The evolving optical frequency comb [Invited]

Scott A. Diddams

National Institute of Standards and Technology, Time and Frequency Division, 325 Broadway, Boulder, Colorado 80305, USA (sdiddams@boulder.nist.gov)

Received July 13, 2010; revised September 11, 2010; accepted September 17, 2010; posted September 22, 2010 (Doc. ID 131588); published October 22, 2010

In the past decade we have witnessed remarkable advances associated with the frequency stabilization of the comb present in the output of a mode-locked femtosecond laser. While proving itself to be fantastically successful in its role as the "gears" of optical atomic clocks, the optical frequency comb has further evolved into a valuable tool for a wide range of applications, including ultraviolet and infrared spectroscopy, frequency synthesis, optical and microwave waveform generation, astronomical spectrograph calibration, and attosecond pulse generation, to name a few. In this review, I will trace several of these developments while attempting to offer perspective on the challenges and opportunities for frequency combs that might lie ahead in the next decade.

OCIS codes: 320.7090, 140.4050, 120.3940.

1. INTRODUCTION: THE MODE-LOCKED LASER—A FREQUENCY COMB LOOKING FOR STABILIZATION

Much as natural historians search for the first roots of our human race, curiosity drives the laser scientist to pursue the roots of his or her own field. This is especially the case in this year where our community celebrates the 50th anniversary of the demonstration of the first laser. If the experiments of Maiman [1], Javan [2], and other early laser pioneers represent the spark of energy that brought the field of laser physics to life, then the seed for the frequency comb was planted in 1964 with the introduction of the active mode-locked laser [3]. In fact, one could already recognize the distinct comb-like frequency structure in the optical spectra of that early helium-neon laser. Developments continued with passively Q-switched mode-locked [4] and continuous wave (cw) mode-locked [5] sources over the intervening decade, and the frequency comb began to sprout in the experiments of Hänsch in the late 1970's [6]. However, it did not assume its presently recognized form until nearly 20 years later.

Tracing this development from the very first laser sources is a fascinating story, but it is not the goal of this article to retell the history of the birth of the frequency comb. That history has already been wonderfully recounted in several reviews [7,8] and the Nobel lectures of Hänsch [9] and Hall [10]. Rather, in this article we will focus our attention on the rapidly evolving developments of the past decade—the story of the numerous applications that have arisen (sometimes unexpectedly) and formed new branches—indeed new areas of research in some cases. From one perspective, the rapid development of the frequency comb and its applications over the past ten years is captured in the "evolutionary tree" of Fig. 1.

With some hindsight, the branching tree helps visualize the progression of the development of the field; however, our experiences in the laboratory teach us that research and technological development rarely proceed in a purely linear manner. In reality, many of the branches of this evolutionary tree are interconnected—both among those drawn and among the many other associated topics in optical physics that are not drawn. Nonetheless, the main point of this figure is to highlight that, in just the past decade, several new avenues of research have been opened by the introduction of the optical frequency comb. In this article, a summary of advances in several of these areas will be presented.

If the mode-locked femtosecond laser is the root of this history, as shown in Fig. 1, then what is it that transforms a mode-locked femtosecond laser into a frequency comb? There are hundreds of demonstrations of different femtosecond lasers, but only in the past decade have some of these lasers been classified as frequency combs. It has been well established that the time-averaged frequency-domain output of a mode-locked laser is described by the simple formula $\nu_n = n \cdot f_r + f_o$, where f_r refers to the repetition rate of the mode-locked laser, f_o is the carrier-envelope offset frequency, and n is the integral mode index. If this description applies to all mode-locked lasers, then it is essentially a functional definition to state that a frequency comb is the optical output of a mode-locked laser for which the parameters ν_n, n, f_r, and f_o are measured and controlled. While this may seem like an insignificant distinction, for the various applications shown in Fig. 1, the apparently "simple" act of measuring and controlling the vast number of frequency comb teeth lends tremendous value to existing applications and opens the possibility of several new applications of the mode-locked femtosecond laser.

For many years the repetition rate of a femtosecond laser had been measured and controlled, but it was only in 1999 that the sources and nonlinear fiber optics progressed to the point where optical spectra exceeding one octave could be readily generated with unamplified mode-locked laser oscillators [11,12]. This was a key requirement for the measurement and control of f_0 [13], and a defining moment in the development of the optical frequency comb, as it permitted the straightforward con-

