Raman-assisted octave-spanning continuum generation in single-crystal diamond for sub-cycle pulse synthesis

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Abstract: A coherent continuum is generated by Raman-assisted four-wave mixing in single-crystal diamond of an 8 fs laser pulse. The bandwidth of the spectrum will support the synthesis of an isolated single-cycle femtosecond pulse.

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1. Introduction

Subfemtosecond to attosecond pulses are highly desirable for elucidating ultrafast processes in atoms, molecules and condensed matter. One particularly promising approach to generating such pulses is the molecular modulation technique which has the potential for synthesizing optical pulses as short as a fraction of a fs [1]. This Raman technique, originally proposed for molecular gases, has been demonstrated to be suitable for efficient production of ultrashort pulses in the near-visible spectral region, where such pulses inevitably express single-cycle nature and allow non-sinusoidal field synthesis [2].

When narrowband nanosecond driving fields were used in gas phase molecular modulation, long trains of ~10^6 relatively low-energy (nanojoule level) subcycle pulses are generated. To increase the pulse energy, the molecular modulation method has been extended to Raman-active crystals pumped with mode-locked laser pulses, leading to a possibility of generating a few ultrashort pulses per pulse train [3]. In particular, by selectively exciting the Raman transitions in a lead tungstate crystal using a pair of time-delayed linearly chirped pulses, strong molecular coherence has led to the generation of up to 40 discrete anti-Stokes and 5 Stokes sidebands assisted by phase-matching and favorable near-resonance to the 191 cm^{-1} Raman mode of the crystal [4].

In order to generate isolated ultrashort pulses that could facilitate studies of ultrafast dynamics that employ the pump-and-probe technique, a continuous spectrum is desired [1]. The large Raman mode spacing of single-crystalline diamond at 1332 cm^{-1} and diamond's extraordinary properties such as high Raman gain, wide band gap of 5.8 eV, and high damage threshold make it an excellent candidate for molecular modulation-assisted generation of ultrashort pulses. Using 30-50 fs pulses chirped to about 1 ps, a discrete 5-6 member frequency comb with comb spacing near the 1332 cm^{-1} Raman mode has been demonstrated [4,5]. A continuous and broad spectrum could be generated in diamond by starting with a pulse with a spectrum wider than the Raman mode of 1332 cm^{-1}. In this paper, we report successful generation of an octave-spanning coherent continuum in single-crystal diamond with a chirped 7.7 fs pulse. This is a key step toward the synthesis of isolated ultrafast pulses in the visible-uv region.

2. Experimental Setup

We began with intense few-cycle laser pulses from a commercial chirp-pulse-amplified Ti-Sapphire laser system (Femtolaser). The output from the system is an infrared beam of pulses centered at λ = 790 nm of 23 fs pulse duration with 1 kHz repetition rate and ~2 W average power. The pulse spectrum was broadened by self-phase modulation in He inside a hollow waveguide and the pulses were compressed to a near-transform-limited duration of 7.7 fs by a set of chirp mirror. After that the pulses were positively-chirped by passing through a 4 mm thick piece of glass to about 370 fs FWHM. Each chirped pulse was then split in a pseudo-Michelson interferometer setting into two pulses of nearly equal intensity by a broadband beamsplitter designed for use at low second order dispersion and later recombined after one pulse is delayed by the translation of a retro-reflector located in one arm of the interferometer. The combined pulses were then focused and crossed each other at an angle of 3^o in a 2mm x 4mm by 0.5 mm thick CVD diamond. The average power of the chirped beams incident onto the diamond crystal were 2.68 mW and 1.44 mW respectively.

3. Experimental results and discussion

When the time delay between the pulse pair is changed, at some time delay the instantaneous frequency difference would equal the Raman frequency in diamond so that the Raman mode of the crystal is driven to enhance four-wave frequency mixing in the crystal. The optimal time delay is a function of the crossing angle between the two pulses. We find that the best results were for a crossing angle of 3^{o} between the two beams. At this angle, the optimal time delay was 232 fs. At this time delay, when off-angle phase-matching condition is satisfied at least 10 anti-Stokes [AS] beams are generated. The total conversion efficiency from the pump beams to the sidebands is 14 %. The average power of the output obtained is limited by optical damage of the diamond crystal which is estimated at 10^{12} W/cm^2. The average power can be scaled to a higher value by employing a looser focus at the crystal. Figure 1 is a picture of the image of the output beams taken on a screen placed 20 cm from the diamond crystal.

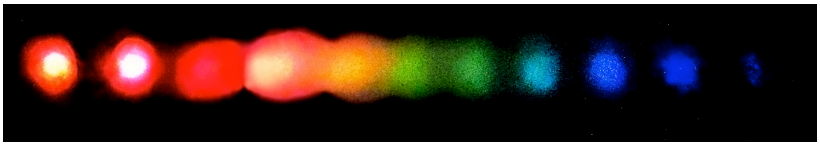

Fig 1. Raman assisted four-wave mixing from single crystal diamond. The two spots on the left are the inputs. Anti-Stokes orders from 1 to 9 are shown.

In our experiment the width of the input spectrum is larger than the Raman spacing of the diamond crystal. The spectrum of sidebands are therefore broad and overlaps with the adjacent sideband spectrum to produce a coherent continuum spectrum. As shown in figure 2, the combined frequency span of the anti-Stokes sidebands is > 15000 cm^{-1} which is more than one octave. Even wider frequency spacing and hence a broader overall spectrum are expected by operating at a larger crossing angle. Interferometric cross-correlation of adjacent orders confirm relative coherence of the anti-Stokes orders. The results suggest that isolated sub-cycle pulses can be synthesized with the generated octave-spanning pulses.

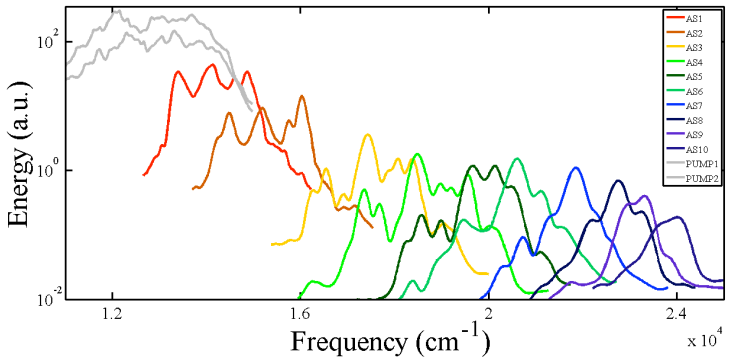

Fig 2. Spectrum of the generated anti Stokes four-wave mixing orders, showing a continuum span of more than 15000 cm^{-1}.

4. Conclusion

We have realized Raman-assisted octave-spanning continuum generation in CVD single crystal diamond by using chirped few-cycle pulses. This will lead to the generation of isolated sub-femtosecond pulses.

5. References

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