Optical frequency combs were developed nearly two decades ago to support the world’s most precise atomic clocks. Acting as precision optical synthesizers, frequency combs enable the precise transfer of phase and frequency information from a high-stability reference to hundreds of thousands of tones in the optical domain. This versatility, coupled with near-continuous spectroscopic coverage from microwave frequencies to the extreme ultra-violet, has enabled precision measurement capabilities in both fundamental and applied contexts. This review takes a tutorial approach to illustrate how 20 years of source development and technology has facilitated the journey of optical frequency combs from the lab into the field.
single mode-locked laser (MLL), enabling vast simplification to precision optical measurement and rapid progress and development into optical atomic standards.

OFCs were developed by drawing on single-frequency laser stabilization techniques and applying them to mode-locked (pulsed) laser systems. The result was a system that could synthesize 10^3–10^6 harmonically related optical modes from either an electronic or optical reference with a fidelity better than 1 part in 10^18. More importantly, OFCs enabled the direct conversion of optical-to-microwave frequencies and vice versa, enabling the extraction of microwave timing signals from optical atomic clocks. Beyond their application to precision optical metrology, OFCs were quickly recognized for their versatility as high-fidelity optical frequency converters and as sources of precisely timed ultra-short pulses. More broadly, by taking advantage of the nonlinearities of optical fibers and free-space, OFCs enable synthesis over the broad spectral regions including the near-infrared, the visible and ultraviolet (XUV). Generation of pulses. More broadly, by taking advantage of the nonlinearities that are bene

The comb equation. The deterministic behavior of the OFC spectrum described above is most succinctly described by the comb equation. To understand the comb equation, we will begin by exploring the relatively simple mathematics that describe the optical field output from a MLL (see Fig. 1). The optical field of the laser pulse train can be described by a carrier frequency, \( v_\nu = \omega_\nu / (2\pi) \), that is modulated by a periodic pulse envelope, \( A(t) \). Typically, the time between optical pulses range between 1 and 10 ns. Due to the pulse periodicity, the optical field can also be described as a periodic Fourier series of optical modes, \( \nu_N = \omega_N / (2\pi) \), with Fourier amplitude components, \( A_N \), and mode number, \( N \), such that

\[
E(t) = A(\nu) e^{i\nu t} = \sum_{N=N_i}^{N_f} A_N e^{i \nu_N t}.
\]

Because \( v_\nu \) is not necessarily an exact multiple of the mode spacing, \( f_r \), the individual Fourier frequencies are shifted from integer multiples of \( f_r \) by a common offset, \( f_0 \), such that

\[
\nu_N = N \cdot f_r + f_0,
\]

where \( N \) is an integer mode number between 100,000 and 1,000,000, that multiplies \( f_r \) from the microwave domain to the optical domain.

Equation (2) is referred to as the comb equation. What the comb equation states is that while an OFC consists of up to a million optical modes, spanning hundreds of terahertz in the optical domain, only two degrees of freedom: (1) the repetition rate, \( f_r \), and (2) the laser offset frequency, \( f_0 \), are needed to define the frequency of each individual optical mode, \( \nu_N \). This ability to completely define optical frequencies in terms of microwave frequencies was the original claim to fame for OFCs in precision optical metrology. To summarize, MLLs can enable near-perfect coherent division of optical frequencies to the microwave domain, and coherent multiplication of microwave frequencies to the optical domain.

The repetition rate \((f_r)\). The microwave mode that ties the spectrum together harmonically is the laser repetition rate, \( f_r \), which is the inverse of the pulse-to-pulse timing, \( T_r \). Pulses exit the laser cavity once per round trip such that the pulse repetition period, \( T_r = 2L/v_g \), where \( v_g \) is the pulse group velocity in the laser cavity, is defined and controlled via actuation of the laser cavity length, \( L \). Changes in \( f_r \) result in an accordion-like expansion and contraction of the frequency modes.

The offset frequency \((f_0)\). Pulse formation necessarily requires that every longitudinal laser mode is perfectly equidistant in frequency and shares a common phase. This unlikely condition is enforced by nonlinearities in the laser cavity that underlie pulse formation and spectral broadening via self-phase modulation and four-wave mixing. The harmonic and coherent connection between laser modes is manifest as a common and additive frequency offset, \( f_0 \). In the frequency domain, \( f_0 \) translates all the laser modes simultaneously. Because this offset frequency is a measure of coherence it also relates to time-changes of the optical carrier phase relative to the pulse envelope, \( \phi_{CEO}(t) \),

\[
f_0 = (1/2\pi) \cdot d \phi_{CEO} / dt
\]

that result due to dispersion induced phase- and group-velocity differences.

What is an OFC and how does it work

The traditional answer is that an OFC is a phase-stabilized MLL. While different generation methods have been developed over the past 20 years, MLLs were the original OFC sources. Because of their historical relevance and operational simplicity, we use them here as a starting point to explain the basics of OFC generation.

The utility of MLLs within the context of optical metrology was recognized as early as the late 1980s. The optical pulses from MLLs result from the coherent addition of 100s to millions of resonant longitudinal optical cavity modes, spanning up to 100 nm in the optical domain. While the broad optical bandwidth is immediately attractive for spectroscopic applications, the mode-locked optical spectrum has unique properties that are beneficial for precision optical metrology: (1) all the optical modes are harmonically related (perfectly equidistant in frequency) and (2) all-optical modes are phase coherent with one another (share a common phase evolution). The consequence of this is that the evolution of the electric field, and consequently the phase and frequency dynamics of every optical mode in the laser OFC spectrum is deterministic. As a result, knowledge about the absolute frequency of one mode can be used to determine the absolute frequency of any other mode.
In the simplest terms, $f_r$ controls the pulse-to-pulse timing, and hence the periodicity of the pulse train, $T_r$, controlling the periodicity of the pulse train, and connects the optical and microwave domains via $N f_r$. The offset frequency, $f_0$, controls the carrier-phase of the pulse train, providing fine optical frequency tuning. The detection and control of the laser offset frequency $f_0$ is the key for allowing precise frequency determination of the comb modes, as well as for control of the pulse electric field in high-field physics and attosecond science experiments. More specifically, with knowledge of $f_r$ alone, a single optical mode can only be known to $\pm f_r$. On an optical frequency, this represents an error of parts in $10^6$ depending on the mode spacing.