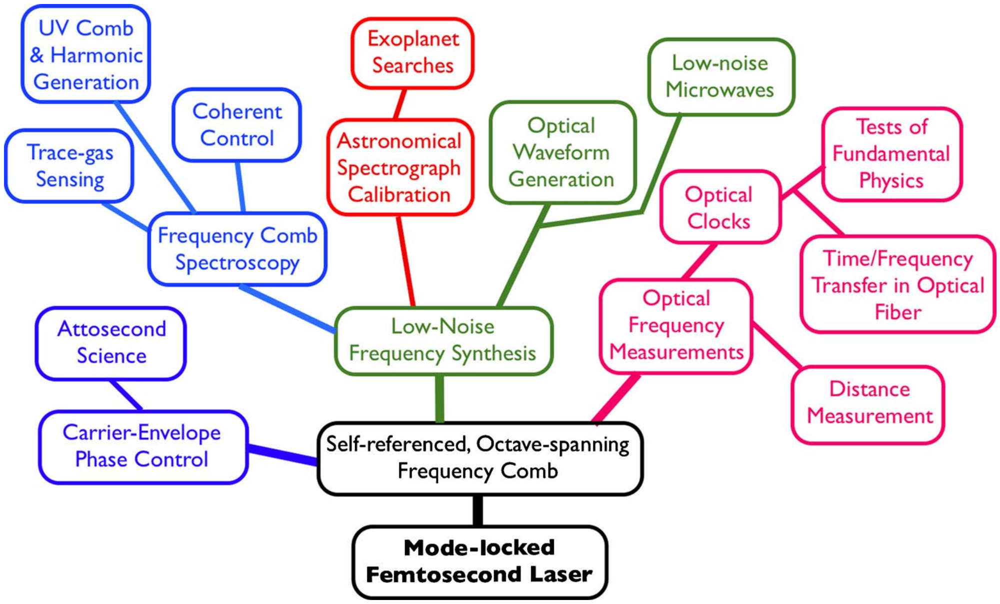

Fig. 1. (Color online) Frequency comb evolutionary tree.

nection between microwave and optical domains. Figure 2 graphically depicts the octave-spanning frequency comb and illustrates approaches to its stabilization. In nearly all cases, f_o is measured by frequency-doubling an infrared portion of the octave-spanning spectrum and heterodyning it with the existing visible comb elements [14]. Measuring f_o with narrower spectra is also possible, but with the requirement of a higher-order nonlinearity, e.g., third harmonic versus second harmonic [13,15]. The physical origin of f_o is the difference between group and

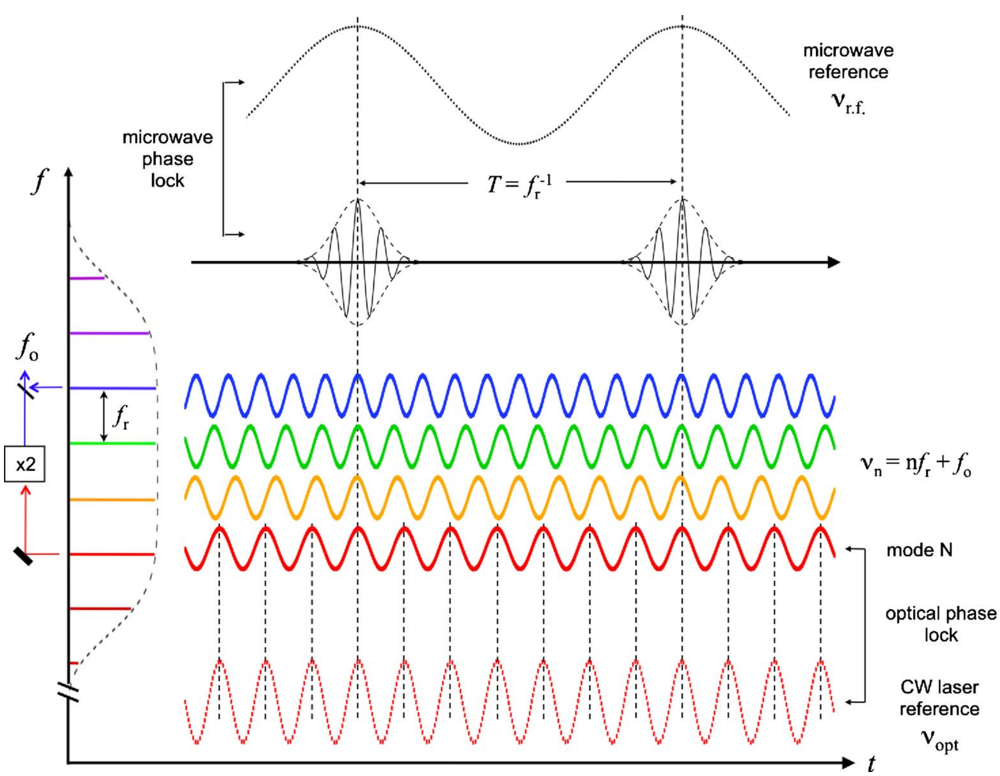

Fig. 2. (Color online) Time-frequency interpretation of the output of the mode-locked laser. The frequency modes $\nu_n = n \cdot f_r + f_o$ of the mode-locked laser are essentially a series of cw oscillators tightly locked in phase that coherently add to form a short pulse at a period T, which is the inverse of the repetition rate. The carrier-envelope offset frequency f_o is measured via the heterodyne between the second harmonic ($\times 2$) of the infrared and visible comb components. For simplicity, f_o is shown with the value of zero, such that every pulse is identical. The repetition rate (mode spacing) f_r can be measured with a fast photodiode and compared or phase-locked to a microwave reference $\nu_{r.f.}$. Alternatively, mode N of the comb can be heterodyned with an external cw laser at frequency ν_{opt}. Active feedback to the frequency comb can force the comb mode to oscillate in phase with the cw laser, and as the other modes are already locked in phase via the mode-locking mechanism, f_r assumes the value of $f_r = (\nu_{opt} + f_o)/N$.

phase velocities inside the laser cavity, and the time-domain manifestation is a pulse-to-pulse slippage between the carrier phase and the pulse envelope. As discussed by Chang and Corkum in this issue [16] the electric field phase relative to the pulse envelope is an important parameter in high-order nonlinear optics and schemes for the generation of isolated attosecond pulses [17]. For such experiments, the measurement and control of f_0 is a prerequisite.

