# Generation of octave-spanning supercontinuum by Raman-assisted four-wave mixing in singlecrystal diamond

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**Abstract:** An octave-spanning coherent supercontinuum is generated by non-collinear Raman-assisted four-wave mixing in single-crystal diamond using 7.7 fs laser pulses that have been chirped to about 420 fs in duration. The use of ultrabroad bandwidth pulses as input results in substantial overlap of the generated spectrum of the anti-Stokes sidebands, creating a phase-locked supercontinuum when all the sidebands are combined to overlap in time and space. The overall bandwidth of the generated supercontinuum is sufficient to support its compression to isolated few-tosingle cycle attosecond transients. The significant spectral overlap of adjacent anti-Stokes sidebands allows the utilization of straight-forward spectral interferometry to test the relative phase coherence of the anti-Stokes outputs and is demonstrated here for two adjacent pairs of sidebands. The method can subsequently be employed to set the relative phase of the sidebands for pulse compression and for the synthesis of arbitrary field transients.

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## 1. Introduction

Optical transients in the single-cycle to sub-cycle regime are femtosecond to attosecond fields that are invaluable in elucidating ultrafast electronic processes in atoms, molecules and condensed matter and for realizing nonlinear optics in the sub-cycle regime. One prerequisite to the successful synthesis of these transient fields is to have a coherent light source that possesses an octave-spanning spectrum. Such a light source can be generated by focusing intense millijoule level femtosecond laser pulses into a gas or a solid medium to make use of laser-induced Kerr nonlinearity to broaden the pulse's spectrum [1]. Indeed with this approach attosecond transients have recently been synthesized successfully [2,3]. An alternative approach for generating a broad spectrum is the molecular modulation technique [4]. Using molecular gases driven in the steady state regime by narrowband nanosecond lasers, this technique has successfully produced frequency combs that consist of up to a couple hundred Stokes and anti-Stokes sidebands [5] and has been used to demonstrate the synthesis of a periodic train of ultrashort optical fields of arbitrary shape in the visible-UV spectral region, where such pulses inevitably express single-cycle nature, and allow non-sinusoidal field synthesis [6].

Transient or impulsive Raman generation, and phase-matched cascaded four-wave mixing (CFWM) using two-color femtosecond (fs) pulses or a pair of time-delayed chirped pulses has also produced broadband spectra constituting multicolor Stokes and anti-Stokes sidebands that span up to an octave in bulk crystals or in the gas phase [7–20]. When the difference in frequency of the two-color pulses or of the chirped pulses is near a Raman-type molecular resonance, contribution of the molecular resonance that boosts the efficiency of the generation process is observed [9,17]. The short pulse envelope of the fs pulses has led to the synthesis of pulse trains consisting of just a few pulses or a nearly isolated single pulse [21,22]. For the next step, in order to produce isolated ultrashort pulses as desired by many applications, it is necessary to have an octave-spanning continuous spectrum with minimal periodic structural peaks or a supercontinuum.

An octave-spanning supercontinuum may be obtained by replicating the spectrum of a driving pulse whose frequency bandwidth is broader than the frequency spacing between adjacent CFWM generated sidebands and then combining the sidebands into one pulse. We report here generation of an octave-spanning supercontinuum in single-crystal diamond using a pair of chirped pulses whose transform-limited duration is 7.7 fs by Raman-assisted CFWM. Diamond is chosen because of its large Raman transition frequency of 1332 cm<sup>-1</sup> and its superior optical properties including high optical damage threshold and large optical bandgap. Previous published reports have already shown that diamond is an effective medium for CFWM [14,23]. We have extended its utility to the generation of a phase-locked supercontinuum. In addition, the substantial spectral overlap of adjacent anti-Stokes sidebands allows the utilization of straight-forward spectral interferometry to determine the relative phase coherence of the anti-Stokes outputs. We demonstrate this by using multi-wavelength interferometric measurements to confirm phase-locking among the spectral components of the sidebands.

