths of the supercontinuum pulse generated from an 8-ps mode-locked glass laser has been measured by a 2-ps time resolution streak camera. The experimental result clearly supports the SPM mechanism and demonstrates one of SPM signature: positive chirp.

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