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Review

Femtosecond optical parametric oscillator frequency combs

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Abstract

Techniques to measure and manipulate the carrier-envelope phase within femtosecond optical parametric oscillators (OPOs) allow their outputs to be stabilized in a way that produces a frequency comb structure, potentially tunable throughout the transparency band of the gain material. In this review we describe the fundamental principles of phase control, on which the development of singly- and doubly-resonant OPO frequency combs is based. We give examples of practical embodiments of such combs, and discuss in detail several applications, including spectroscopy, metrology, quantum computation and astrophotonics.

Keywords: frequency combs, optical parametric oscillators, femtosecond lasers

1. Introduction

Second-order nonlinear frequency conversion offers a means of extending frequency comb technology to wavelength regions not addressable directly by common femtosecond laser comb sources such as Ti:sapphire [1], Yb:fibre [2], Er:fibre [3], Tm:fibre [4], Cr:ZnS [5], or Cr:ZnSe [6] lasers. Single-pass difference frequency generation (DFG) with femtosecond pulses makes it possible to produce combs having a stable carrier-envelope phase (CEP) [7], even allowing high conversion efficiency to be achieved when using amplifiers in both pumping channels [8]. Constructing a resonator around such a DFG crystal so that oscillation is established from parametric fluorescence with only a single pump laser and by synchronous feedback of the signal and / or idler pulses was first demonstrated in the femtosecond regime in 1989 [9], laying the groundwork for the synchronously pumped optical parametric oscillator (OPO) systems described in this article.

Femtosecond OPOs are natural systems for generating frequency combs, whose applications tend to emphasize the importance of bandwidth and wavelength coverage, for example their use in optical metrology [10], spectroscopy [11], optical clockworks [12] or for comparisons between different time and frequency standards [13]. Unlike laser media, the wavelengths generated in a parametric frequency conversion process are not limited by a prescribed set of electronic energy levels, allowing OPOs to be both widely tunable and broadband, only limited by the absorption characteristics of the nonlinear gain material.

The parametric gain process is also intrinsically quiet. While in a femtosecond laser, contributions from spontaneous emission can increase the noise power spectral density (PSD) [14, 15], in an OPO the gain process is instantaneous and the contribution from parametric fluorescence is typically negligible. Furthermore, the synchronously pumped nature of femtosecond OPOs means that they do not need to utilize significant levels of Kerr nonlinearity in order to support

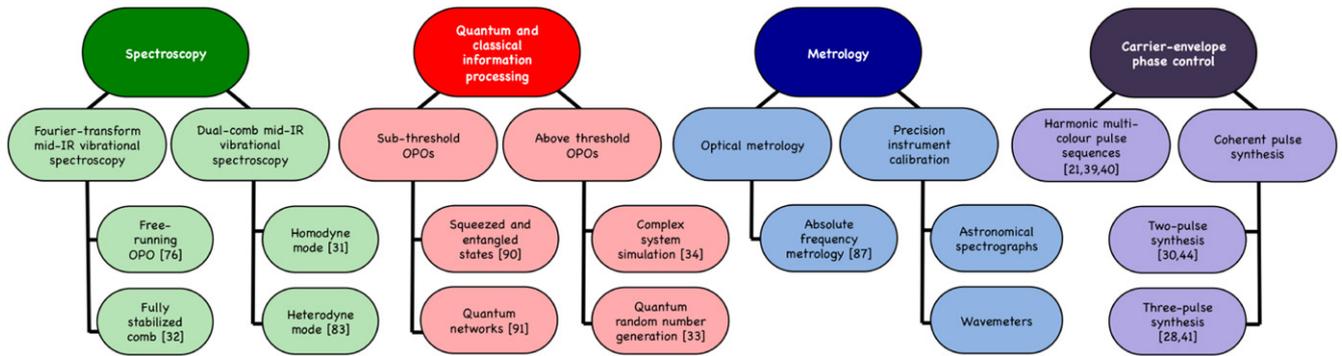


Figure 1. A perspective on femtosecond OPO frequency combs, classifying their main application areas.

modelocking, in contrast to many passively modelocked lasers. Femtosecond OPOs typically employ millimetre-length nonlinear crystals, which are orders of magnitude shorter than the gain media used in fibre frequency combs operating at similar near-infrared wavelengths. This means that the coupling of intensity fluctuations into phase noise within an OPO comb can be much lower than in fibre combs. When a high quality pump laser is used, this feature allows OPO frequency combs to be stabilized to operate with carrier-envelope-offset (CEO) frequency beat linewidths of a few Hz [16], rather than the hundreds of kHz associated with competitive laser combs operating in the same spectral region such as Er:fibre [3] and Cr:forsterite [17], although we note that with suitable techniques the offset-frequency linewidth in Er:fibre combs can be reduced to Hz levels [18]. As well as the differences in the intrinsic noise processes between OPOs and lasers, this difference in performance can also be attributed substantially to the presence of phase noise at the extremes of the super-continuum used for detecting the comb-offset beat frequency with the f -to- $2f$ technique [19], which may be lower in an OPO comb which can use more central super-continuum wavelengths for locking, depending on the specific wavelengths of interest [20].

Optical parametric down-conversion is further characterized by the mutual coherence between the interacting fields, studies of which paved the way to the original demonstration of femtosecond OPO frequency combs in the form of relative phase measurements between OPO harmonic outputs [21] and the energy conservation in the parametric process [22]. This phase coherence now plays an important role in the techniques used to stabilize the offsets of OPO frequency combs [23] and in the interpretation and design of degenerate OPO frequency combs [24].

Since their first demonstration in 2007 [20], femtosecond OPO combs have been subject to increasing development. Notable results include the demonstration of average powers exceeding 1 W [25], zero-offset combs offering phase coherence among all the parametrically related fields [26], GHz operation [27], tunability into the visible [28] and mid-infrared [25], pulses as short as five cycles [29], Hz-level offset-frequency linewidths [16] and as low as 30 attosecond timing jitter between pulses in the same parametrically related comb [30]. Figure 1 presents an overview of the scientific areas already impacted by femtosecond OPO frequency

combs. In some of these, the OPO comb offers a technical advantage because alternative laser technology does not yet exist (e.g. in mid-IR vibrational spectroscopy, for which solid-state femtosecond laser sources operating at wavelengths $>2.5\ \mu\text{m}$ do not yet exist). In other areas the femtosecond OPO comb is fundamentally unique, for example in quantum optical applications which exploit the properties of parametric entanglement.

Before going further it is necessary to define what we mean by a ‘frequency comb’. Formally, a frequency comb is an ensemble of equally spaced narrow frequencies, whose relative phases are constant and whose absolute positions in frequency are fixed. By ‘narrow’ we mean that the linewidth of an individual frequency is much narrower than the spacing between adjacent frequencies. A modelocked laser naturally satisfies the requirement for fixed relative phases and equal frequency spacing, but freezing the absolute positions of these frequencies is only achieved in practice with the additional step of CEO frequency stabilization, often requiring considerable additional technical effort. While not every example we describe here demands full CEO stabilization, the majority require the offset frequency to have sufficient passive stability for the comb to be static in frequency during the lifetime of the measurement, for example in dual-comb OPO spectroscopy. Others (e.g. coherent synthesis) require that the relative offsets of different combs are controlled.

The remainder of this article develops the core concepts underpinning femtosecond OPO combs, starting from an understanding of the fundamental phase relationships between the pump, signal and idler pulses in a synchronously pumped femtosecond OPO. We highlight the recent achievements in this research area, which can now be divided into singly-resonant and doubly-resonant OPOs (DROs), which are each characterized by a distinct set of performance features and technical approaches. These separate comb configurations are explained in detail, allowing their relative performance to be critically compared. As the technology matures, the uses of femtosecond OPO combs are becoming increasingly diverse and ambitious. Applications employing some or all of the phase-stability characteristics of femtosecond OPOs are discussed in detail and include dual-comb spectroscopy [31], Fourier-transform spectroscopy [32], photonic random number generation [33], coherent pulse synthesis [30], quantum simulation [34] and even the generation of entangled states [35].