## 2. Experimental

We begin with 23 fs long pulses centered at  $\lambda = 790$  nm from a commercial 1 kHz chirp-pulse amplified mode-locked Ti:sapphire laser (Femtopower HE PRO CEP). We expand the spectrum of these pulses by self-phase modulation in helium inside a hollow waveguide (inner core diameter 250 µm) to obtain wavelength coverage from 690 nm to 870 nm at the 50% intensity level (a width of 3000 cm<sup>-1</sup>). A spectrum of this pulse is shown in Fig. 1. The broadened pulse is first compressed with negatively-chirped mirrors to a near-transformlimited duration of 7.7 fs. The main portion of this pulse is used for a high harmonic generation experiment. We used about 2% of the compressed laser power for the purpose of the experiments described here. The 2% value was chosen because we found that this value is sufficient for demonstrating the idea being put forward in this paper while leaving sufficient laser power for other experiments. The 2% pulse was chirped to about 420 fs FWHM (chirp rate 7400 fs<sup>2</sup>) by passing the pulse through a 4 mm thick SF56A glass. For CFWM in condensed phase it is prudent to lengthen the pulse to avoid self-phase modulation and optical



Fig. 1. Schematic of the experimental setup. BS1: 2% beamsplitter; BS2: 50/50 beam-splitter; CP: compensation plate; Stretcher: 4 mm thick SF56A glass. The focusing mirror has a radius of curvature of 40 cm. About 2% of the compressed output from the hollow fiber is stretched and split into two and then recombined at the diamond crystal with a relative time delay between the two pulses provided by the delay stage. A spectrum of the pulse incident onto the crystal is shown in the upper left quadrant.

damage in the medium [18]. We determined experimentally that the present chirp rate is optimal for the pulse power used in this experiment. A smaller chirp would cause optical damage in our set-up and with a larger chirp the generated bandwidth and overall efficiency would have suffered.

Each chirped pulse was then split into two pulses by a broadband ~50% beamsplitter coated for minimal second-order dispersion. One of the split pulses served as the pump pulse and traveled on a path that can be delayed (see Fig. 1). The other pulse (signal) passed through a chirp compensator to equalize the chirp acquired by each split pulse after their separation. The split pulses were then focused and recombined to cross each other at an angle over a spot of diameter 40  $\mu$ m in a 0.5 mm thick CVD-grown single-crystal diamond. The polarization of the pulses is parallel to the [110] direction of the crystal. After attenuation and accounting for losses in the beam paths, the average power of the chirped beams incident onto the diamond crystal was 3.3 mW (pump) and 3.0 mW (signal) respectively. The corresponding incident intensities were  $6.25 \times 10^{11}$  W/cm<sup>2</sup> and  $5.7 \times 10^{11}$  W/cm<sup>2</sup>.

A time delay between the two chirped pulses creates an instantaneous frequency difference between them when they cross each other in the crystal [13]. The pulses interact by CFWM to produce sidebands that exited from the diamond crystal in a range of angles away from the two crossing incident pulses as depicted in Fig. 1. The intensity and spectrum of each generated sideband were recorded and analysed with an optical spectrometer equipped with a calibrated diode array detector (Ocean Optics HR4000).

#### 3. Results and discussion

We did experiments at several external crossing angles between the two input pulses from  $\sim 2^{\circ}$  to  $\sim 5^{\circ}$ , recording the anti-Stokes sidebands as we scanned the relative delay time between the two pulses. We found that, for all incident angles, intense CFWM output consisting of several Stokes and anti-Stokes sidebands occurs only for a span of time delay that is short compared to the chirped pulse duration of 420 fs (see Fig. 2). In CFWM the generation of each sideband is determined by off-angle phase-matching of the frequencies involved [8,15]. This result means that for chirped pulses there is only a limited span of time delay for which the instantaneous frequencies of the pulses can satisfy the phase-matching condition.

We further found that the 3° crossing angle produces the largest number of Stokes and anti-Stokes sidebands. At this angle, more than three Stokes and ten anti-Stokes sidebands were generated. Due to a smaller dispersion on the red side, the first two Stokes beams exiting the diamond crystal spatially overlapped in part with the pump. Recording the spectrum of the Stokes spectra inevitably included a portion of the more intense pump spectrum. Analysis of the Stokes sidebands becomes quite ambiguous. Therefore, only the anti-Stokes sidebands have been studied in greater detail and reported below.