Full frequency stabilization of the comb is achieved using negative feedback to the laser cavity length and intra-cavity dispersion to physically control $f_r$ and $f_0$. Ensuring good mechanical stability and engineering of the stabilization loops, the above methods can enable control of the average cavity length at resolutions below a femtometer, the diameter of the proton. Applications with the highest stability requirements, or ones that require long-term accuracy and averaging, generally require...
The offset frequency and measurement of the comb parameters. While it is impossible to count optical frequencies directly, optical difference frequencies are easily measured as long as they fall within the bandwidth limit of precision frequency counters (<10 GHz). As a simple example, consider two optical carriers close in frequency, \(v_1\) and \(v_2\), that can be interfered to produce an optical carrier with an amplitude modulation at the difference frequency, \(v_\Delta = v_1 - v_2\), (see Fig. 1a). When this signal is incident on a photodetector, the detector produces a voltage proportional to the amplitude modulation. This signal is often referred to in the literature as a heterodyne optical beat frequency.

This technique of difference frequency measurement is at the heart of nearly all measurement techniques with optical OFCs, and enables access to its characteristic frequencies, \(v_0\) and \(v_r\). Detection of \(v_0\) is the extreme case of the example of two mode beating in the optical comb spectrum. The offset frequency \(v_0\) is common to each mode, or \(v_0 = N \cdot v_f + f_0 = (N - M) \cdot v_f\) where \(f_0\) is the pulse train that is output from an MLL. While high-energy ultra-short laser pulses were being explored in the 1990s for few-cycle pulse generation and spectroscopic applications, it was developments in highly engineered low-dispersion optical fibers that enabled continuum generation at lower pulse energies. These small core (1–3 μm) silica fibers balanced material dispersion with a waveguide dispersion enabled via fiber tapering or fiber cladding supported by glasses. When ultra-short pulses from a Ti:sapphire laser were launched into these fibers, the combination of small cross section, and low dispersion allowed for high-pulse intensities to be maintained over interaction lengths of several centimeters, and up to several meters. The result is coherent white light continuum generation, that, quoting directly from the text in Birks et al., “has the brightness of a laser with the bandwidth of a light bulb.” It was only a matter of months after these first demonstrations that OFCs with Ti: sapphire lasers were fully realized.

The ability to directly convert optical frequencies to the microwave domain and vice versa resulted in a rapid advance in precision metrology capabilities. Within the first four years of their realization nearly everything that could be proposed with OFCs was demonstrated. This included carrier-envelope phase control, the first all-optical atomic clocks, absolute optical frequency measurements and the measurement of optical atomic frequency ratios, searches for variation of fundamental constants, precision distance measurement, coherent bandwidth extension and single cycle pulse synthesis, attosecond control of field-sensitive processes, coherent and direct microscopy, direct molecular spectroscopy, the development of molecular frequency reference, and optical synthesis of precision electronic signals. It was absolute madness!

Emergence of comb sources, frequency generation, and new architectures
Evolution of solid-state and fiber-based mode-locked combs. Once the stabilization techniques were understood, any MLL system that had sufficiently high-pulse energy (~1 nJ) for broadening, could be converted to an OFC. As a result, highly nonlinear fibers for continuum generation were studied extensively to enable extension to different wavelengths. Advances in fiber technology along with the desire for more energy efficient OFCs yielded diode-pumped solid-state lasers systems near 1 μm based on Er:LiSAF, Yb:CALGO, Yb:KGW, Er:Yb:glass and Yb:KYW, as well as diode-pumped fiber systems emitting light in the telecommunication band around 1550 nm. The latter fiber lasers, built with off-the-shelf all-fiber components, allowed for even more compact, energy efficient and robust systems. Subsequently these Er: fiber OFCs have seen the most commercial success and are the most commonly used OFCs system to date.

Other notable fiber lasers include the high power 1 μm Yb: fiber, which when combined with high power Ytterbium amplifiers are ideal candidates for high-harmonic generation in the XUV for direct comb spectroscopy, as well as the realization of 2 μm Thulium doped fiber OFCs. In more recent years, bandwidth extension to the mid-IR has resulted in the exploration of non-silica based fiber lasers such as those based on Er3+:fluoride.

The use of the above solid-state and fiber-based systems, combined with continuum generation in non-silica based nonlinear fibers currently provides near-continuous and coherent spectroscopic coverage from 400 nm to ~4 μm. In many ways, frequency generation with short-pulsed laser systems enables the only means for broad spectroscopic coverage at some wavelengths with high brightness. This is particularly true in the mid-IR to the terahertz and the ultraviolet (UV) to the XUV. The more extreme demonstration of bandwidth extension has been to wavelengths as long as 27 μm using a combination of difference frequency generation (DFG) and/or optical parametric oscillation (OPO). Difference frequency generation relies on phase matching in standard nonlinear crystals to down-convert two photons of higher energy, a pump, \(v_1\), and signal, \(v_2\), to an idler mid-IR photon via \(v_3 = v_1 - v_2\). Optical parametric oscillators can perform either DFG or sum

stabilization of both \(f_r\) and \(f_0\). As will be discussed later in the text, out-of-lab applications that use OFCs to measure Doppler broadened molecular linewidths, or applications that do not benefit from perfectly controlled environments, can use OFCs with lower stability and accuracy (see “Optical frequency combs beyond the laboratory”).
frequency generation ($\nu_5 = \nu_1 + \nu_2$) in a resonant optical cavity, which permits highly efficient and extremely versatile frequency conversion. Below 400 nm, researchers borrowed techniques from pulsed tabletop X-ray sources to generate high optical harmonics by focusing cavity-enhanced optical pulses into a jet of noble gas for the production of UV and XUV frequencies. The highest achievable coherent frequency generation resulted from the 91st harmonic at 11 nm from a 60 W Yb:glass laser that can have been investigated as OFC sources including quantum photonically integrated OFC sources17,60.

Current state-of-the-art in MLL sources: After their first demonstrations, much effort in MLL OFCs design was placed on satisfying the dual goals of higher performance and lower SWAP. To keep pace with improvements in optical atomic clock development, common-mode measurement architectures and higher-bandwidth actuators have enabled long-term stability in dilution refrigerators. To finally to monolithic, high-performance, sub-100 fs fiber lasers focused into a gas jet of argon31.