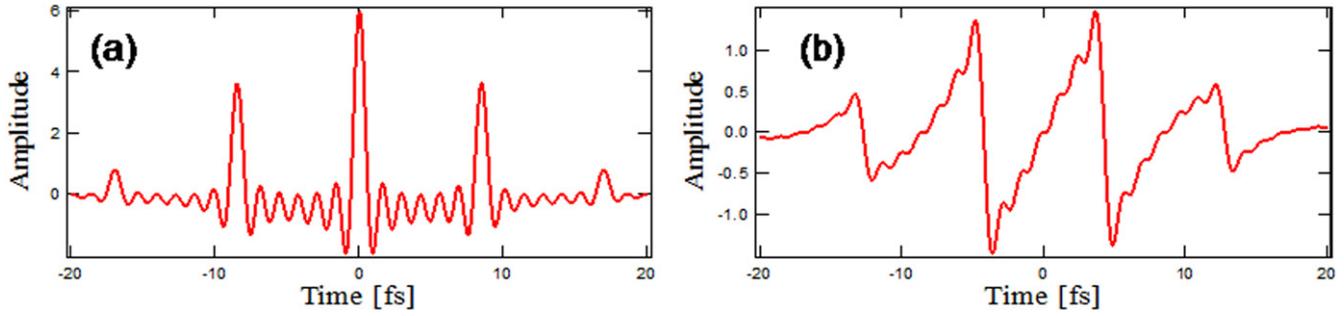


Figure 2. Example illustrating the electric field synthesis of six harmonics pulses to form an attosecond pulse train (a) and a saw-tooth wave (b) with wavelengths of 2550, 1275, 850, 638, 510, and 425 nm.

2. CEP control in femtosecond OPO combs

In an optical parametric process, where a pump beam produces a signal and an idler beam, for both the continuous and pulsed modes of operation the optical phase relationship between all three waves is given (modulo 2π) by

$$\theta_p = \theta_s + \theta_i + \frac{\pi}{2}, \quad (1)$$

where θ_p , θ_s , and θ_i are the phases of the pump, signal, and idler, respectively. In a doubly-resonant OPO it is well known that, as a result of the above-mentioned phase relationship, the oscillation is sensitive to the cavity length [36]. When we consider the phase relationship in a singly-resonant OPO (SRO) pumped by a mode-locked pulse sequence, the pulse repetition frequencies of both the pump and signal need to be the same. Here, the optical phase of the signal is determined from the cavity length and the intracavity dispersion, and that of the idler by the phase relation in equation (1). This assumption was experimentally verified in 2000 [21]. The optical phase of a femtosecond OPO can also refer to the CEP, which is defined as the phase difference of the carrier wave relative to the pulse envelope.

In the frequency domain, a femtosecond pulse sequence is represented by a series of equally spaced spectral lines and forms a frequency comb. The position of each comb tooth exhibits a CEO frequency which corresponds to the CEP slip of successive pulses. The optical frequencies of individual comb teeth in the pump, f_p , signal, f_s , and idler, f_i , beams can be expressed as

$$f_p = f_{0p} + lf_{\text{rep}}, \quad (2a)$$

$$f_s = f_{0s} + mf_{\text{rep}}, \quad (2b)$$

$$f_i = f_{0i} + nf_{\text{rep}}, \quad (2c)$$

where l, m, n are integers, f_{rep} is the pulse repetition frequency and f_{0p} , f_{0s} , and f_{0i} are the CEO frequencies of the pump, signal and idler beams, respectively. Since the three optical fields also satisfy the simple equation of energy conservation, $f_p = f_s + f_i$, the CEO frequency relation can be expressed as

$$f_{0p} = f_{0s} + f_{0i}. \quad (3)$$

For a given wave, j , the CEO frequency and the CEP slip of successive pulses ($\Delta\theta_j$) are tied together by a simple

relation

$$f_{0j} = \frac{\Delta\theta_j}{2\pi} f_{\text{rep}}. \quad (4)$$

From equation (1), the CEP slip relation between the pump, the signal, and the idler then follows

$$\Delta\theta_p = \Delta\theta_s + \Delta\theta_i. \quad (5)$$

Thus, controlling the relative CEO frequencies in equation (3) is equivalent to controlling the relative CEP slips of equation (5). However, as three fields are involved in the generation of a frequency comb in an OPO, to establish a fixed CEP frequency relationship between all three requires the absolute phase difference between them to be controlled.

By controlling the relative phase relation it is possible to realize numerous applications in various research areas. One promising application is the coherent synthesis of femtosecond pulses as originally envisioned by Hänsch and Shimoda [37, 38]. The artificial coherent addition of different coloured pulses enables arbitrary waveform generation to be achieved to produce, for example, attosecond pulse trains or saw-tooth waves, which could be applied to the coherent control of chemical reactions. An illustration of coherent synthesis is shown in figure 2, which depicts theoretical results achieved by combining six harmonic pulses to generate specific target waveforms.

For coherent synthesis, it can be beneficial to set the optical frequency ratio between the pump, the signal, and the idler to 3:2:1. Such a sub-harmonic OPO is not only relevant to applications such as arbitrary waveform generation, but also facilitates the direct observation and the control of the optical phase relation. Figure 3 illustrates how the relative CEP slip in the sub-harmonic femtosecond OPO can be measured and controlled [39].

Here, we consider a sub-harmonic OPO pumped by a Ti:sapphire laser at a centre wavelength of 850 nm. The pump, the signal, and the idler frequencies are defined as 3ω , 2ω , and ω , respectively. The generation of these harmonics is readily available by exploiting the additional frequency conversion processes that occur when using a periodically-poled lithium niobate (PPLN) crystal as the nonlinear medium in the OPO. Interactions between the pump, signal and idler pulses

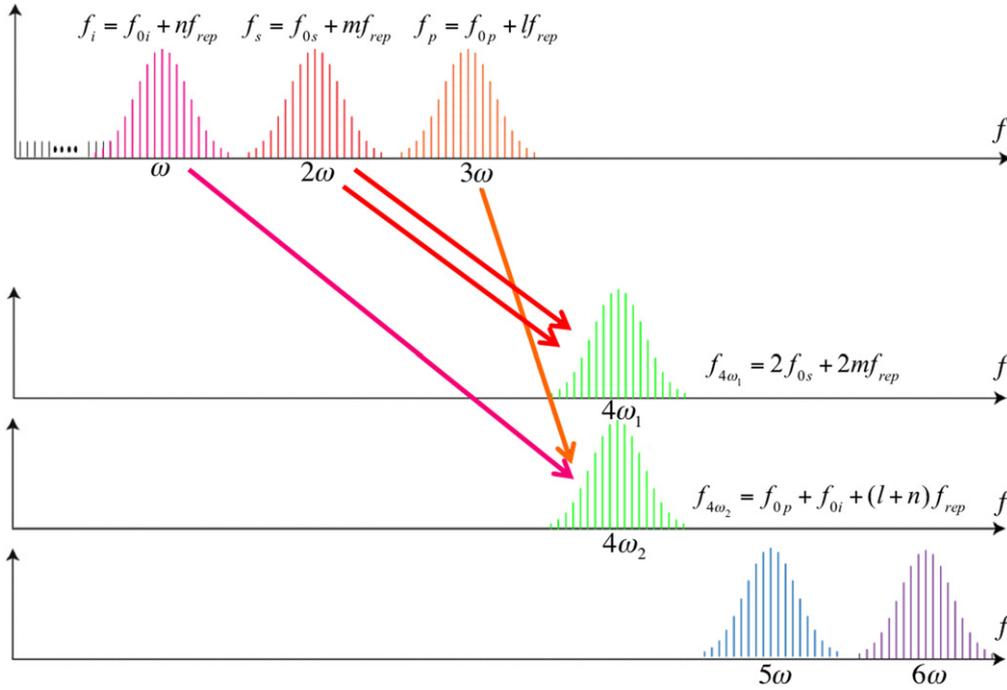


Figure 3. Relative CEP relations between sub-harmonics in a 3:2:1 OPO. The pump (3ω), signal (2ω) and idler (ω) combs can mix either by sum-frequency generation or second-harmonic generation to form two new combs around the same frequency ($4\omega_1$ and $4\omega_2$), whose heterodyne beat reveals the relative offsets between the pump, signal and idler combs. The same approach can be extended to higher harmonics (5ω and 6ω). See text for full details.