In our experiment the largest number of anti-Stokes sidebands produced occurred at a relative time delay of  $220 \pm 10$  fs. A mirror image of the angular distribution of the sidebands appeared at negative time delay of the same value due to symmetry of the set up. This provided an excellent way to determine the zero time with an accuracy of a few fs for the experiment. Figure 2 shows the intensity of the first 5 anti-Stokes sidebands generated as a function of relative time delay between the two incident pulses at the 3° crossing angle. As can be seen, the third and higher anti-Stokes orders appear only within a relative time delay window of 60 fs (FWHM) centered at about 220 fs. The instantaneous frequency difference corresponding to the time delay of 220 fs is  $1221 \pm 200$  cm<sup>-1</sup>. This value is within experimental uncertainty equal to the 1332 cm<sup>-1</sup> Raman frequency of diamond. The proximity of the two values is not accidental. Evidently successful generation of many sidebands needs good conversion efficiency so that it is a prerequisite that both the phase-matching condition and near-resonance Raman enhancement are satisfied in the CFWM process.

In our experiment the spectral width of the input is substantially wider than the Raman spacing of the diamond crystal. From the Sellmeier equation of diamond [24] we calculated that the CFWM phase-matching acceptance bandwidth for the 0.5 mm long crystal is sufficiently broad so that a good portion of the entire input spectrum can be sequentially upconverted to the next several anti-Stokes sidebands. The spectrum of each sideband would therefore be broad and would overlap with the adjacent sideband spectrum to produce a coherent supercontinuum. We recorded the spectrum of every anti-Stokes sideband. They are jointly plotted into a combined spectrum as shown in Fig. 3.



Fig. 2. Anti-Stokes output generated as a function of the relative delay between the two input pulses for a 3° crossing angle between the two input pulses. ASn stands for n<sup>th</sup> anti-Stokes sideband. From top to bottom: AS1, AS2, AS3, AS4 and AS5.



Fig. 3. Combined spectrum of the input and generated anti-Stokes sidebands, showing a continuous span in excess of  $13000 \text{ cm}^{-1}$ .

The figure confirms that the broad input spectrum can be replicated to most of the sidebands. The structures seen in Fig. 3 originate mainly from those present in the input spectrum that is shown in Fig. 1. The sidebands do gradually get narrower at higher anti-Stokes orders when increasing dispersion towards the blue side of the spectrum naturally

tightens the phase-matching bandwidth. A distinctive feature in Fig. 3 is that there is significant overlap in the spectra of all the adjacent sidebands as was initially conjectured. Every frequency is present in at least two different CFWM sidebands. The pulse energy of sidebands higher than the tenth falls off rapidly as dispersion causes CFWM to become far from being phase-matched.

The frequency span of the combined sidebands is larger than 13000 cm<sup>-1</sup>, spanning more than one octave. This spectral width is sufficient for the Fourier synthesis of single isolated few-cycle to single-cycle transient fields. Matter of fact this generated total bandwidth is similar to previously reported CFWM work cited above. However the difference is that in the present case there is better spectral continuity and coverage with few periodic structural peaks. Such discrete structures are inevitable when inputs narrower than the Raman spacing were used as pump. Suppression of these structural peaks due to the improved continuity in spectral coverage that is achieved in this experiment will help to realize pedestal-free femtosecond transient fields.

The sidebands generated by CFWM are expected to be mutually coherent. Since the sidebands overlap there should be optical interference when heterodyning pairs of adjacent sidebands. We set up a Michelson interferometer to demonstrate this effect. The beam of the first anti-Stokes sideband (AS1) pulses was aligned collinearly with that of the second anti-Stokes sideband (AS2). The combined beam was fiber-coupled to the spectrometer-diode array detector system. The resulting spectrum was recorded while the AS1 pulse was delay-scanned with respect to the AS2 pulse. The result is a two-dimensional multi-wavelength interferogram that simultaneously records the interference of many wavelengths as a function of time delay between the two anti-Stokes sideband pulses as shown in Fig. 4. The blue curve in Fig. 4(a) is spectrum recorded when AS1 and AS2 pulses were temporally separated. This spectrum is simply the sum of the individual spectra of the AS1 and AS2 pulses. Figure 4(a) also shows that the spectra of AS1 and of AS2 overlap for as much as 90 nm, from ~620 nm to ~710 nm.

