Reprinted from Applied Optics, Vol. *26*, page 2700, July 15, 1987

Supercontinuum pulse generation and propagation in a liquid carbontetrachloride

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Received 7 March 1987.
0003-6935/87/142700-03$02.00/0.

Over the past seventeen years, the generation of the ultrafast supercontinuum (USC) has been investigated[1–8] experimentally and theoretically. The generation location and propagation of USC pulse have not been described in any detail. It is commonly believed that the pulse duration of USC pulse will become broadened due to the group velocity dispersion (GVD). This has been expounded in studies of ultrashort laser pulses in optical fibers. Recently, we have demonstrated new properties of pulse propagation of USC in semiconductors,[7] which appears to have a different dispersion relationship than the well-known group velocity propagation. This has led us to reexamine the USC pulse propagation and generation region. In this Letter, the duration of the incident and the pulse delay of USC pulses generated by an 8-ps laser pulse in a 20-cm long liquid CCl_4 cell were directly measured with a 2-ps resolution streak camera. Information on the temporal behavior of the USC generation, propagation length, and pulse compression in condensed matter is presented.

Dominant mechanisms responsible for the generation of the USC[1,9,10,11] are self-phase modulation (SPM), four-photon parametric generation (FPPG), cross-phase modulation (XPM),[10] and stimulated Raman scattering (SRS). In the SPM process, a newly generated wavelength could have bandwidth-limited duration at the well-defined time location[9,12] in the pulse envelope. While in the FPPG and SRS process, the duration of the USC pulse could be shorter than the pump pulse duration due to the higher gain about the peak of the pulse. In either case, the USC pulse will be shorter than the incident pulse at the local spatial point of generation. After that, these pulses will be broadened in time due to the GVD in a condensed matter. That is, the Stokes wavelengths travel ahead of anti-Stokes wavelengths due to normal dispersion. A salient feature not been discussed in detail is the continued generation of spectra within and outside the interaction length by the pump pulse until the pump depletion sets in.

The experimental setup used to characterize and compress the USC pulses has been previously described.[5,7,11] A mode-locked Nd:glass laser system (an oscillator, single-pulse selector, amplifier, and second harmonic generation), SC generator, optical delay pulse compressor,[11] 2-ps resolution Hamamatsu 1587 streak camera,[13] and an OMA2 for data processing were used. A single 8-ps pulse at 530 nm was weakly focused by an $f = 30$-cm lens into a 20-cm long cell filled with CCl_4 to generate the ultrafast USC pulses. Typical data of the time delay of 10-nm bandwidth pulses centered at 530-, 650-, and 450-nm wavelengths of the USC without compression are displayed in Fig. 1. The absolute time of these three pulses was compared with a reference pulse traveling through air. The peak locations of 530, 650, and 450 nm are −49, −63, and −30 ps, respectively.

The salient features displayed in Fig. 1 indicate that the duration of all 10-nm band USC pulses are only ~6 ps, which is shorter than the incident pulse of 8 ps, and the Stokes side (650 nm) of the USC pulse travels ahead of 530 nm by 14 ps, and the anti-Stokes side (450 nm) of the USC pulse lags the 530 nm by 19 ps.

If the USC could be generated throughout the entire length of the sample (assume that the power density of the incident pulse remains about the same), the Stokes USC pulse generated by a 530-nm incident pulse at $z \sim 0$ cm of the sample would be ahead of the 530-nm incident pulse after propagating through the length of CCl_4. Over this path, 530 nm could continuously generate the USC pulse. Thereby the Stokes USC pulse generated at the end of the CCl_4 coincides in time with the 530-nm incident pulse, and the Stokes USC generated at $z = 0$ travels ahead due to the group velocity dispersion (GVD). In this manner, an USC pulse centered at a particular Stokes frequency could have a pulse duration greater than the incident pulse extending in time

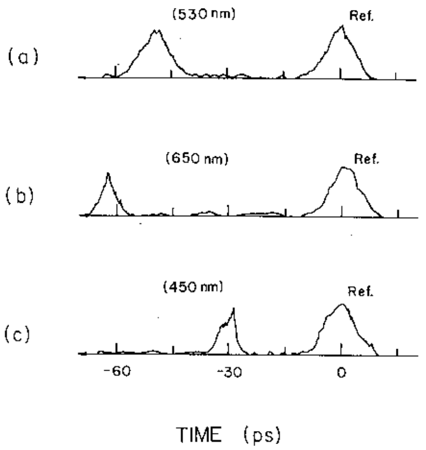

Fig. 1. Temporal profiles and pulse locations of a selected 10-nm band of a supercontinuum pulse at different wavelengths propagated through a 20-cm long CCl_4 cell: (a) $\lambda = 530$ nm; (b) 650 nm; (c) 450 nm. Amplitudes of these pulses are adjusted for display. The time zero is set for a reference pulse traveling in air. Filter effects were compensated. Different wavelengths were selected using narrowband filters of the same thickness.