produce new pulse trains due to sum-frequency mixing (SFM) and second-harmonic generation (SHG). The new corresponding frequency combs can be described by

$$f_{\text{SFM/SHG}} = qf_{0p} + rf_{0s} + sf_{\text{rep}}, \quad (6)$$

with the idler CEP not being explicitly shown as it can be eliminated using equation (3). Along with the generation of the signal and the idler, the second harmonic of the signal ($4\omega_1$) and the sum frequency between the pump and the idler ($4\omega_2$) are produced and their optical frequencies can be set to be equal. The resulting beat frequency (f_{beat}) from the two generated 4ω signals can be expressed as:

$$f_{\text{beat}} = 2f_{0s} - (f_{0p} + f_{0i}) = 3f_{0s} - 2f_{0p} = f_{0s} - 2f_{0i}. \quad (7)$$

If the beat frequency is zero, i.e. the offset frequencies have a ratio of $f_{0i}:f_{0s}:f_{0p} = 1:2:3$, then according to equations (3) and (5), the CEP slips follow the same ratio $\Delta\theta_i: \Delta\theta_s: \Delta\theta_p = 1:2:3$. Since the wavelength ratio between the pump, the signal, and the idler, in this order, is 1:2:3, the CEP slip incurred during successive pulses remains the same in the time domain. In this case, the synthesized electric field shape does not change from pulse-to-pulse. Only the position of the synthesized field relative to the synthesized pulse envelope shifts, in accordance to the absolute value of f_{0p} . The sum frequency between the pump and the signal (5ω), and the second harmonic of the pump (6ω), are also generated. The femtosecond PPLN OPO pumped by a Ti:sapphire laser thus generates seven differently coloured pulses simultaneously. The offset frequency relation between the pump and the

signal can be controlled by tuning the cavity length. Slight changes to the cavity length during laser oscillation do not affect the repetition rate. Only the carrier frequency shifts to compensate the intracavity dispersion to realize the same repetition frequency as the pump. Under these conditions the slight cavity-length change possible by using a piezoelectric transducer (PZT) causes the phase relation to change in the OPO.

This method for observing and controlling the phase relation was first proposed and demonstrated in the early 2000s [40, 41]. Around the same time, the observation and the control of the CEO frequency in Ti:sapphire lasers was achieved, which led to the realization of the optical frequency comb and CEP control in oscillators [42, 43]. For coherent synthesis, establishing a phase relation of $\Delta\theta_i: \Delta\theta_s: \Delta\theta_p = 1:2:3$ fixes both the relative phase relation in the time domain and the synthesized electric field shape shift with respect to the phase slip of the idler. If the relative phase is fixed to $\Delta\theta_s = \Delta\theta_p$ then the pump and the signal have the same offset frequency, which implies that they can be synthesized as a single pulse. CEP control and subsequent superposition of two-colour pulses in an OPO makes it possible to realize a synthesized envelope shape which is shorter than those of the original pulses [44]. The combination of relative phase measurements with the absolute phase measurement of the pump makes it possible to fix all the parameters. By using a similar technique, one can superimpose independent laser pulses with different wavelengths coherently for field synthesis. For example, the phase control of a Ti:sapphire laser and another type of mode-locked laser with different colour can

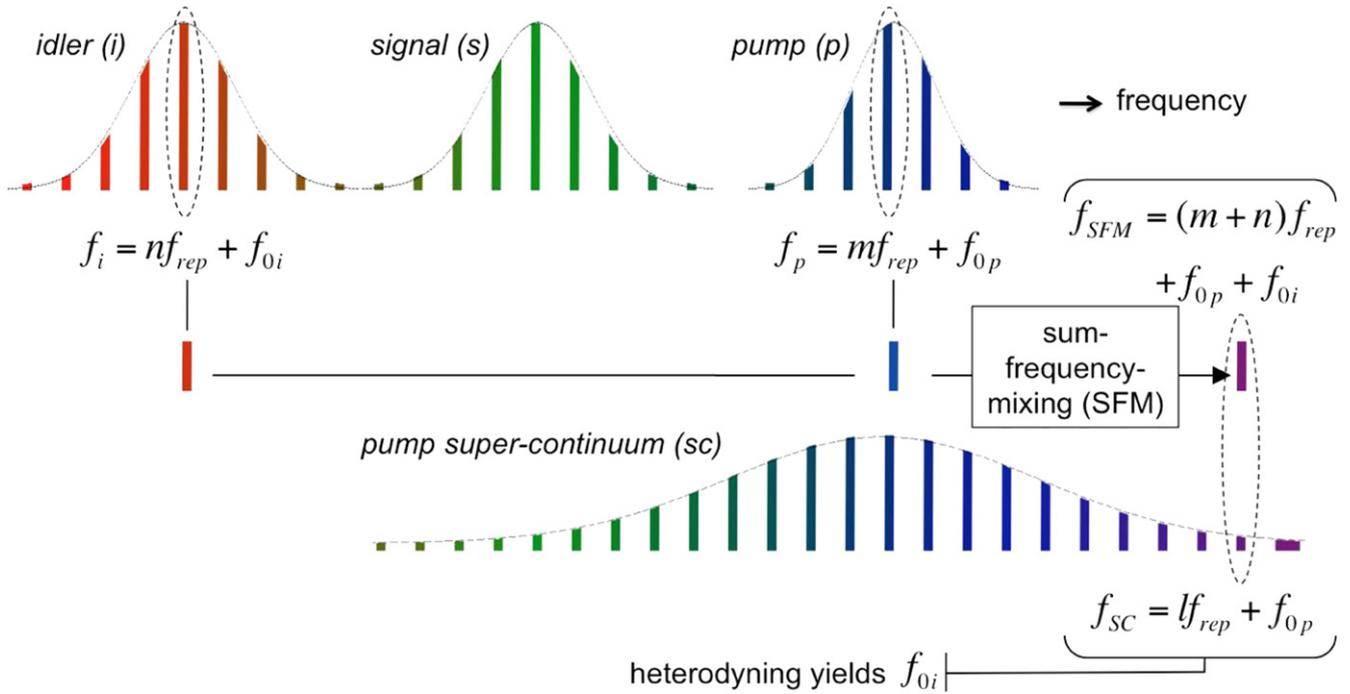


Figure 4. Illustration of the measurement of the idler-comb carrier-envelope offset frequency, f_{0i} . Sum-frequency mixing between the pump (p) and idler (i) combs yields a secondary comb with an offset of $f_{0p} + f_{0i}$. Heterodyning this comb with pump super-continuum light of the same wavelength (comb offset f_{0p}) generates a beat frequency which eliminates the pump comb offset leaving only frequencies at the harmonics of the repetition rate and $\pm f_{0i}$.

lead to the realization of an ultra-broadband spectrum for ultrashort pulse generation [45–49]. Direct frequency conversion from a long-wavelength laser is another possible candidate for producing phase-coherent multi-colour pulses. Recently, Cr-doped crystals have been shown to be suitable candidates for the production of mid-infrared femtosecond pulses [5, 6]. Tm-doped fibres [4] are also promising possibilities, since the pulses can be amplified to high average powers.