As the two pulses began to overlap in time, interference in the form of a beat signal across the overlapping spectral region of AS1 and AS2 can clearly be discerned. The portion from 670 nm to 685 nm where the largest modulation depths were produced is shown in Fig. 4(b). A portion of Fig. 4(b) is expanded for display in Fig. 4(c) to show clear oscillations at every resolvable wavelength. The slanted pattern in Figs. 4(b) and 4(c) is an expected consequence of different phase-coherent wavelengths propagating simultaneously in space. The interferograms shown in Fig. 4 took several minutes to complete. The clarity of the interference patterns indicates that the phase locking between the two pulses is extremely stable.

We repeated the measurements for the third (AS3) and fourth (AS4) sidebands and obtained similar results (Figs. 4(d)-4(f)). By induction the result can be used to interpret that phases are locked simultaneously at all wavelengths across the entire supercontinuum. While phase coherence is fully expected from a CFWM process, this is the first time that the coherence and its stability are displayed by experiment over a broad spectral region simultaneously. From the interferogram, relative spectral phase across many wavelengths can be determined. Thus, these measurements demonstrate that for sidebands generated by broadbandwidth CFWM it is sufficient to use simple spectral interferometry to determine the relative phases without having to resort to nonlinear optical processes that are a lot less efficient. This technique can eventually be used to set or correct the relative phase of all the anti-Stokes sidebands by inclusion of phase adjustment devices such as spatial light modulators [25] or chirp mirrors to facilitate compression to an ultrafast field transient.

The measured average power of the output were 344  $\mu$ W, 68  $\mu$ W, 20  $\mu$ W, 6  $\mu$ W, 4.8  $\mu$ W, and 3.6  $\mu$ W for the first to the sixth anti-Stokes sideband respectively. This is >13% power conversion from the pump pulse to the anti-Stokes sidebands. The power of higher anti-Stokes sidebands were less than 1 $\mu$ W and are not listed here. These sideband powers are scalable with the input power.



Fig. 4. Result of phase coherence tests of the generated sidebands: (a) Spectrum of AS1 and AS2 recorded separately (red: AS1; green: AS2) and combined with the two temporally separated (blue); (b) Spectrally-resolved intensity pattern of combined AS1 and AS2 from 670 nm to 685 nm as a function of relative time delay between the two sidebands. Relative delay time of zero is when the phases are aligned at all wavelengths; (c) portion of (b) expanded in time to show the result of optical interference. (d) to (f) are repeat of (a) to (e) for AS3 and AS4 with similar result.

By the nature of off-angle phase-matched four-wave mixing, the generated sidebands exit the diamond crystal at slightly different angles and are spatially chirped in the phasematching plane. These effects have to be compensated when it comes time to synthesize ultrashort pulses with these sidebands. One solution is to bring the sidebands together in a focused region with a pair of parabolic mirrors similar to the arrangement exercised in Ref. [21] and Ref. [25] or use MEMS mirrors to perform the compensation. An alternative is to substitute off-angle mixing with collinear four-wave mixing. This however is an inferior solution due to a lower efficiency and some reduction in bandwidth of the continuum.

## 4. Summary

In summary, we have advanced the technique of CFWM to efficiently produce an octavespanning supercontinuum with input pulses whose bandwidth is wide compared to the Raman mode of the generating medium and using single-crystal diamond as the generating medium. We demonstrated that optical interferometry is effective for simultaneous multi-wavelength measurement of the relative phases of the generated output. With further development the interferometric technique could be used for simultaneous adjustment of the phases. A logical next step would be to spatially and temporally overlap all the sidebands to realize transient field synthesis with the generated supercontinuum. When perfectly compressed this supercontinuum will provide a single-cycle transient field with minimal side-lobes and an envelope of 2.6 fs. It will also be of interest to perform more systematic studies to quantify the contribution of the Raman effect in this broadband chirped-pulse CFWM process.

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