from the emerging of 530-nm pulse to position where the Stokes frequency originally was produced at $z \sim 0$ cm. In a similar consideration, the anti-Stokes USC pulse will also be broadened in this case at the leading time. However, no slow asymmetric tail for the Stokes or rise for the anti-Stokes pulses are displayed in Fig. 1. These observations suggest the local generation of USC pulses.

A model to describe the generation and propagation features of the USC pusle is formulated based on local generation. The relative time delay of Stokes and anti-Stokes USC pulses to the 530-nm pump pulse is accounted for by the filaments formation at ~5 cm from the sample cell entrance window. The 5-cm location is calculated from data in Fig. 1 by using the equation

$$T_{530} - T_{\mathrm{USC}} = \Delta x(1/V_{530} - 1/V_{\mathrm{USC}}), \tag{1}$$

where Δx is the total length of the USC pulse traveled in CCl_4 after the generation, T_{530} and T_{USC} are the 530-nm and USC pulse peak time locations in Fig. 1, V_{530} and V_{USC} are the group velocity of 530-nm and USC pulses, respectively. T_{USC} has been corrected for the SPM generation process over the pump pulse envelope location.[5] For example, the time separation between 650 and 530 nm in Fig. 1 is 14 ps. However, due to the SPM generation mechanism, a 650-nm USC pulse was introduced at the 3-ps leading edge of the pump pulse. Therefore, $T_{650} = 14\ \mathrm{ps} - 3\ \mathrm{ps}$ for the T_{USC} of Eq. 1 at $\lambda = 650$ nm.

The duration of the USC pulse right at the generation location is either limited by the bandwidth of the measurement from the SPM process or shortened by the parametric generation process. In either case, a 10-nm bandwidth USC pulse will have a shorter duration than the incident pulse. After being generated, each of these 10-nm bandwidth USC pulses traveled through the rest of the CCl_4 liquid and would be continuously generated by the incident 530 nm over a certain interaction length before these two pulses walked off. The interaction length can be calculated as[9,10]

$$l = \tau V_{530} V_{\mathrm{USC}}/(V_{530} - V_{\mathrm{USC}}), \tag{2}$$

where l is the interaction length over which the pump and USC pulses stay spatially coincident by less than the duration (full width at half-maximum) of the incident pump pulse, and τ is the duration of the USC pulse envelope. From Eq. (2), one can estimate the interaction length from the measured τ of the USC pulse. Using parameters $\tau_{\mathrm{USC}} = 6$ ps, $V_{530} = c/1.4868$, and $V_{650} = c/1.4656$, the interaction length $l = 8.45$ cm is calculated. This length agrees well with the measured beam waist length of 8 cm for the pump pulse in CCl_4.

Since no long tails were observed from USC pulses to the dispersion delay times of the Stokes and anti-Stokes USC pulses, the USC was not generated over the entire length of 20 cm but only over $z = 1$–9 cm. This length is equivalent to the beam waist length of the laser in CCl_4. The length of the local SPM generation over a distance of 8.45 cm yields a possible explanation for the 6-ps USC pulse duration. In addition, a pulse broadening of 0.3 ps calculated from the GVD of a 10-nm band at 650-nm USC traveling over a 20 cm of liquid CCl_4 is negligible in our case. Further experimental work to strengthen the proposed USC generation model will be to study the dependence of the location of the USC pulse duration as a function of the beam waist length and focal point.