Ultimately, a difficulty still remains in the direct measurement of the shape of the synthesized electric field when using the low-energy pulses available from OPOs. The superposition of parallel optical parametric amplifiers makes it possible to increase the peak power to estimate the absolute phase relation between different colour pulses. Recently, multi-colour OPAs were developed for high-intensity coherent synthesis at 1 kHz repetition frequency [48, 49] and higher repetition-rate OPAs have also been demonstrated [50].

3. Single-resonant femtosecond OPO frequency combs

In a SRO, the CEO frequency of the signal (or idler) pulses can be measured by using interference between a pump super-continuum and the pump + signal (or pump + idler) SFM light. As an example, figure 4 illustrates how the heterodyne beat between the pump super-continuum comb, $f_{SC} = l f_{rep} + f_{0p}$, and the comb corresponding to the pump + idler SFM light, $f_{SFM} = (m + n) f_{rep} + f_{0p} + f_{0i}$, contains the idler carrier-envelope offset frequency f_{0i} , and harmonics of

the laser repetition frequency, f_{rep} . By detecting this heterodyne beat experimentally and locking it to a stable frequency reference it is possible to stabilize the carrier-envelope offset frequency of an SRO femtosecond idler frequency comb. An equivalent approach can also be taken to stabilize the signal comb by beating pump + signal sum-frequency light with the pump super-continuum. In either case, with the additional constraint of stabilizing the pump laser repetition frequency, a fully stabilized frequency comb can be realized, in which the frequencies of every optical mode can be precisely known.

SRO frequency combs were first demonstrated in 2007 by Sun *et al* [20], in which a composite frequency comb spanning 0.4–2.4 μm was achieved by phase locking a femtosecond Ti:sapphire laser and an OPO to a common super-continuum reference. A custom-built Kerr-lens modelocked Ti:sapphire laser (200 MHz, 800 nm, 1.3 W, 50 fs) was used to synchronously-pump an OPO based on a 1 mm long MgO:PPLN crystal. An end mirror of the pump laser was mounted on a PZT to lock the repetition rate to a stabilized external 200 MHz reference. Approximately 20% of the pump power was launched into a 30 cm long photonic crystal fibre (PCF) to obtain an octave-spanning super-continuum for f -to- $2f$ self-referencing [42]. The detected CEO frequency f_{0p} was controlled using a feedback loop, with an acousto-optic modulator in the Ti:sapphire pump beam used to modulate the intracavity dispersion of the laser. The remaining 80% of the pump power was used to pump the OPO, which was resonant from 1.2 to 1.37 μm . Last-coherence length and higher-order phase-matching in the OPO crystal produced a number of

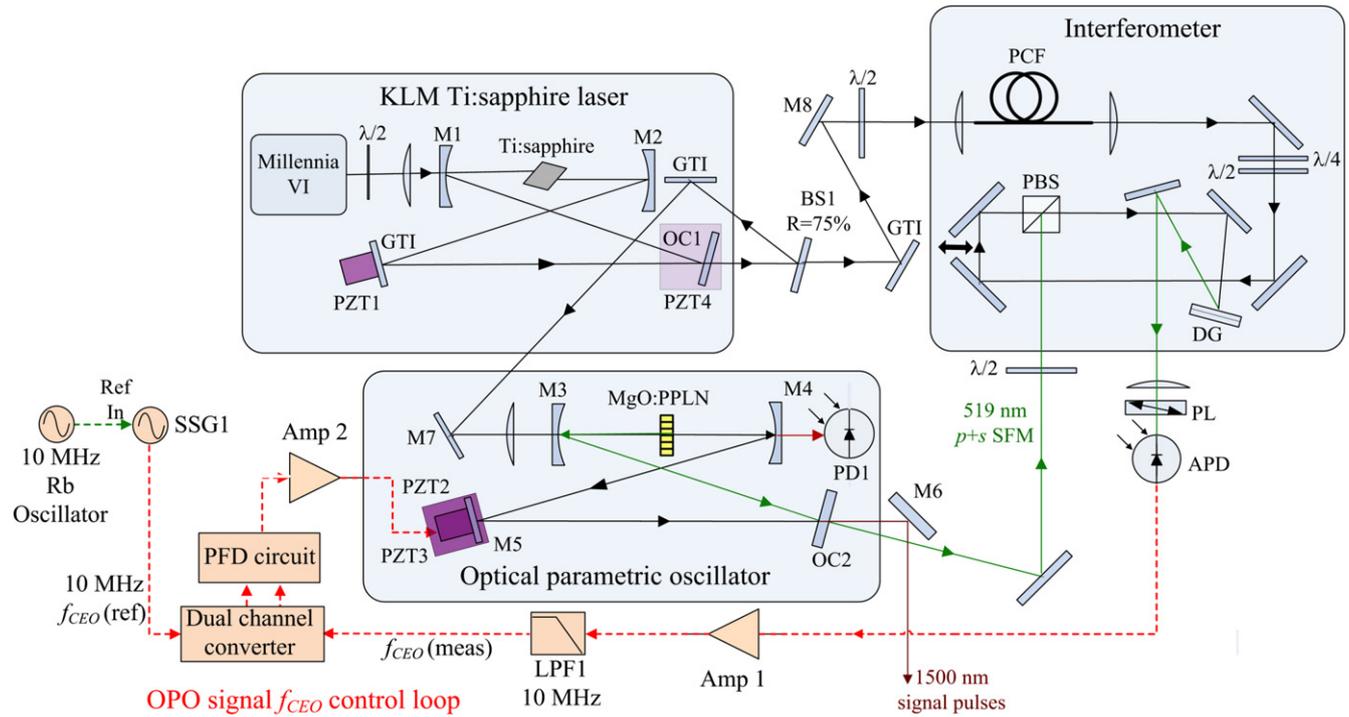


Figure 5. Schematic of the optical and electronic layouts used to generate an OPO frequency comb without f -to- $2f$ self-referencing. Reprinted with permission from [51].

SHG and SFM pulses which, combined with the non-resonant idler, spanned the visible to mid-IR spectral regions.

The SHG signal ($2s$) was heterodyned against the pump super-continuum in a nonlinear interferometer to obtain a beat frequency at $2f_{0s} - f_{0p}$, which was used to drive PZT2 in the OPO cavity and complete the phase-locking loops for the pump and $2s$ pulses, providing full CEO control of all generated frequency combs. A heterodyne frequency between the pump super-continuum and pump + idler (pi) SFM pulses was used to assess the locking quality of the experiment, which was found to stabilize the comb lines to a near-instrument-limited linewidth of 1.2 kHz.

In 2009 Adler *et al* [25] used an amplified Yb:fibre laser (136 MHz, $1.07 \mu\text{m}$, 10 W, 100 fs) to pump an OPO with a 7 mm long MgO:PPLN crystal. The OPO cavity had high finesse at the resonant signal wavelength, and produced up to 1.5 W of idler power, tunable from 2.8 to $4.8 \mu\text{m}$. The higher pump power and intracavity signal power generated an increased number of parasitic mixing pulses at the mW-level, including wavelengths as low as 360 nm ($4s$). The pump super-continuum heterodyning method described above was implemented with both ps and pi pulses, with the f_{0s} beat from the former used to stabilize the OPO cavity length and the latter f_{0i} beat used to modulate the fibre laser's pump-diode power, which acts on f_{0p} . After stabilization of the Yb:fibre repetition rate, the OPO comb quality was measured using an out-of-loop 10 kHz linewidth external cavity diode laser. Using this method the idler comb linewidth was found to be 40 kHz.

