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Extended Abstract

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Ultrabroad band mid-IR frequency combs for sensitive molecular detection: frequency-divide-and-conquer approach

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I will present a new technique for extending frequency combs to the highly desirable mid-IR ?molecular fingerprint? spectral range. The technique is based on subharmonic optical parametric oscillation (OPO) in quasi-phase-matched GaAs crystal. This process can be considered as a reverse of the second harmonic generation: the frequency comb of a pump laser (Tm-fiber 2-?m laser or Cr:ZnS 2.4-?m laser) is transposed to half of its central frequency and simultaneously spectrally augmented to more than an octave, thanks to the enormous gain bandwidth near OPO degeneracy and cross-coupling between the laser and the OPO frequency comb lines. With this source we demonstrate massively-parallel trace detection of molecular mixtures and achieve part-per-billion sensitivity levels. Our present focus is both biomedical and security applications of mid-IR spectral combs.

Some 20 years ago, optical frequency combs emerged as a radically new laser technology that culminated in the 2005 Nobel Prize in Physics. Frequency combs, which can be generated in phase-stabilized mode-locked lasers, possess unique properties in both the spectral and time domains. In the spectral domain, they correspond to a manifold of several hundred thousand equally spaced narrow spectral 'comb' lines. In time, they relate to a periodic train of pulses with an extremely stable repetition rate and carrier-envelope phase. These combs hold much promise for high-resolution spectroscopy and spectroscopic detection. A laser comb can directly interrogate a vast number of molecular transitions, resulting in an immense parallelism of spectral measurements, thereby enabling quick measurements of a variety of chemicals. Frequency combs combined with Fourier transform spectroscopy (FTS) are especially advantageous because one can retain massive parallelism of spectral measurements, as well as high accuracy and sensitivity, using just one optical detector. FTS can be performed using a single comb source combined with a Michelson interferometer, or two mutually phase-locked frequency combs with a small offset in their pulse repetition rate. In the latter case (so-called dual-comb spectroscopy), an enormous advantage in data acquisition speed may be achieved due to the absence of moving parts.
We have implemented a new technique that enables the use of octave-wide frequency combs to be extended into the highly desirable, yet challenging, mid-IR fingerprint spectral range, leading to the development of ultrabroadband mid-IR combs. We used a short (few-100μm) second-order nonlinear crystal as the gain element in a synchronously pumped optical parametric oscillator (OPO) that operates very close to its degeneracy, where gain bandwidth is very broad and hence enables a very broad instantaneous spectrum to be obtained in the output. The role of the OPO is to rigorously both down-convert and augment the spectrum of a pump frequency comb, such that the laser comb spectrum is transferred to half of its central frequency and dramatically broadened.

To prove the viability of this technique, we used a variety of OPO gain materials in combination with ultrafast pump lasers. Broadband (>1000nm) instantaneous outputs were reported in three setups: an erbium-fiber laser-pumped periodically poled lithium niobate subharmonic OPO with a center frequency near 3μm, a thulium-fiber-laser-pumped orientation-patterned gallium arsenide (OP-GaAs) OPO with center frequency ~4μm and a chromium-doped zinc-selenide-laser-pumped OP-GaAs OPO with center frequency ~5μm. In addition to the low pump threshold (<10mW) associated with their doubly resonant nature, these OPOs have the important property of preserving pump laser coherence. The latter characteristic is extremely important for performing dual-comb spectroscopy.

Our new mid-IR comb technology could have a major impact in a wide range of applications based on real-time spectroscopic molecular detection. These span from trace explosive and chemical/biological hazard detection to many industrial and commercial applications in areas including environmental monitoring, medicine, planetary exploration, and analytical chemistry. In future work, we hope to develop a frequency comb with an instantaneous bandwidth of two octaves (2.5–10μm) by using special 'chirped' dielectric mirrors in the OPO resonator, enabling compensation of group velocity variation over the whole spectrum. We also intend to build a 'twin' OPO system, based on a pair of phase-locked fiber lasers, for dual-comb spectroscopy. This system will provide an enormous advantage in terms of the sensitivity and data acquisition speed of standoff chemical sensing.

Bottom Note: *This work is partly presented at International Conference and Trade fair on Laser Technology, July 20-22, 2015, Orlando, Florida, USA*