Compact and chip-scale sources. The past 15 years have also seen the development of chip-scale systems based on microresonators and semiconductor systems. Also described in this section are electro-optic frequency combs, which are currently the only source with a highly agile repetition rate. The compact size of these systems yield great excitement about the possibility for chip-scale and vertically emitting semiconductor lasers integrated with semiconductor saturable absorber mirrors, which help induce modelocking. When optically pumped, MIXSELs can produce sub-100 fs pulses and greater than 1 W of optical power. In addition, the integrated semiconductor platform is a potential candidate for mass production with substantially reduced fabrication costs and high-efficiency, and can be engineered to operate from 800 nm to the near-IR. Whereas the operating wavelength of a traditional semiconductor laser is determined by the bandgap of the material, QCLs rely on sandwiched quantum-well heterostructures that behave as engineered bandgap materials. As a result, the QCL offers a versatile system based on four-wave mixing17 for the generation of mid-IR to terahertz radiation with variable mode spacing from 5 to 50 GHz. While QCL-combs do not produce optical pulses, which results in serious challenges to nonlinear broadening, stability, and regularity in mode spacing, they currently offer the only OFC platform with direct electrical pumping.

Microresonator systems: OFCs based on microresonators, or micro-combs, differ significantly in operation from MLLs because they are not lasers, but low-loss, optical resonators. The first of these systems developed as OFCs were based on suspended silica micro-toroids14, and machined and hand-polished crystalline CaF$_2$ micro-rods67. Microresonator architectures have since expanded to more easily integrated and lithographically engineered and patterned waveguides based on a multitude of materials, whose various properties are summarized in ref. 62. Micro-resonators act as build up cavities that enable high nonlinearity over long storage times, or equivalently long interaction lengths, in very much the same way as do nonlinear fibers. Via degenerate- and non-degenerate four-wave mixing, a resonantly coupled single-frequency pump source is converted to a comb of optical frequencies. Coherent optical bandwidths for self-referencing of $f_0$ have only been demonstrated directly from
resonators whose mode spacing exceeds 200 GHz\textsuperscript{68,69}. Unfortunately, the optical bandwidth narrows significantly for resonators with accessible mode spacing closer to 20 GHz. Unfortunately, this bandwidth narrowing at lower repetition rate necessitates the use of external amplification and broadening. While micro-combs enable chip-scale comb generation, they do not necessarily yield optical pulses. Because pulse formation is crucial for coherent comb formation, much of the early microresonator work was aimed at understanding the temporal dynamics of stable optical soliton production, which is now regularly realized via systematic and careful control of the pump laser detuning\textsuperscript{62,70}. 

**Electro-optic comb generators:** The equally spaced optical modes of frequency comb generators based on a phase-modulated single-frequency laser were used in precise optical metrology prior to 2000\textsuperscript{71} to bridge and measure smaller frequency gaps (<10s of terahertz) between the last multiplication stage of the frequency chain to an unknown transition of interest (see “Clock comparisons”).\textsuperscript{71} Because of their simple optical architecture and the possibility for multi-gigahertz mode spacing derived directly from a vacuum electronic synthesizer, these sources have been revisited in recent years primarily in the context of arbitrary optical waveform generation\textsuperscript{72}, high bit-rate optical communication and microwave photonics\textsuperscript{51}. Perhaps the most versatile feature of electro-optic combs (EO-combs) is the fact that they are the only OFC source that offers wide-band and agile tuning of the mode spacing. An additional benefit of these systems is the availability of high-speed electro-optic modulators that operate at pump wavelengths spanning 780 nm to 2 μm. More recently, electro-optic combs (EO-combs) have yielded access to \(f_0\) for full stabilization\textsuperscript{60}. Electro-optic comb generation using a cavity stabilized seed laser has lead to the application of an EO-comb to the calibration of an astronomical spectrograph\textsuperscript{73}, which requires long-term frequency stability at parts in \(10^{11}\) and an ultra-wide and extremely flat optical spectrum (see “Calibration of astronomical spectrographs”).

**Super-continuum generation below 200 pf:** To date, nearly all high repetition rate and compact frequency comb systems\textsuperscript{60,74,75} required high-power optical amplification to enable fiber-based continuum generation for detection of \(f_0\). This is because the combination of high repetition rate, low output power, narrow optical bandwidth, as well as no modelocking mechanism in the case of EO- or QCL-combs, results in pulse energies that are at least 100 times lower than those possible with MLLs. Continuum generation in lithographically patterned photonic waveguides and nanowires (as well as extremely small core chalcogenide fibers\textsuperscript{47}) have recently helped to mitigate these difficulties. These photonic waveguides have been to compact combs, what nonlinear fiber was to MLLs. The use of extremely small cross-sections (<1 μm × 1 μm), waveguide dispersion engineering, broad transparency windows, and a higher nonlinear index as compared to silica fiber, have permitted super-continuum generation at much lower pulse energies (<200 pJ)\textsuperscript{76}. Much like the developments that took place with nonlinear fibers, the last decade has seen the remarkable engineering of photonic waveguides for continuum generation from the visible to the mid-IR\textsuperscript{62,76–82}. Because patterned waveguides are lithographically produced, they offer more versatility than optical fibers in terms of spectral shaping, but on the downside suffer significantly higher insertion and propagation losses. 

**Current state-of-the-art in compact and chip-scale sources:** Although gains have been made in the coherence generation techniques described above, semiconductor and microresonators systems have yet to achieve their full potential as compact systems for precision optical comb generation. Optically pumped MIXSELs have been fully stabilized, with low residual noise, using extended cavities (\(f_0 < 2 \text{GHz}\)) and external amplification\textsuperscript{74}, but at the compromise of SWAP and system complexity. Of the compact sources, QCL-based OFCs currently offer the lowest SWAP due to the fact that they are electrically pumped. Despite the fact that QCLs currently do not offer access to \(f_0\) for precision operation, their low-SWAP and access from the mid-IR to terahertz have allowed these sources to emerge as a commercially available platforms for dual-comb spectrometers\textsuperscript{83}.

In microresonators, stable soliton formation has been understood and can now be regularly controlled, and broadening to an octave at mode spacing (<30 GHz) has been achieved to enable coherent detection of \(f_0\)\textsuperscript{5}. However, soliton formation in microresonator systems requires significant detuning of the pump away from the resonator modes, resulting in an optical efficiency of less than 3%\textsuperscript{85}. Consequently, the low optical output power necessitates the use of high-power external optical amplifiers and/or “helper” lasers for frequency doubling to enable \(f_0\) detection\textsuperscript{68,69,75}. These auxiliary optical components currently limit the photonic integration of micro-combs and increases their SWAP. Despite these challenges, microresonators have been applied to many of the experiments that traditionally employ MLLs, but with degradation in performance\textsuperscript{12,69,84,85}. Currently the largest obstacle in achieving low-drift and narrow optical linewidths in microresonators is thermorefractive noise, which results because microscopic systems naturally exhibit high sensitivity to both temperature and pressure. An additional blue-detuned auxiliary laser can be used to correct thermally induced changes in the material index of refraction by nearly 10 dB\textsuperscript{86}, but once again at the expense of power consumption and complexity.