In 2010 Ferreiro *et al* [23] demonstrated that the CEO frequency of the pump laser does not need to be controlled in

order to achieve high-quality stabilization of the OPO signal pulses. A free-running Ti:sapphire laser (280 MHz, 793 nm, 1.4 W, 50 fs) was used to pump an OPO based on 1 mm of MgO:PPLN which was signal resonant over $1.42\text{--}1.60 \mu\text{m}$ (figure 5). 350 mW of pump light was used to generate a super-continuum which was interfered with ps pulses at 520 nm. The resulting beat provided the CEO frequency of the signal pulses, and was controlled through a PZT mounted on an OPO cavity mirror. The locked beat had an instrument-limited bandwidth of 1 kHz, and phase noise PSD measurements showed a cumulative phase error of 0.38 rad in 1 s, integrated up to 1 MHz.

Further investigations of these results [16] were carried out and the cavity length of the Ti:sapphire laser stabilized to the 8th harmonic of the repetition rate (2.24 GHz), referenced to a synthesized signal generator. Both the signal generator and the f_{0s} beat were referenced to the same 10 MHz Rb clock. Locking the pump repetition rate reduced the linewidth of the offset frequency beat, f_{0s} , which was measured to be 15 Hz at the -3 dB point with an instrument resolution of 10 Hz. The cumulative phase noise in this system was measured to be 0.56 rads in 1 s, integrated over 2 MHz. Two-sample frequency deviation measurements [52] of the comb instability implied fractional stabilities of 2×10^{-11} (f_{rep}) and 1×10^{-15} (f_{0s}), comparable with the 2×10^{-11} instability in the Rb clock.

At Heriot-Watt University we have recently demonstrated a set of fully stabilized frequency combs from a 1 GHz fundamentally-pumped OPO that spanned the visible and IR. CEO frequency stabilization of the Ti:sapphire laser (Gigajet, Laser Quantum) was achieved in a standard f -to- $2f$ interferometer, with f_{0p} controlled via diode-current modulation in

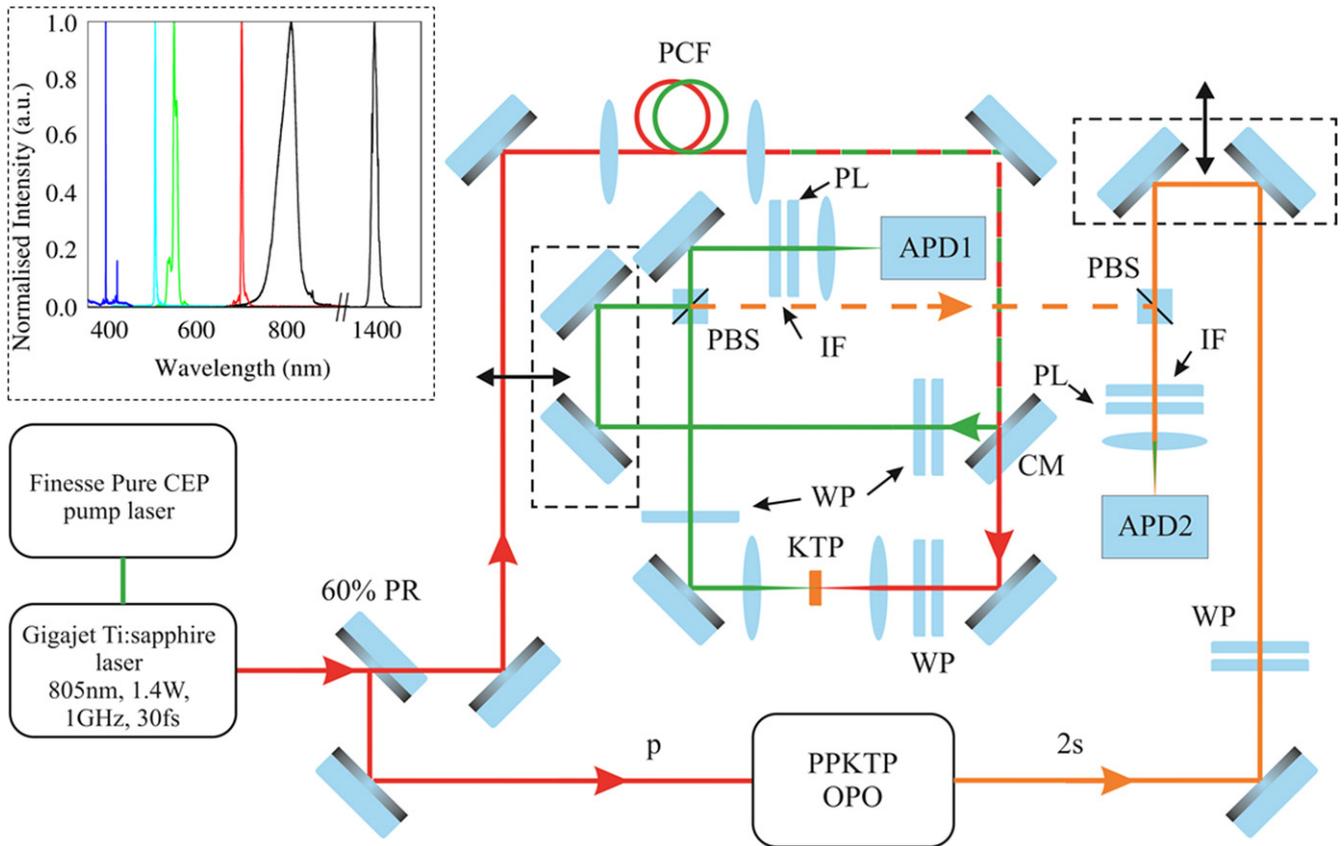


Figure 6. Optical layout of the scheme used to simultaneously lock all OPO frequency combs. APD, avalanche photodiode; CM, cold mirror; IF, interference filter; PBS, polarizing beam splitter; PCF, photonic crystal fibre; PL, polarizer; PR, partial reflector; WP, wave plate. Inset: spectra of the composite comb comprising SHG pump (400 nm); pump + signal (510 nm) and pump + idler (565 nm) SFM light; SHG signal (700 nm); pump (805 nm); and signal (1400 nm). The idler spectrum (1900 nm) is not shown.

the pump laser (Finesse Pure CEP, Laser Quantum). In a complementary arrangement, $2s$ pulses were heterodyned against a different portion of the pump super-continuum to obtain a beat that was then used to lock the difference between the pump and signal CEO frequencies. Both CEO frequencies were locked to a 10 MHz Rb clock, which also acted as the frequency reference for the repetition-rate stabilization of the Ti:sapphire laser. The optical layout is shown in figure 6, with the spectra of the composite comb inset. Controlling both the Ti:sapphire laser and the OPO frequency combs automatically stabilizes all other combs generated on the bench, as is evident from equation (6).

PSD measurements of the Ti:sapphire locking loop showed a cumulative phase noise of 0.4 rad over 64 kHz, with the dominant noise at 6 and 33 kHz attributable to RIN fluctuations of the DPSS pump laser. The cumulative phase noise for the OPO was 1.6 rad over 64 kHz, with stability limited by the corner frequency of the PZT used for cavity length control. The high repetition rate of the Ti:sapphire laser resulted in low peak power pulses, requiring significant levels of average power (>500 mW) to generate a coherent octave spanning super-continuum for f -to- $2f$ locking. The accompanying decrease in the available average power for the OPO restricted its operational range to 1375–1425 nm.

Partially-stabilized SRO combs have also been demonstrated in a number of forms. Kobayashi *et al* achieved phase locking among various harmonically related outputs of an OPO operating in a 3:2:1 pump:signal:idler frequency ratio [39, 40]. Sun *et al* extended this to an OPO with no fundamental wavelength constraints by exploiting dual-colour operation, which is possible when the net cavity group delay of the OPO is identical for two different signal wavelengths [53]. Internal beat frequencies were observed in the visible at wavelengths corresponding to ps , $2s$ and pi , which could be stabilized for use in coherent pulse synthesis [44].

