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High energy, low repetition rate, photonic crystal fiber generated supercontinuum for nanosecond to millisecond transient absorption spectroscopy

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ABSTRACT

High energy density per pulse (-15 dBm nm^{-1}) supercontinuum (SC) source has been developed as a probe for transient absorption (TrA) spectroscopy of systems with lifetimes from nanoseconds to a few milliseconds. We have generated a 600–1600 nm, broadband SC by pumping a 15 m photonic crystal fiber (PCF) with relatively high power, 7 ns, 1064 nm pulses. The SC generated at peak pump power of 7.1 kW was randomly polarized and maintained a stable output (6.5% rms average power; 9.1% rms shot-to-shot power). Co-pumping with both 1064 and 532 nm light extended the wavelength range of the SC by about 20%, to 500–1700 nm. Power conversion efficiency and spectral flatness were improved as well. In the visible range, the single-pump SC shows a flatness of 5 dB while the dual-pump SC exhibits 3 dB. In the NIR (1100–1600 nm), the flatness in single- and dual-pump configurations were 3 and 2 dB, respectively. Optically induced fiber breakdown was characterized.

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1. Introduction

SC pulses are widely exploited in pump-probe, TrA spectroscopy methods. In these techniques, a molecular system is excited with a laser pulse into a higher energy electronic state, or collection of states, and then the UV/Vis/NIR absorption spectrum is probed as a function of the time delay. The resulting absorption of light by the transient intermediate species yields important structural and dynamical information about photoactive systems. This technique has been useful in elucidating the molecular basis of vision, photosynthesis, solar energy harvesting, photoswitchable molecules, and many other important research areas. Pump-probe methods are such an important application of SCs that the first ever SC generated in fiber, reported by Lin and Stolen in 1976 [1], was developed for ns TrA spectroscopy.

The development of the ns, broadband SC source reported here was motivated by research into the photochemistry and photophysics of solar energy harvesting complexes that exhibit cascades of dynamical processes. Understanding the photochemistry of such systems is critically important for realizing a future based on renewable energy [2]. One very promising chemical system that mimics biological photosynthesis involves supramolecular assemblies of porphyrin and fullerene derivatives [3–8]. In these self-assembled complexes, a photoexcited porphyrin acts as an electron donor to a fullerene. The transient intermediate states adopted by the complex subsequent to photoexcitation possess time constants that range from femtoseconds (fs) to micro- (μs), and UV/Vis/NIR absorption signatures from 400 nm to 1300 nm. A variety of spacers and ligands can be used to fine-tune the dynamics, with a major goal of increasing the lifetime of the final charge-separated state to the ms regime. A broadband ns SC source would, therefore, be very useful for probing the longer-lived of these states.

The pump-probe arrangement is an elegant and efficient tool for spectroscopically exploring photophysical and photochemical processes. On the fs timescale, nonlinear optical processes can provide stable broadband pulses that enable multi-wavelength probing. On ns to ms time scales, however, the probe step is accomplished either point-by-point, with a second laser, or spectrally resolved with a broadband flash lamp. The single wavelength approach can be quite tedious, at best; at worst, it can result in misleading data. Flash lamps allow broad spectral acquisition, but at the cost of time resolution. Currently, the fastest flash lamps have pulse durations on the scale of a few hundred ns, and they lack spatial coherence. One can gate the acquisition to gain temporal resolution, but this makes the system cumbersome and expensive. The most elegant solution is to have

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an electronically triggered broadband SC laser with pulses of a few ns in duration. A new PCF SC source was, therefore, constructed to efficiently and robustly measure lifetimes from about 2 ns to a few ms in the ~500–1300 nm wavelength range. The SC laser reported here represents a significant step forward by providing a source of 6 ± 0.5 ns, white-light probe pulses in this region of the electromagnetic spectrum.

The source we have constructed takes advantage of the relatively recent advent of SC generation using PCFs [9–13]. The laser pulses from this new system possess the highest spectral density per pulse (-15 dBm nm^{-1}) of any system reported in the literature, to the best of our knowledge. The system consists of a 15 m PCF with a zero-dispersion wavelength (ZDW) at 1040 nm that is pumped into the anomalous group velocity dispersion (GVD) regime with the 7 ns, 1064 nm output from a commercial Nd:YAG laser. Since our pump-probe spectroscopy must access time delays as long as a few ms, we have selected a laser source with a repetition rate of 20 Hz. The configuration was optimized for SC spectral width, power, uniformity and stability in the absence of optical damage. The best configuration generates SC from 450 to 1700 nm using a dual-pump system, with peak pump power of about 7.2 kW. With these properties, this SC source is very suitable for ns–ms TrA experiments for a wide range of photochemically and photophysically important molecular systems.

2. Experimental setup

A Q-switched Nd:YAG laser (1064 nm, 7 ns pulse duration, 20 Hz repetition rate, 420 mJ/pulse; Quanta-Ray INDI, Spectra-Physics) provided the pump pulse for the SC generation stage. As illustrated in Fig. 1, a glass wedge (Thorlabs WG11050) was used to split off approximately 10% of the beam and direct it to gold steering mirrors. A half-wave plate (Thorlabs WPQ05M-1064) and a polarizer (Thorlabs GL10-C) were employed as a variable attenuator, and the beam intensity was further reduced with a 0.9 OD ND filter, resulting in a pump power density of range 1.3×10^{-3} to 0.01 W cm^{-2}. A f=50 mm lens was used to couple the free-space 1064 nm beam into the fiber system. The SC was generated in a 15 m PCF (d_{core}=4.8 μm; Perfos HF148MIC; ZDW=1040 nm see dispersion characteristics in Fig. 2) [14].