Although compact sources based on microresonator and semiconductor systems still face challenges to full optical integration, to date these platforms offer the only possible architectures for chip-based and integrated comb systems. The future integration of compact OFCs with CMOS compatible photonic waveguides based on, for example lithium niobate, silicon or silicon nitride, diode lasers and miniature optical clocks might one day enable sub-watt systems for both optical\textsuperscript{69} and microwave synthesis, enabling cost-efficient production for dissemination of OFC sources and products to larger commercial markets.

**OFC applications**

In the following sections we explain how OFCs have impacted various applications and their evolution. For simplicity, we group applications into two categories based on application stability requirement. The first sections focuses on the application of OFCs to high-precision frequency synthesis and measurement, which require the most stringent requirements to enable the comparison and dissemination of signals for atomic clocks (see “Combs for atomic clock comparisons”). Section “Optical frequency combs beyond the laboratory” encompasses research that have begun to move from the laboratory toward more commercial applications. These latter applications typically require more robust and less environmentally sensitive sources, and can tolerate lower stability requirements because of their use in less controlled environments. While the work discussed here does not encompass the full application space enabled by OFCs, we limit the discussion to these topics due to the length constraints of the manuscript.

**Comb referencing, stabilization, and performance.** Because the optical spectrum from a MLL is not stable on its own, precise knowledge and control of both comb parameters is required to harness the laser’s full potential for precision metrology. The measurement of any frequency, optical or microwave, requires
comparison against a second frequency reference. Here we explore how the chosen references and how the frequency (microwave or optical) at which a comb is referenced impacts the stability and noise of its modes. Knowledge and control of the comb modes can be achieved by comparing \( f_r \) and \( f_0 \) directly to microwave references, by constraining the two degrees of freedom in the optical domain to optical references, or via a combination of the two methods. While OFCs can be referenced in either the optical or microwave domain, stabilization of an optical mode of the OFC to an optical reference is achieved by comparing and/or locking the small difference frequency, \( \Delta f = f_r - f_0 \), to a microwave reference. Aside from full phase stabilization, passive optical and electronic phase noise removal from the comb parameters can be used for frequency measurement using of an OFC. These methods include DFG for the creation of optical combs with vanishing \( f_0 \), and synchronous electronic “mix-out” of \( f_0 \) and \( f_r \) using what is called the transfer oscillator method. Full frequency stabilization uses negative feedback to the laser cavity length and intra-cavity dispersion to actively and physically control the values of \( f_r \) and \( f_0 \) via comparison to chosen frequency references. These are most typically achieved using piezoelectric actuators for length control and modulation of the laser pump power and intra-cavity gain, which actuates on the laser intracavity dispersion. In contrast, the transfer oscillator method loosely controls the drift of \( f_0 \) and \( f_r \) so that their electronic signals can be precisely tracked to correct their noise excursion across the microwave and optical modes of the comb.

**Passive \( f_0 \) removal via DFG.** 
DFG can be used to create a frequency comb with vanishing \( f_0 \) information. The result is an "offset-free" comb with optical modes that are described by multiples of \( f_r \) only, whereby \( \nu_k = \nu_N - v_M = (N - M) \cdot f_r + f_0 = N \cdot f_r. \) This passive removal of \( f_0 \) eliminates the need for its measurement and referencing (while OPOs can synthesize idler frequencies based on DFG, \( f_0 \) cancellation is not necessarily a given because the signal is created via nonlinear conversion from the pump in the OPO). Creating optical combs via DFG requires \( \nu_M \) and \( \nu_K \) to be separated by large difference frequencies. For example, a MLL centered at 1550 nm can produce an "offset-free" comb at 3 μm, but requires an octave of bandwidth (~1000 nm). Additionally, the low-efficiency and phase matching requirements of the nonlinear crystal results in low-power and narrow bandwidth DFG combs. As a result, the DFG comb often requires both optical amplification and nonlinear broadening for power and bandwidth recovery.

Figure 3 shows how the OFC noise performance varies as a function of different referencing schemes. For readers that are more familiar with noise analysis, the arguments here are constrained to systems where the noise does not significantly overwhelm the optical carrier. The stabilization schemes depicted by the blue and red traces can be used to constrain \( f_0 \) for a system where its detection is not accessible. The performance in black and green traces are only possible with combs whereby \( f_0 \) is directly accessible, or for "offset-free" combs. In the red trace in Fig. 3 we see that the noise equivalent linewidth, \( \delta \nu \), of the comb modes is constrained between the two lock points (for \( N \leq K \leq M \)), but diverges outside the lock points (for \( K < N \) and \( K > N \)) with a slope \( \delta \nu/\delta K \) given by the noise of the references (added in quadrature), divided by their spacing.

\[
\delta \nu/\delta K = \sqrt{\delta \nu_{\text{ref1}}^2 + \delta \nu_{\text{ref2}}^2}/\sqrt{(M - N)}. \tag{5}
\]

Equation (5) indicates that the further apart the lock points, the better the frequency leverage to constrain all the comb modes. As a result, the self-referenced stabilization method (black trace) exhibits the highest performance by having lock points separated by 100s of terahertz, with \( f_r \) constrained via stabilization of a mode in the optical domain, and by direct stabilization of \( f_0 \) to a microwave reference near 0.