A multi-octave-spanning phase coherent composite OPO frequency comb was recently reported by McCracken *et al* [28]. The CEO frequencies of the pump, signal and idler pulses were locked to the same frequency comb grid by using a zero-offset frequency locking technique [26]. Through parametric energy conservation [22], all parasitic SHG and SFM combs generated in the OPO were also constrained to this same frequency grid. The natural synchronisation of the OPO with the pump laser allowed the composite frequency comb to be realized without the need for complex locking electronics between separate oscillators [54, 55], making this system an ideal candidate for high-repetition rate coherent pulse synthesis.

Recently, Balskus *et al* extended the control of SRO frequency combs to the harmonically-pumped regime [27]. Femtosecond OPOs are typically synchronous with their pump laser, with the cavity length of the former matched to the latter. In harmonic pumping the OPO cavity length is chosen to be an integer [56, 57] or integer fraction [58, 59] of the pump cavity length, allowing high repetition rate pulse trains to be generated without the need for high repetition rate oscillator. Balskus *et al* used a 333 MHz Ti:sapphire laser to pump an OPO constructed in a fundamental (333 MHz) and harmonic (1 GHz) configuration, locking the CEO frequency of the signal in both cases, and compared the locking stability. The fractional stability of the locked f_{0s} for both OPOs was 1.35×10^{-15} over a 1 s gate time, demonstrating that harmonic pumping does not reduce the comb quality despite the signal pulses making multiple trips round the cavity, which might be expected to increase environmental noise contributions. It should be noted that while the harmonic OPO comb itself is stable, the underlying comb structure will contain additional sidebands at multiples of the pump repetition rate.

Progress in SRO frequency combs has been encouraging, with instabilities in the positions of the comb lines limited only by the clocks used as a locking reference. Phase instabilities below 200 mrad are readily achievable, making OPO frequency combs competitive with the best laser combs operating in the near-infrared region. Future developments will undoubtedly include extensions to longer wavelength regions which are ideal for spectroscopy and other sensing applications, as well as pushing the boundaries of fundamental pumping using high repetition rate oscillators [60].

4. Doubly-resonant femtosecond OPO frequency combs

In DROs the signal and idler waves resonate in the OPO, and the simultaneous cavity field enhancement for the signal and idler can lead to low oscillation thresholds. The threshold of a DRO can be lower than an SRO with the same gain by a factor of half the round-trip power loss [61]. This feature of DROs reduces the gain requirement and consequently compact, low-power, and quiet pump sources can be used. Moreover, shorter nonlinear crystals can be used, which usually results in broad phase matching bandwidths.

The frequencies of the signal and idler must satisfy two criteria: the energy conservation condition and the resonance condition. Therefore, unlike SROs, the wavelength tunability is limited. In most cases, the cavity is shared between the signal and idler, and scanning the cavity length results in discrete oscillation peaks, with widths in the order of a tenth of the wavelength. Achieving stable operation for long times usually requires active stabilization, and can be challenging in the presence of environmental noise or frequency noise on the pump, especially if the frequencies of the signal and idler are far apart.

In the frequency comb regime, the pump is a frequency comb, and the signal and idler are expected to be frequency

combs as illustrated in figure 7(a). In the non-degenerate operation, the signal and idler are well separated from each other. They can become close to each other to spectrally overlap, which is shown as partly-degenerate operation in figure 7(a). In this condition, the signal and idler can be independent frequency combs with different CEO frequencies resulting in beating in the overlapping portion of the spectrum [62]. It has been shown by numerical simulations and in experiments that the signal and idler can become a single frequency comb, where the beating disappears [63, 64]. The rich physics of the nonlinear interactions between the combs in the transition from non-degenerate to degenerate operation is yet to be studied comprehensively.

At degeneracy, the signal and idler waves become indistinguishable, and as a consequence, the phase and frequency of the output is automatically locked to the pump. This is an important feature of degenerate femtosecond OPOs because they can offer mid-infrared frequency combs at their output which are intrinsically phase and frequency locked to the near-IR pump frequency comb [65]. When both the repetition and CEO frequencies of the pump laser are stabilized then the entire pump-OPO system becomes a fully locked frequency comb.

In this regime, the optical carrier frequency of the signal pulses is half the carrier frequency of the pump pulses (the opposite of SHG). The relationship between the phases of pump and signal is given, modulo 2π , by:

$$\theta_p = 2\theta_s + \frac{\pi}{2}, \quad (8)$$

where θ_p and θ_s are the optical carrier phases of pump and signal, respectively [65]. The comb lines of the output frequency comb at degeneracy are defined by:

$$f_m = \frac{f_{0p}}{2} + mf_{\text{rep}}, \quad (9a)$$

or

$$f_m = \frac{f_{0p}}{2} + mf_{\text{rep}} + \frac{f_{\text{rep}}}{2}. \quad (9b)$$

These two frequency states are different by a shift of half the repetition rate, and the OPO operates in one of them, depending on which one is supported by the resonator. Switching from one state to another is achieved by changing the round-trip cavity length by approximately half of the OPO centre wavelength [65].

Identifying the degenerate operation of OPOs in the frequency comb regime, where the above conditions are satisfied, is sometimes challenging. For instance, operation of femtosecond DROs at many times above the oscillation threshold can result in multiple frequency comb outputs, similar to the micro-resonator-based OPOs [66]. Also, in the partly degenerate and non-degenerate OPO (figure 7(a)) independent signal and idler frequency combs can be observed which do not follow the above equations [67]. In general, where the signal and idler cannot be distinguished spectrally, and the OPO is operating at only a few times above