In the most robust configuration, short lengths of SMF (Thorlabs SMF28) were spliced on both ends of the PCF to act as an optical buffer and prevent damage to the PCF. The splices resulted in additional losses of 0.4 dB. In the final system configuration, the Nd:YAG 1064 nm pulses were directly coupled into the bare SMF end. Any particle contamination resulted in failure at the bare fiber tip due to localized absorption and thermal breakdown. These occur even at input pulse powers far below the damage threshold of the SMF. A protective enclosure was, therefore,

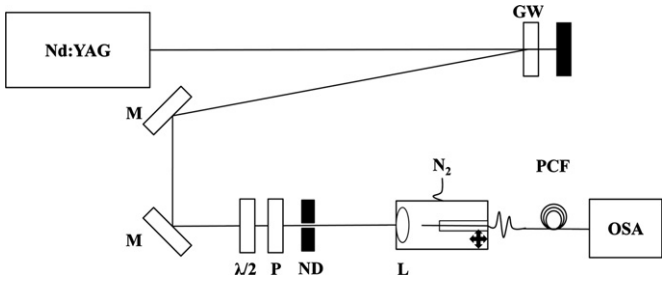

Fig. 1. Experimental setup: Nd:YAG laser (Spectra-Physics Indi) at 1064 nm, 20 Hz, 7 ns; GW, glass wedge at 10% reflection; M, mirrors; $\lambda/2$, half-wave-plate; P, polarizer; ND, neutral-density filter; L, 50 mm focusing lens; PCF, 15 m photonic crystal fiber.

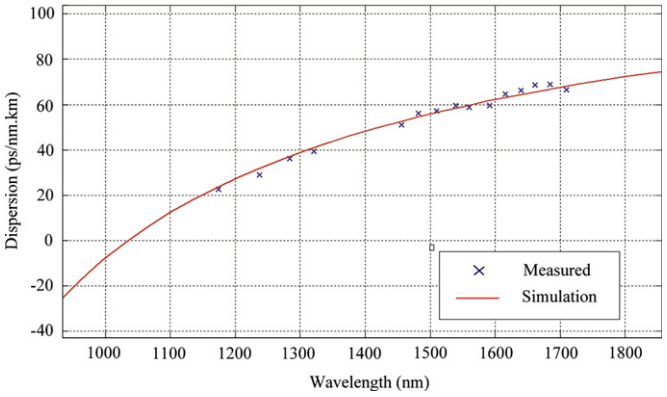

Fig. 2. (Color online) Dispersion profile of the 15 m Perfos HF148MIC PCF used to generate the SC.

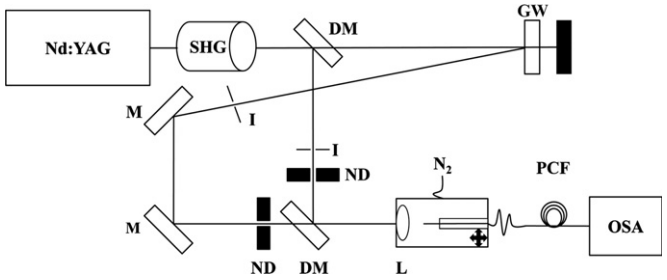

Fig. 3. Experimental setup for double-pumping: Nd:YAG laser (Spectra-Physics Indi); SHG, second harmonic generation crystal for 532 nm; DM, dichroic mirror reflects 532 nm; GW, glass wedge at 10% reflection; I, iris; ND, neutral-density filter; L, 50 mm focusing lens; N_2, nitrogen gas; PCF, 15 m photonic crystal fiber; OSA, optical spectrum analyzer (Andor AQ6315A).

constructed to isolate the injection stage. This housing was purged with N_2 gas before (30 min @ 7 L/min) and continuously during (3 L/min) experiments to eliminate dust particles at the fiber tip.

The alignment procedure began with the Nd:YAG laser in the 50 ns long-pulse mode. This reduced the pulse power and avoided fiber damage resulting from focusing on the edge of the fiber optic. The fiber tip was roughly aligned to approximately 50 mm away from the focusing lens, the N_2 gas purge described above was initiated, and the input coupling was then optimized to 30% efficiency. At this point, we can switch to short pulse (7 ns) mode without damaging the system. We believe we are at the limit of the optical energy that can be injected into the fiber given our pulse rate and duration, as failure to follow exactly the procedure outlined above resulted in fiber damage.

The spectral content of the SC was assessed using an ANDO-AQ6315A optical spectrum analyzer with a spectral resolution of 5 nm. Spectral stability was studied by recording measurements every 15 min over several hours. Pump and SC pulse durations were directly measured with a Si PIN photodiode and a Tektronix 2022B oscilloscope.

To study the effects of dual-wavelength pumping, a second configuration with simultaneous input at 1064 and 532 nm was also constructed (Fig. 3). The second harmonic of the Nd:YAG output was generated with a conversion efficiency of 30%. The double-wavelength beam was first directed onto a 532 nm dichroic mirror (Photonic Solutions, 105-0022) to transmit the 1064 nm beam and reflect the 532 nm onto a second dichroic mirror. The two pump beams were then spatially superimposed. A glass wedge was used to attenuate the 1064 nm beam, and irises and ND filters were employed to independently control the power of each wavelength coupled into the fiber.

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