The self-referenced locking scheme also enables optical frequency division (OFD), whereby both the frequency and noise of the optical reference are divided down to the microwave domain. In this scheme, the divided optical signal is detected via the repetition rate, which carries the noise of the optical reference at mode \( M \) divided by \( M^2 \), \( \delta f_r = \delta f_r/\sqrt{M^2}. \) To put this division into context, an optical reference at 300 THz divided down and detected on a repetition frequency of 10 GHz, would
yield a reduction of the optical phase noise to the microwave domain by 90 dB (≈ 10 · Log(300 THz/10 GHz)), or nine orders of magnitude. The green trace in Fig. 3, which also exhibits a linear dependence between noise and mode number, uses frequency multiplication to transfer a microwave reference to the optical domain. In both schemes the phase noise power spectral density (PSD) of mode number \( k \), \( S_{\phi, k} \) is equal to the phase noise PSD of the reference, \( S_{\phi, \text{ref}} \) (at mode \( M \)), scaled by the square of the ratio of the mode numbers, or
\[
S_{\phi, k} \sim S_{\phi, \text{ref}} \left( \frac{K^2}{M^2} \right). \tag{6}
\]
Because both the frequency and noise scale equally in multiplication and division, use of either the green or black traces for stabilization yields preservation of the fractional frequency stability of the reference in its transfer to all comb modes, or
\[
\sigma_f = \frac{\delta \nu_{\text{ref}}}{\nu_{\text{ref}}} = \frac{\delta \nu_{K}}{\nu_{K}}. \tag{7}
\]

**Combs for atomic clock comparisons.** The full characterization of the optical spectrum of the MLL in the year 2000 allowed for frequency multiplier chains to be replaced by a single MLL. As will be discussed in more detail in the following section, this increased measurement capability was marked by the rapid characterization of unknown optical clock transition frequencies. The large bandwidth of OFCs also allowed for the relative measurement of different species of developing optical atomic clocks, enabling frequency comparisons below the limit imposed by the \(^{133}\text{Cs}\) primary reference.

**Clock comparisons: Absolute frequency measurement.** As mentioned briefly in the introduction, optical atomic clocks were developed because their higher transition frequencies enabled better frequency/time resolution than their microwave counterparts. Generally, as described in Eq. (7), the resolution of an oscillator is defined as the uncertainty with which the frequency can be defined, \( \delta \nu \), scaled by its center frequency, \( \nu \). More specifically, the frequency uncertainty of atomic clocks is limited by the clock transition sensitivity to external environmental fields, as well as how well these fields can be controlled and/or measured. Because the measurement and control of the latter physical parameters (temperature, E- and B-fields, optical power, and background pressure) is roughly the same for microwave and optical clocks, the benefit of the high transition frequency in optical clocks is two-fold; one can achieve both higher resolution with less stringent environmental control. To put this into perspective, to achieve \( 10^{-16} \) fractional stability on a 500 THz optical clock requires control of the optical transition frequency at the 50 mHz level. Conversely, the same fractional stability on a 10 GHz signal requires control of a microwave transition frequency at the 1 mHz level. The upshot is that optical atomic clocks still have significant room for improvement, both in terms of SWAP and performance, whereas the performance of microwave clocks has plateaued in recent years.

As seen in Fig. 4, frequency combs and comb generators measure frequency in much the same way that rulers measure distance. They work by generating optical frequencies on an equidistant grid, with a known grid spacing. An unknown optical frequency, \( \nu_{\text{opt}} \), is determined by measuring the difference frequency, \( \Delta f \), between it and the nearest comb mode. Prior to the year 2000, frequency comb generators were used to bridge smaller gaps in frequency. This enabled a significant simplification to multiplier chains that required additional oscillators to frequency shift various multiplication stages such that the last stage fell within 50 GHz of the transition frequency of interest (limited by photodetector speeds). Although these comb generators could not measure \( f_0 \) directly, \( f_0 \) could be constrained by locking an optical mode to the output from the last multiplication stage of a frequency chain, and by subsequent stabilization of the mode spacing directly to a microwave reference, see Fig. 4. This stabilization scheme yields a performance similar to the blue trace in Fig. 3 with the exception that the optical output from the frequency chain had a noise at least 100 times higher than that of \( \nu_{\text{opt}} \) in Fig. 3.

Where comb generators required referencing in the optical domain, OFCs, which yield direct access to \( f_0 \), allow for direct referencing and connection to the microwave domain. As such, an optical transition, \( \nu_{\text{opt}} \), measured with an OFC can be expressed entirely in terms of microwave frequencies via the comb equation such that
\[
\nu_{\text{opt}} = \nu_N + \Delta f = N \cdot f_{\text{rep}} + f_0 + \Delta f. \tag{8}
\]
To use Eq. (8), the value of \( N \) can be determined using a wavemeter to loosely measure the unknown optical frequency, such that \( N = \text{Nearest Integer} \left[ \nu_{\text{opt}}(\text{wavemeter})/f_0 \right] \). Wavemeters can yield frequency resolutions of parts in 10\(^8\), or approximately 50 MHz on an optical frequency. To ensure there is no ambiguity in the mode number, OFCs with a repetition rate of >150 MHz are typical used in atomic clock measurements.

**Optical clock comparisons.** The measurement of absolute optical frequencies is naturally limited by the lower stability microwave references that measure them. Otherwise said, the measurement of an optical clock in terms of the Hertz is limited by the current definition of the Hertz at parts in \( 10^{16} \). This stability limitation has the additional drawback that averaging periods of up to a month are required to reach the maximum fractional accuracy near 1 part in \( 10^{16} \) of the microwave references. To circumvent this limitation optical clocks can achieve relative measurements against one another via optical synthesis with an OFC. Because optical clocks have short-term resolutions 10 to 100 times higher than their microwave counterparts, a relative uncertainty of \( 10^{-16} \) can be achieved in a matter of seconds.