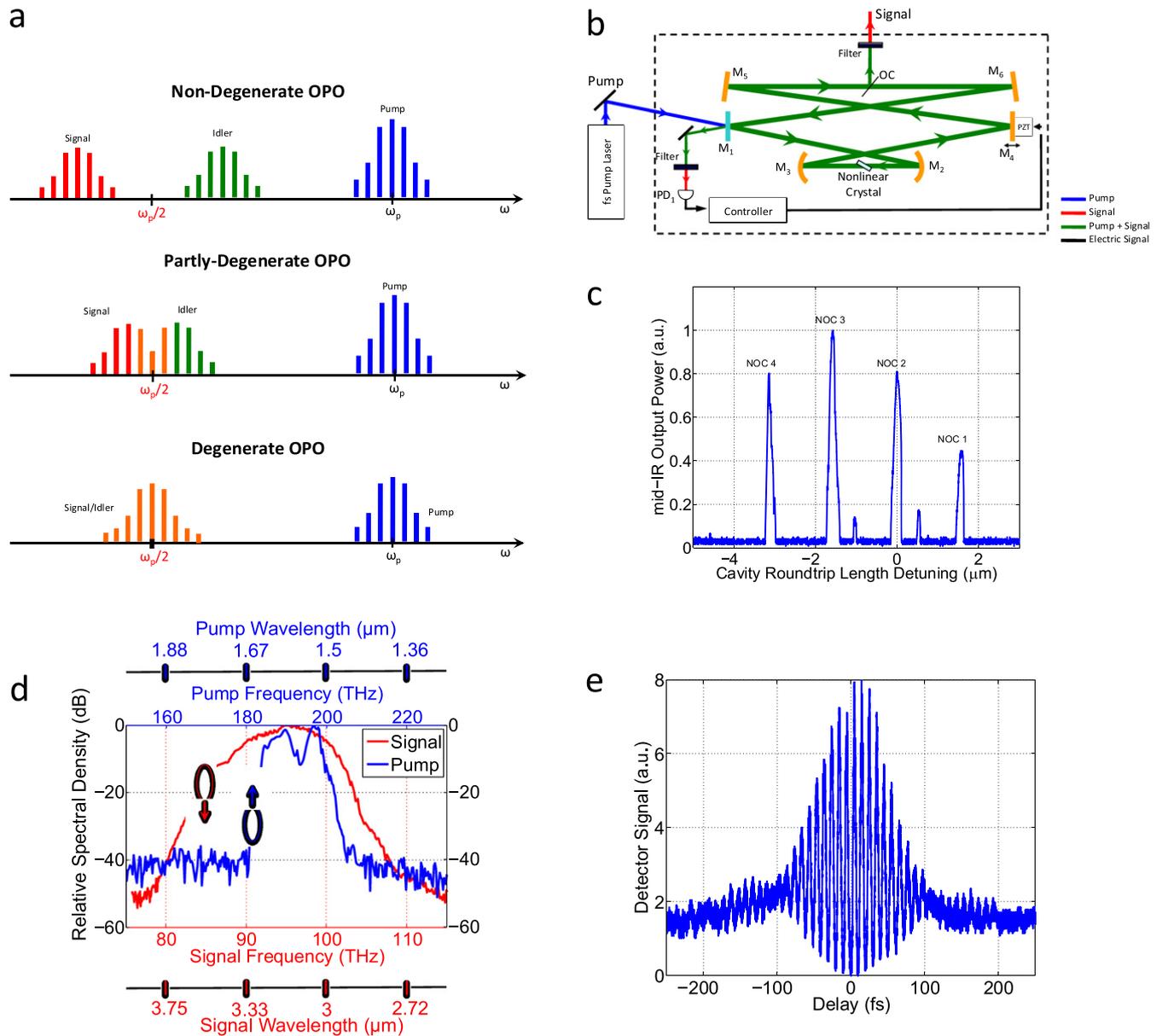


Figure 7. (a) Illustration of the pump, signal and idler frequency combs in non-degenerate, partly degenerate, and degenerate frequency comb OPOs, (b) schematic of a degenerate OPO pumped by a femtosecond pump laser, (c) oscillation peaks of a frequency comb OPO operating around degeneracy, (d) pump and output spectra of a degenerate OPO [65], (e) autocorrelation trace of a degenerate OPO output at 3.1 μm [65].

the oscillation threshold, the operation is most likely degenerate with established phase and frequency locking over the spectrum [65].

Several degenerate frequency comb OPOs have been demonstrated over the past few years for extending the well-established mode-locked frequency comb sources to longer wavelengths with salient features of low pump power requirement, phase- and frequency- locked operation, and broadband outputs. Different femtosecond pump lasers, such as Ti:sapphire at around 800 nm, mode-locked fibres lasers at around 1 μm (Yb-doped), 1.5 μm (Er-doped), and 2 μm (Tm-doped) have been exploited to generate femtosecond frequency combs at 1.5 μm [68, 69], 2 μm [70], 3 μm [65], and 4 μm [29]. Different nonlinear crystals have been used for

realization of such OPOs ranging from PPLN [65, 68, 70] to orientation-patterned GaAs [71] and BiBO [69].

The dominant resonator designs for these OPOs have been the bowtie ring resonator or equivalent folded versions, as depicted in figure 7(b), with a dichroic dielectric mirror (M_1) employed for introducing the pump light. The remaining mirrors are usually metal-coated for maximum reflectance in mid-IR. The group velocity dispersion in the cavity is usually compensated to be close to zero and it has been shown that fine-tuning the intracavity dispersion can lead to few-cycle signal pulses in the mid-IR [29]. The crystal length is usually chosen to be short (close to the walk-off length of the pump and signal wavelengths) providing broad gain bandwidth and limiting the dispersion in the cavity. Mode-

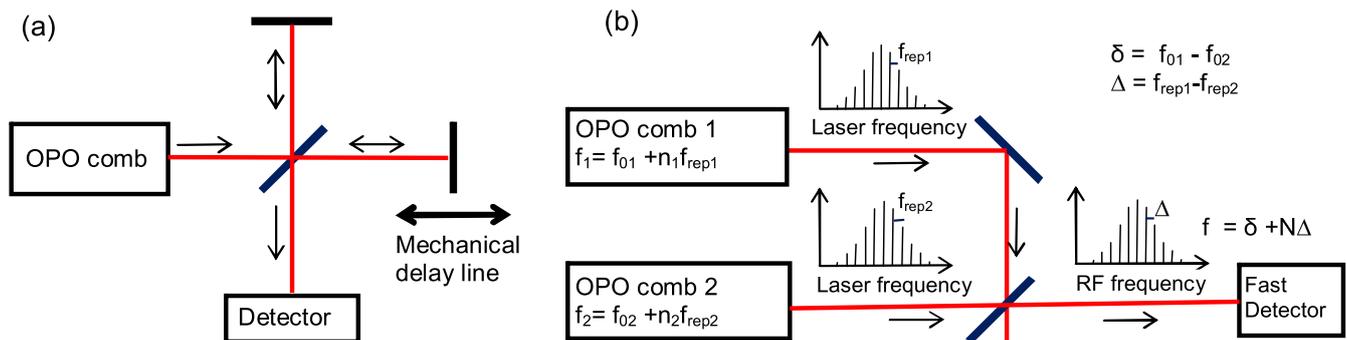


Figure 8. Layout of (a) a single-comb FTIR spectrometer and (b) a dual-comb FTIR spectrometer.

matching lenses are usually employed to achieve optimum pumping.

Similar to synchronously-pumped SROs, the round-trip time of the resonator is matched to the repetition period of the pump, however, because of the phase-sensitive nature of the gain at degeneracy, when the cavity lengths of such OPOs are scanned, multiple oscillation peaks are observed, as depicted in figure 7(c). These peaks are usually separated by half the wavelength of the signal, corresponding to the round-trip phase shift steps of π . Higher-order spatial operation can be observed as extra peaks. For instance, the two weakest peaks of figure 7(c) correspond to operation at TEM_{01} , and the rest are TEM_{00} . Depending on the intracavity dispersion, pump power, and the spectral response of the cavity, different cavity lengths can correspond to non-degenerate, partly-degenerate, and degenerate operation, and the OPO can be locked to a peak for continuous operation. Active stabilization is usually implemented to achieve stable long-term operation lasting for days [65]. In some cases, thermal stabilization has resulted in stable operation over shorter timescales [68].

The dominantly used stabilization method is the top-of-fringe locking that has been widely used for stabilizing cavities and interferometers. A small length modulation (~ 10 nm in the kHz range) is applied to the OPO cavity, and the error signal is generated by phase-sensitive detection of the applied modulation frequency in the output signal through a lock-in amplifier or an electronic mixer. The error is then applied to a PZT on the OPO cavity (such as M4 in figure 7(b)) to achieve stable operation at the centre of one of the oscillation peaks [65]. Figure 7(d) shows the pump and output spectrum of a degenerate femtosecond OPO pumped by an Er-doped mode-locked fibre laser, and the corresponding autocorrelation trace is depicted in figure 7(e), suggesting shorter than 100 fs pulses centred at $3.1 \mu\text{m}$.

The phase and frequency locking is verified experimentally by achieving stable interference between the output femtosecond pulses of two independent degenerate OPOs pumped by the same pump [65]. It is also verified experimentally that for a free-running mode-locked pump laser, the second-harmonic of the OPO output has a comb linewidth of about 140 kHz which is the same as the comb linewidth of the free-running pump [72]. Preliminary results suggest that this behaviour can be present in cases where sub-Hz linewidths are used for the pump [73], however a comprehensive study

of the strength of this intrinsic phase and frequency locking is yet to be undertaken.