In a nonguided system such as a liquid, due to strong self-focusing, it may be difficult to lead a perfect linear chirped SPM as in guided fibers. This noncontrollable filament will lead to a large fluctuation in compression value from shot to shot and a failure to obtain the bandwidth-limited short duration in nonguided condensed matter. The following experiments were performed to measure the pulse compression in condensed matter. The measurement of pulse compression of the 530-nm and USC pulses was optically delayed and sent through a pair of parallel volume phase transmission holographic gratings. The spatial frequency of the holograms is 1130 lines/mm. Further details about the gratings are described in Ref. 13. The output compressed USC pulse was also directed into a streak camera to measure the pulse duration.[14] The absolute arrival time of the USC pulse was determined using a reference pulse.[7] Twenty laser shots were used for the average of each experimental condition. Narrowband filters (with 10-nm pass bandwidth) were used in front of the entrance of the streak camera to select the desired wavelength. The location of the second hologram was adjusted on an x-y translation stage. The y-direction variation was used to compensate for the wavelength difference between the alignment He–Ne laser and the USC wavelength. The x-direction adjustment was used to control the separation of gratings to achieve the optimum location for pulse compression. The pulse compression $\Delta\tau$ (defined as the difference between the measured pulse durations before and after compression) of the incident 530-nm laser pulse was determined as a function of the grating pair slanted distance b. When b was <15 cm, $\Delta\tau$ was found to be linearly proportional to b. However, when $b > 20$ cm, $\Delta\tau$ approached a constant of ~3.2 ps (see Fig. 2). The incident pulse duration (8 ps) minus $\Delta\tau$ (3.2 ps) gives an averaged compressed 4.8-ps duration of the 530-nm pulse. This 4.8-ps limitation of the compressed duration of the incident 530-nm pulse may be account for by the imperfect mode locking, multiple pulses substructure,[9] and nonlinear chirp of the laser. However, a 10-nm band of the USC pulse from a CCl_4 liquid of effective interaction length 8.45 cm at 620 nm was compressed down to our time resolution limit. The temporal profile of USC pulse measured with and without the compressor is displayed in Fig. 3. A 10-nm bandwidth USC pulse centered at 620 nm is shown in Fig. 3(a) without compression. The measured average duration of the selected 10-

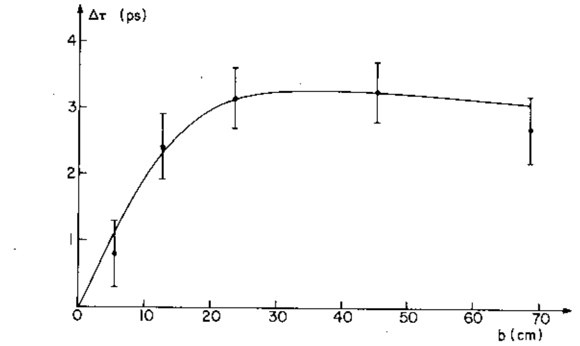

Fig. 2. Compression value of a 530-nmn pulse Δτ (the difference between the measured pulse duration before and after compression) as a function of the slant distance b between two gratings where $\Delta\tau \sim$ 2.5 nm. The laser pulse duration before compression was 8 ps.

nm band USC pulses was 6.4 ± 0.6 ps, which was shorter than the incident 530-nm pulse. Without a 10-nm bandpass filter, the duration of the entire USC was ~30 ps. A compressed 10-nm band centered at a 620-nm USC pulse was measured to be as short as 2 ps [Fig. 3(b)] which was the time resolution of the streak camera.

In conclusion, the USC generated in a 20-cm long CCl_4 liquid by a weakly focused 8-ps 100-μJ laser pulse was measured directly by a streak camera. The USC pulse was found to have a shorter duration than the pump and to be generated over a local spatial domain in the liquid cell. A selected portion of the USC pulse at 620 nm in liquids was compressed down to the time resolution limitation of the streak camera of ~2 ps.

This research is supported in part by NSF-OIR 841322, AFOSR 86-0031, PSC/BHE, and Hamamatsu Photonics K.K.

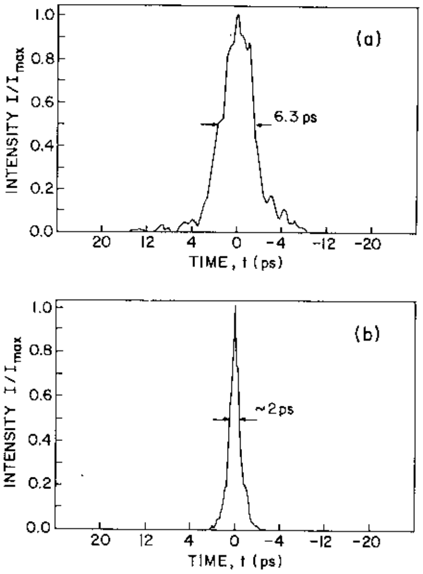

Fig. 3. USC pulse profile center at λ = 620 nm with a 10-nm wavelength bandwidth: (a) pulse profile before compression; (b) pulse profile after compression.

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