These relative clock comparisons are generally reported in the form of frequency ratios, \( R = \nu_{\text{opt1}}/\nu_{\text{opt2}} \), which have both practical and fundamental applications to clock comparisons. Via a quick ratio measurement (See Fig. 4), the absolute measurement of an unknown optical clock, \( \nu_{\text{opt2}}(\text{Cs}) \), can be measured against a known one, \( \nu_{\text{opt1}}(\text{Cs}) \) as follows:
\[
\nu_{\text{opt2}}(\text{Cs}) = R_{2,1} \cdot \nu_{\text{opt1}}(\text{Cs}). \tag{9}
\]
Here \( \nu_{\text{opt1}}(\text{Cs}) \) and \( \nu_{\text{opt2}}(\text{Cs}) \) are fractionally limited by the definition of the Hertz, but because the ratio is unit-less, \( R_{2,1} \) is independent of this \( 10^{-16} \) limit. As a result, frequency ratio uncertainties are limited by the optical transition frequencies alone. Due to the fact that optical clock transition frequencies can be controlled at levels near parts in \( 10^{-16} \), frequency ratios permit the highest precision physical measurements to date. Additionally, because the atomic transition frequency and its dynamics are governed by the physical laws and the universal fundamental constants, atomic clock comparisons enable platforms for exquisite tests of fundamental physics. For instance, by looking for variations in the historical time records of clock ratios, ratio measurements have placed the highest constraints on violations of special relativity and searches into time variation of fundamental constants. Atomic clock comparisons have also been used to search for ultralight dark matter particles that are theorized to induce time-dependent changes in the value of the fine structure constant, and hence in the transition frequencies of some clock species. Beyond tests of fundamental physics, atomic clocks currently exhibit sensitivity to...
Fig. 4 How frequency chains and frequency combs measure an unknown optical frequency. Frequency generators bridge frequency gaps by counting the number of equidistant modes between the last multiplication state of the frequency chain and the optical transition of interest. $v_{\text{opt}}$. Because the offset frequency of these systems is unknown, $f_0$ is constrained by locking a single mode of the comb generator to the output stage of the frequency chain. The measurement is limited by the stability of the $^{133}\text{Cs}$ reference. An optical frequency comb (OFC) eliminates the need for a frequency chain by directly connecting optical frequencies to the microwave domain via access to $^{133}\text{Cs}$, that once against limits the measurement stability. Finally, an OFC compares two optical clocks by measuring the difference frequencies, $\Delta f$ and $\Delta f_0$, with regards to their nearest OFC modes, $N$ and $M$, respectively. Optical comparisons can be determined via the mode numbers and difference frequencies alone. For this reason, optical comparisons yield amazing precision because microwave reference errors on $\Delta f$ and $\Delta f_0$ are additive to the optical frequency. A $10^{-12}$ fractional error on $\Delta f = 100$ MHz, only yields a frequency error of 100 μHz. This error on a 500 THz optical carrier only contributes fractionally at 2 parts in $10^{19}$. Put differently, using a ruler one can split a centimeter marker by about a factor of 20, while difference frequency measurement can split 1 GHz-spaced OFC optical modes by parts in $10^{14}$.

Timing, synchronization, and atomic clock networks. Many of the applications mentioned in the previous section could be extended and benefit from global networking of optical atomic clocks. For instance, contrasting the values of published atomic clock ratios from different national metrology laboratories currently represents the only means by which clock networks on separate continents can be compared. Additionally, the primary motivator for the development of optical clock networks is the realization of an optical SI second. However, this realization will require the networking of an international ensemble of atomic clocks for a distributed and democratic realization of optical-universal coordinated time. While microwave clocks are currently linked internationally by the Global Positioning Systems (GPS) and 2-way satellite time transfer, the stability of these systems can only achieve a transfer stability of $10^{-16}$ after one month of averaging, which is insufficient to support the timing capabilities of the best atomic clocks.

Aside from clock-based applications, the dissemination of ultra-high-stability timing signals is also a necessity in the context of large-scale science facilities for remote synchronization of physical events and data collection in high-resolution measurements. As a consequence, optical fiber frequency transfer was developed to facilitate both higher stability optical clock comparisons and higher resolution timing distribution and synchronization. In the context of free-electron laser facilities, the distribution of timing signals from OFCs has enabled all-optical synchronization of remotely located lasers, accelerators, RF electronics and ultrafast X-ray experiments to within 30-fs facility-wide. Additionally, over the past 15 years, fiber networks in Europe have been expanded to enable inter-city and inter-continental clock comparisons of primary frequency standards at parts in $10^{16}$ and optical clocks at the mid-$10^{-17}$ level.

Extension of clock comparisons over long-haul fibers as a means to connect clocks from North America and Asia to Europe is unlikely due to the cost and technical difficulty to upgrade the existing infrastructure. Two-way optical time/frequency transfer (TWOTFT) across open-air paths using OFCs has been under development since 2013. These optical links allow for the comparison and synchronization of spatially separated clocks at locations where fiber networks are not readily available. Time/frequency communication is achieved by comparing counter-propagating optical pulse trains from OFCs stabilized to clocks at remote locations. Linear-optical sampling (LOS), whereby each local pulse train at one site samples the incoming pulse train from the other remote site, produces interferograms that yield the relative OFC timing and frequency information (see also Fig. 5). This permits measurement of the OFC pulse arrival times from the remote site with femtosecond-level resolution using megahertz bandwidth electronics. The arrival times from both sites are used in the two-way transfer to either discern differences in clock rates, or optical path lengths fluctuations caused by platform motion and air turbulence. Because the optical path length fluctuations are reciprocal, and thus
equally sampled by the counter-propagating OFC pulse trains, comparison of optical pulse arrival times from the two sites can be used to obtain the clock rate difference independent of turbulence. Consequently, TWOTFT enables the measurement of clock ratios, the frequency equalization of two clocks (syntonization), as well as the absolute elimination of time offset between two clocks (synchronization)\textsuperscript{115} with minimal residual time/frequency errors.

For example, TWOTFT links have been implemented up to several kilometers and have demonstrated timing errors as low as 7 fs after 1 s of averaging\textsuperscript{116}. While all OFC-based TWOTFT demonstrations to date used kilometer-scale terrestrial links, it has been inferred\textsuperscript{117} that a TWOTFT ground-to-satellite link should only increase the turbulence-induced timing noise to a few femtoseconds. Finally, the low duty cycle of the optical pulses make the link significantly less sensitive to interruptions (for

Fig. 5 Dual-comb ranging and linear-optical sampling. In direct time of flight ranging with a single comb, the range is determined by measuring the delay between pulses traveling to a known reference (green pulses) and a target (red pulses). Ideally, the distance resolution, $\delta L$, should be limited by the width of the optical pulses, $T_p$. However, when the optical pulses are detected directly, it is the much slower response time of the photodetector, $T_{\text{resp}} \gg T_p$, that limits the distance/timing resolution. By employing a second comb, linear-optical sampling (LOS) is used to circumvent the photodetector response limit. LOS creates an interferogram between two OFCs with slightly offset repetition rates, $\Delta f_r$. This optical cross-correlation between the blue and red pulses yields information about the relative optical pulse envelopes, $T_p$, and the optical carriers (interferometric fringes), $\delta T'$, accessible with standard microwave electronics and megahertz sampling rates. This interferometric LOS improves the coarse distance resolution of direct time-of-flight measurements from the millimeter- to nanometer scale. Linear-optical sampling also enables the measurement of high-resolution time/frequency information. In two-way optical time/frequency transfer, interferograms from LOS are collected at remote sites to compare, syntonize, and synchronize remote clocks, yielding time/frequency measurement accuracy and precision of parts $10^{-17}$.
times less than $1/f_p$, usually a few nanoseconds) than those based on continuous wave signals.