In principle, cw OPOs can be very efficient and pump depletions greater than 90% have been reported. It is expected that the efficiency of half-harmonic generation in cw degenerate OPOs can be as high as 90% and beyond [74]. So far, a conversion efficiency of slightly higher than 60% has been achieved for half-harmonic generation of sub-50 fs femtosecond pulses at $2 \mu\text{m}$ [75].

5. Current and future applications of femtosecond OPO frequency combs

Stabilized OPO frequency combs, with their exceptional versatility in spectral coverage spanning the visible, near-infrared and mid-infrared wave bands, present opportunities for innovative applications in frequency metrology, spectroscopy and quantum optics. Here we outline some emerging and potential applications which make use of the unique properties of SRO and DRO frequency combs.

5.1. Spectroscopy

The mid-infrared (mid-IR) spectral region is well suited for sensing molecules, since it contains strong characteristic rovibrational transitions of most molecular species. As already discussed, broadband OPOs are readily designed to generate combs in this important spectroscopic region.

Using broadband mid-IR OPO frequency combs, spectroscopy has been conducted with two different schemes. One way is directly using OPO combs as broadband photonic sources to replace widely used thermal light sources in a Michelson-interferometer based FTIR spectrometer, consisting of a moving mirror mounted on a mechanical delay line, as illustrated in figure 8(a). This is called single-comb FTIR spectroscopy. In contrast with an FTIR using an incoherent thermal light source, an FTIR spectrometer using a spatially coherent OPO frequency comb source dramatically reduces the measurement time owing to the high spectral brightness. An FTIR spectrometer based on an OPO frequency comb source supports the use of a multi-pass cell for enhancing detection sensitivity, and allows long-range open-path sensing and FTIR micro-spectroscopy. FTIR spectroscopy with an

unstabilized OPO comb was first demonstrated by Tillman *et al* [76]. Adler *et al* reported the first implementation of high-resolution gas spectroscopy with a fully-stabilized OPO comb, achieving a spectral resolution of 0.0056 cm^{-1} and part-per-billion detection limits in 30 s of integration time [32]. Using intracavity spectroscopy, Haakestad *et al* demonstrated ultrasensitive detection of several different gas species in the spectral range $2.5\text{--}5\text{ }\mu\text{m}$ by use of a broadband DRO comb, sharing coherence with its pump laser but not stabilized to an absolute reference [77].

A new approach for implementing frequency-comb spectroscopy is the dual-comb spectrometer, illustrated in figure 8(b), in which two asynchronous OPO combs with slightly different repetition rates (or mode spacings) are heterodyned before or after passing through a sample, yielding a down converted radio-frequency comb that contains information on the absorption experienced by the combs. The absence of moving parts in dual-comb spectrometers overcomes the speed and resolution limitations of a Michelson-based spectrometer, and its resolution is only limited by the comb line-spacings. The recording time is the inverse of the difference in the repetition rates of combs. Dual-comb spectroscopy was initially demonstrated in mid-IR region by low power DFG combs [78, 79] or in near-IR region by mode-locked laser frequency combs [80]. Zhang *et al* demonstrated the first OPO-based mid-infrared dual-comb spectroscopy by use of a unique scheme, in which two asynchronous mid-IR combs originated from one single OPO, and shared all the components for mid-IR generation and CEO-frequency detection [31, 81, 82]. Others have extended this approach by using two crystals in a shared OPO cavity produce two idler beams suitable for heterodyne detection of the dual-comb interferogram [83].

5.2. Frequency metrology and calibration

Femtosecond OPO frequency combs are ideal sources for frequency calibration in the near- to mid-IR, for applications in precision spectroscopy. Since stabilized OPO combs are not widely available, in all current demonstrations, frequencies of mid-IR lasers (e.g. quantum cascade lasers or single-frequency OPO sources) were calibrated either indirectly against a near-IR frequency comb via a nonlinear process [84, 85] or directly to a low power DFG comb [86]. Potentially, OPO frequency combs, which can provide higher power and direct coverage in the mid-IR region, will facilitate the implementation of precision mid-IR spectroscopy on a wider scale. In a preliminary study we used a $1.56\text{ }\mu\text{m}$ femtosecond OPO comb to measure the absolute frequency of a Rb-referenced optical frequency standard, demonstrating performance fully matching that of a commercial fibre comb in the same near-infrared wavelength region [87]. In the near future we anticipate that femtosecond OPO combs will be suitable calibration sources for precision wavemeters and Fourier-transform spectrometers operating in the near- and mid-infrared.

5.3. Quantum optics and optical computing

Apart from using femtosecond degenerate OPOs as a tool for frequency-locked translation of the existing frequency comb sources to the longer wavelengths, they have rich quantum properties around the oscillation threshold. These properties are the consequence of the phase-sensitive nonlinear gain, which has been effectively used over the past few decades as a tool for generation of single-mode squeezed vacuum states in the cw regime [88]. It is also predicted that cw degenerate OPOs can potentially provide a qubit-like behaviour around the oscillation threshold [89]. In the frequency comb regime, non-classical behaviours of degenerate OPOs have been demonstrated below threshold [90], and they are utilized for realization of quantum networks [91] and all-optical quantum random number generation [33].

A network of time-multiplexed femtosecond degenerate OPOs have been utilized as a computing machine for Ising problems [34]. These problems map to many real-life combinatorial optimization problems, and belong to the non-deterministic polynomial-time (NP)-hard computational complexity class, for which no efficient classical and quantum algorithm exists. The performance of the OPO network in experiment and numerical simulations is promising as a new path for tackling these problems.

5.4. Astrophotonics

Laser frequency comb technology is emerging as the calibration source of choice for high resolution astronomical spectroscopy, however specific technical challenges stem from the difficulty of providing exceptionally wide comb-line spacings (6–32 GHz) across a three-octave wavelength range from 380 nm (Ca II H and K lines) to $2.4\text{ }\mu\text{m}$ (CO band at $2.3\text{ }\mu\text{m}$). Current solutions employ Fabry–Pérot filter cavities to increase the comb-line spacing, then amplify this filtered comb before launching into highly-nonlinear fibre to generate the required spectral broadening [92]. A technical issue with this approach (so far, only resolved by using multiple cascaded Fabry–Pérot filter cavities) is that nonlinear fibre-optic spectral broadening degrades side-mode suppression and causes side-mode amplitude asymmetry, which translates into wavelength calibration errors [93, 94]. The remarkably low thresholds of DRO combs offer an opportunity to realize high brightness, swept-wavelength calibration combs at multi-GHz repetition rates by direct pumping using multi-GHz Ti:sapphire lasers, such as the 10 GHz system reported in 2008 by Bartels *et al* [60].

6. Conclusions and perspectives

In this article we have highlighted the advances made in the phase control of femtosecond OPOs, which led to the demonstration of OPO frequency combs. Such OPO combs share many common characteristics with their laser counterparts, however—as we have shown here—they also possess unique spectral and temporal properties that allow them to be

exploited in applications beyond those addressable by currently available laser combs. For example, in the future we envisage low-threshold DRO frequency combs operating at high repetition rates, allowing them to be directly referenced to atomic transitions. The inherent quantum entanglement in such systems, and the potential to generate high repetition rate pulses containing only hundreds, rather than *hundreds of thousands* of modes, will make possible new continuous-variable approaches to quantum optics, acting as potential resources for quantum information processing. The extension of OPO combs to the spectral fingerprint region, for example using materials such as CdSiP₂ [95], will also open new opportunities for dual-comb spectroscopy to become a powerful diagnostic tool by providing kHz-rate ultra-high-resolution mid-IR spectroscopy for the analysis of systems characterized by complex and rapidly changing chemical signatures. Phase-stabilized OPO combs, based on the techniques described earlier, may also become useful as seed sources for mid-IR laser amplifiers, enabling CEP-stable μJ pulses to be generated for high-harmonic generation, potentially at high repetition rates.

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