**Ultra-low-noise microwave generation.** As discussed in section "Comb referencing, stabilization and performance", OFCs stabilized to high-stability optical references can enable the derivation of microwave signals with stabilities better than $10^{-13}$, yielding greater than a 100 times improvement over what can be achieved with the best room-temperature electronic oscillators. In principle, the generation of low-noise microwave is relatively straightforward. As shown in Fig. 1, the phase-stabilized optical pulses from an OFC can be converted to a stable electronic pulse train via direct detection with high-speed photodetectors. Subsequent electronic filtering isolates a single harmonic of $f_e$, within the bandwidth of the photodetector producing a sinusoidal signal. Signals derived in this manner can be utilized as microwave frequency representation of optical atomic clocks ("ticks") to facilitate their characterization against primary microwave standard and for the generation of optically-derived microwave timing signals.

An additional benefit of OFD, as described in Eq. (6), is that OFD also divides the noise of the optical reference to the microwave domain, yielding high-spectral purity electronic signals. For example, OFD of high-stability optical cavities have been used to generate 10 GHz signals with ultra-low phase noise of $10^{-15}$ Hz (the spectral noise sidebands falls 10 orders of magnitude when stepping 1 Hz away from the microwave carrier). Ultra-low noise electronic signals have natural applications to military, communications, and test and measurement applications. For example, in X-band RADAR signals with low close-to-carrier noise can facilitate the detection of slow moving objects (with Doppler shifts below 1 kHz), and the measurement of weak return signals from low-cross section objects. Additionally, low noise on local oscillators can enable shorter averaging periods for quicker acquisition of targets and frequency-hopped communications signals.

To put the potential for low-noise into perspective, self-referenced OFCs can produce optical pulses with $>200$ dB of optical dynamic range and sub-femtosecond pulse-to-pulse timing jitter. Ultimately, the biggest challenge in low-noise microwave generation is preservation of these exquisite optical pulse characteristics in their conversion to the microwave domain. Aside from the massive pulse distortion that results due to limited detector response time and power handling, the detection of high-energy optical pulses further exacerbate detector nonlinearities by creating short bursts of extremely high-density photo-carriers in the detector active region. Normally, this results in charge-screening of the applied bias voltage, which reduces detector speed and yields strong conversion of optical amplitude noise to phase noise on the microwave pulse train. Consequently, to attain the highest-spectral purity microwave signals, low-pulse energy, higher-repetition rate OFC sources ($f_e > 1$ GHz) are ideal. The photodetector distortion and saturation effects can be mitigated with heterostructures designed to improve charge transport speeds, and ones that pre-distort the internal potential for high-linearity at high-photocurrents. Impressively, such photodetectors have demonstrated preservation of the optical pulse-to-pulse timing at parts in $10^{17}$ and have enabled microwave signal dynamic ranges as high as $180$ dB. Finally, a significant side benefit of ultra-short optical pulses (<1 ps) is that coherence of the optical modes results in modified shot noise statistics, or cyclo-stationary noise, which permits microwave phase noise floors significantly lower than what is possible with CW lasers.

**Calibration of astronomical spectrographs.** OFCs were first proposed for improved frequency calibration of astronomical spectrographs in 2007. These spectrographs measure Doppler shifts of stellar spectra to determine the radial velocities of celestial bodies, as well as the composition of solar atmospheres. Higher precision measurements that aim to discern cm/s level shifts in these Doppler shifts, can be used to assess the rate of expansion of the universe, as well as detection of the periodic wobble in stellar velocity due to the influence of an orbiting exo-planet. These small Doppler drifts are inaccessible with conventionally calibrated spectrographs due to environmental instability, and are limited by the spectral coverage and stability of traditional calibration sources (wavelength references). Observation of the equidistance of the OFC modes at the spectrograph’s imaging plane allows for calibration of instrument drift, improving its long-term frequency accuracy.

The application specifications require that these metro-OFCs overcome some technical challenges, namely high-mode spacing near 30 GHz for matching to the spectrographs resolving power, as well as very broad and spectrally flat coverage ($\Delta f < 3$ dB) from 380 nm to 2.4 μm for uniform illumination of the spectrograph, and frequency stability at a level of $3 \times 10^{-11}$ corresponding to 1 cm/s radial Doppler drift. As such, different architectures have been developed using mode-filtered fiber, and Ti:sapphire-based OFCs, and more recently electro-optic comb generators. OFCs began deployment to spectrographs in 2008, and can be currently found in at least seven telescopes around the world. Of these, the high-precision OFC calibrations were performed to 2.5 cm/s at HARPS/FOCES and to <10 cm/s at HARPS-N and the Hobby–Eberly Telescope.

**Distance measurements and laser ranging.** The application of optical frequency combs to LIDAR (light detection and ranging) was first demonstrated in 2000 and enables a number of advantages over traditional sources. As seen in Fig. 5, two techniques can be used to discern macroscopic distances: the more coarse direct time-of-flight measurement, and the more fine linear optical sampling (LOS) measurement. As seen in Fig. 5, the maximum distance resolution in direct time of flight measurements is limited by the response time of the photodetector, which is much slower than the optical pulse envelope width, or $T_{\text{resp}} \ll T_p$. Ranging measurements, enabled by dual-comb techniques (see also section “Direct comb-based spectroscopy”) or balanced optical cross-correlation in nonlinear crystals, can bypass the limitations of $T_{\text{resp}}$ via optical down-sampling so that the optical carrier can be detected on low-speed photodetectors (Fig. 5). This permits distance measurements with resolutions limited by the width of the convolution of the two OFC pulse envelopes, $T_p < 100$ s, significantly shorter than the ~1 ns pulses from traditional LIDAR systems. These gains in resolution and precision can be further improved by detecting the optical carrier under the pulse envelope. Interferometric measurements of this kind have demonstrated 10 nm resolution for averaging times of ~60 ms, and nanometer-level precision with longer averaging over second timescales. Finally, the combination of short pulses, and a phase-stabilized optical spectrum has allowed for demonstrations of comb-based distance measurements at kilohertz measurement rates and sub-micrometer ranging precision, while simultaneously enabling unambiguous range measurement to targets at meter to kilometer distances.

The largest drawback of using OFCs directly for ranging is their high cost and high system complexity. Consequently, recent work has pushed toward the development of smaller and simpler systems based on electro-optic and microresonator comb
Yielding femtosecond optical fringe information to megahertz bandwidths that can be processed with off-the-shelf electronics. Fourier transform of the optical interferogram yields a microwave comb RF spectrum with mode spacing.

The signal repeats with a periodicity of 1/Δf_r. All spectral information from the optical domain is mapped to an RF bandwidth ≤ Δf_r/2, corresponding to an optical bandwidth of f_r/Δf_r. Spectral filtering is usually applied to ensure non-ambiguous detection over this spectral range.

As a result, spectroscopic measurement with OFCs has been explored over vast portions of the electro-magnetic spectrum, extending from the terahertz to the UV. A concerted effort has also been devoted on photonically integrated circuits as well as on photonic phased-array (VIPA) systems, similar to that in dual-comb spectroscopy (DCS). Direct comb-based spectroscopy, is that the low-power per optical mode requires a retro-reflecting mirror for sufficient return light powers at distances beyond ~1 m.

Direct comb-based spectroscopy. Frequency combs are attractive sources for spectroscopy because they offer: (1) broad spectral bandwidth, good for detection of multiple molecular species, (2) high spatial coherence, which allows for longer interrogation paths and higher sensitivity, and (3) high frequency resolution and accuracy when measured on a mode by mode basis. As a result, spectroscopic measurement with OFCs has been explored over the terahertz to the UV. A concerted effort has also been devoted toward coverage in the mid-IR region, to access stronger molecular cross-sections, and OFC-based spectroscopy has also been reported on extensively in various review articles (see for example). More specifically, direct spectroscopy with OFCs has been applied to study ultra-cold molecules, human breath analysis, time-resolved spectroscopy, and high-precision molecular spectroscopy.

While multiple comb-based measurement techniques have been explored for molecular spectroscopy, such as comb-assisted Fourier transform infrared spectroscopy (FTIR) and virtually imaged-phased-array (VIPA), the most notable has been DCS. Dual-comb spectroscopy, which was first demonstrated in 2004, uses LOS of two combs with slightly offset repetition rates to down-convert 10s of terahertz of optical bandwidth to 100s of megahertz in the RF domain. This detection scheme preserves the stability and accuracy of the comb, as well as the amplitude and phase information encoded on the individual modes of the comb by the spectroscopic sample. Consequently, the detected RF spectrum can be used to reconstruct the optical molecular absorption spectra.

Direct molecular spectroscopy with DCS enables significant benefits as compared to FTIR. Perhaps the biggest advantage is that DCS does not require any moving parts, thus enabling much faster acquisition times. Stabilization of the OFC spectrum also permits much higher resolutions, as well as SI-traceability for higher accuracy. Finally, because there are no complicated diffraction and imaging optics, measurements do not suffer instrument lineshapes. While there are many benefits to DCS, challenges arise for deployment to the field. The low power per optical mode requires a retro-reflecting mirror to obtain strong enough signals strengths, and spectral magnitude fluctuations pose challenges, especially when interrogating broad molecular transitions, or while probing a cluttered molecular spectrum over the atmosphere.

Perspective and future outlook

While precision measurements may seem like a boutique application primarily reserved for metrology laboratories, this is the area where combs have seen the most commercial success, in part because large-scale laboratory experiments are willing to pay the high price for performance. The adoption of OFC products for replacement of traditional LIDAR and FTIR systems, however,
has yet to find commercial traction. Combs, which offer better precision, resolution and faster acquisition times for both applications, yield a cost and complexity that is still too high to outweigh its benefits. As a result, this cost-benefit trade-off has helped to fuel the development of compact and lower SWAP spectroscopic sources\(^1\)\(^{6,85,83,84,148–150}\).

In precision metrology, the replacement of frequency multiplier chains by OFCs enabled a vast simplification to precision optical measurement, helping the development of new clock species, as well as facilitating continued improvement in optical clock accuracy. In the past 20 years, optical atomic clock accuracies have improved by five orders of magnitude, and now demonstrate performance 100 times better than state-of-the-art primary \(^{133}\text{Cs}\) microwave standards. This rapid progress has prompted discussions about, and the creation of a road-map toward the redefinition of the SI second to optical atomic time\(^1\)^{31}\(^\text{,}\). In the context of future optical timescales, OFCs will provide the clockwork for the derivation and dissemination of highly precise and accurate microwave and optical timing signals.

In the near future, OFCs integrated with transportable optical clocks will be important for supporting optical redefinition of the second by facilitating clock comparisons between remotely located metrology labs\(^1\)^{32}\(^\text{.}\) Aside from global clock comparisons, comb-based inter-continental time/frequency transfer could help to enable high-precision mapping of the geoid, and help to facilitate tests of fundamental physics by leveraging long baselines. To date, while high-precision clock comparisons have yet to uncover dark matter signatures or observe temporal variations of fundamental constants, these tests have been used to verify our understanding of current physical models at parts in \(10^{17}\)\(^\text{.}\) In the future, ranging and time transfer with space-borne combs\(^1\)^{33,154}\(^\text{,}\) might be used for ground-to-satellite clock synchronization for improved timing in GNSS and communications systems, and sensing with combs could be used for atmospheric spectroscopy in broadband occultation. Finally, tests of fundamental physics beyond parts in \(10^{19}\) might one day be enabled by space-borne systems benefiting from operation in a low-vibration environment and outside the earth’s gravitational potential, as well as from non-classical statistics\(^1\)^{35}\(^\text{ enabled by quantum frequency combs\(^1\)^{156}\).}

Data availability

No data set were generated or analyzed during the current study.

Received: 4 April 2019; Accepted: 9 October 2019; Published online: 06 December 2019

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Acknowledgements
The authors acknowledge fruitful discussion with William Loh, Fabrizio Giorgetta, Gabe Ycas, Ian Coddington, William Swann, and Florian Adler. We would also like to thank Fabrizio Giorgetta, Holly Leopardi, Megan Kelleher, Nick Nardelli, Frank Quinlan, Tanja Cuk, Ladan Arissian, and Henry Timmers for thorough reading of the manuscript.

Author contributions
T.M.F. and E.B. equally contributed to the ideas, organization and writing of this review article.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information is available for this paper at https://doi.org/10.1038/s42005-019-0249-y.

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