Figure 2 also illustrates two approaches to measuring and controlling f_r. As already noted, f_r can be measured relative to a microwave oscillator; or, as also depicted in Fig. 2, f_r can be effectively measured and controlled via a heterodyne beat between one tooth of the optical comb and a stable continuous wave (cw) laser. In the former case, one is essentially multiplying up the microwave reference to the optical domain, while in the latter case the frequency comb acts to divide down from optical to the microwave output. As will be discussed, this latter approach can take advantage of the exceedingly low noise of the best cavity-stabilized lasers, which have fractional stability exceeding that available from common radio frequency or microwave oscillators.

The preceding paragraphs have described in general terms the means to measure the parameters associated with the frequency comb from a femtosecond laser, but little has been said about the techniques employed to build the phase-locks shown in Fig. 2 that enable control of the parameters ν_N, f_r, and f_0. A thorough discussion of the frequency control aspect is beyond the scope of this article, but suffice it to say that, in general, one needs two actuators inside the laser cavity to control two of these three parameters. The most common approaches typically involve a piezo-mounted cavity mirror to control f_r and some means to modulate the intracavity power to control f_0. Further details and approaches to optimum control can be found in many of the references.

2. EVOLVING FREQUENCY COMB SOURCES

During the past decade, the increased interest in optical frequency combs has resulted in the development of new femtosecond laser sources, as well as the revisiting of existing sources with a focus on the frequency domain properties. While comb structure was recognized and employed in some experiments with picosecond and femtosecond dye lasers, it is not clear if the noise properties of those lasers would have permitted the nonlinear generation of octave-bandwidth spectra while retaining the sharp comb structure. Indeed, the rapid progress of frequency combs at the turn of the century benefitted greatly from development in the 1990's of robust femtosecond solid state lasers, such as those based on Ti:sapphire. Although Ti:sapphire-based frequency combs were the first ones to be spectrally broadened to an octave and self-referenced, in the past decade several other femtosecond lasers have been employed as frequency combs. In line with the functional definition of the frequency comb provided above, Table 1 provides a summary and comparison of the seven different femtosecond lasers for which f_0 has been directly measured, and Fig. 3 shows some of the typical spectra obtained from a few of these comb sources.

This decade has witnessed an expansion of the frequency comb technology to new femtosecond lasers with some distinct advantages over the original Ti:sapphire. Primarily, erbium and ytterbium-based lasers, which are directly pumped by laser diodes, have emerged as low-cost, compact and robust alternatives. At the same time, the search for the ideal frequency comb continues and

Table 1. List Of Self-Referenced Frequency Combs, Including Some Key Parameters[a]

	Ti:Sapphire [14,18]	Cr:LiSAF [19]	Er:fiber [20,21]	Cr:forsterite [22,23]	Yb:fiber [24,25]	Yb:KYW [26]	Er:Yb:glass [27]
Center λ	800 nm	894 nm	1560 nm	1275 nm	1040 nm	1030 nm	1560 nm
Pulse length	10–50 fs	~50 fs	80–200 fs	30 fs	70–100 fs	290 fs	170 fs
Pump source	532 nm, doubled Nd:YVO	650 nm diode	980 or 1480 nm diode	1075 nm fiber laser	976 nm diode	980 nm diode	976 nm diode
Repetition rate	0.1–10 GHz	93 MHz	50–300 MHz	420 MHz	0.1–1 GHz	160 MHz	75 MHz
Octave-spectrum	~500–1200 nm; direct or in MSF[c]	~550–1100 nm in MSF	~1000–2000 nm in HNLF[d]	~1000–2000 nm in HNLF	700–1400 nm in MSF	700–1400 nm in MSF	~1000–2000 nm in HNLF
Electrical-to-optical efficiency[b]	~0.1%	1–2%	~1%	~0.5%	1–2%	~2–3%	~2–3%
Average optical power	1000 mW	150 mW	25–100 mW	500 mW	100–200 mW	>200 mW	>100 mW

[a]The order in which the different sources were introduced increases from left to right.
[b]Electrical-to-optical efficiency is the ratio of output optical power to input electrical power.
[c]MSF is short for micro-structured optical fiber, which generically refers to nonlinear air-silica holey fiber; sometimes also called photonic crystal fiber (PCF).
[d]HNLF refers to highly nonlinear fiber.

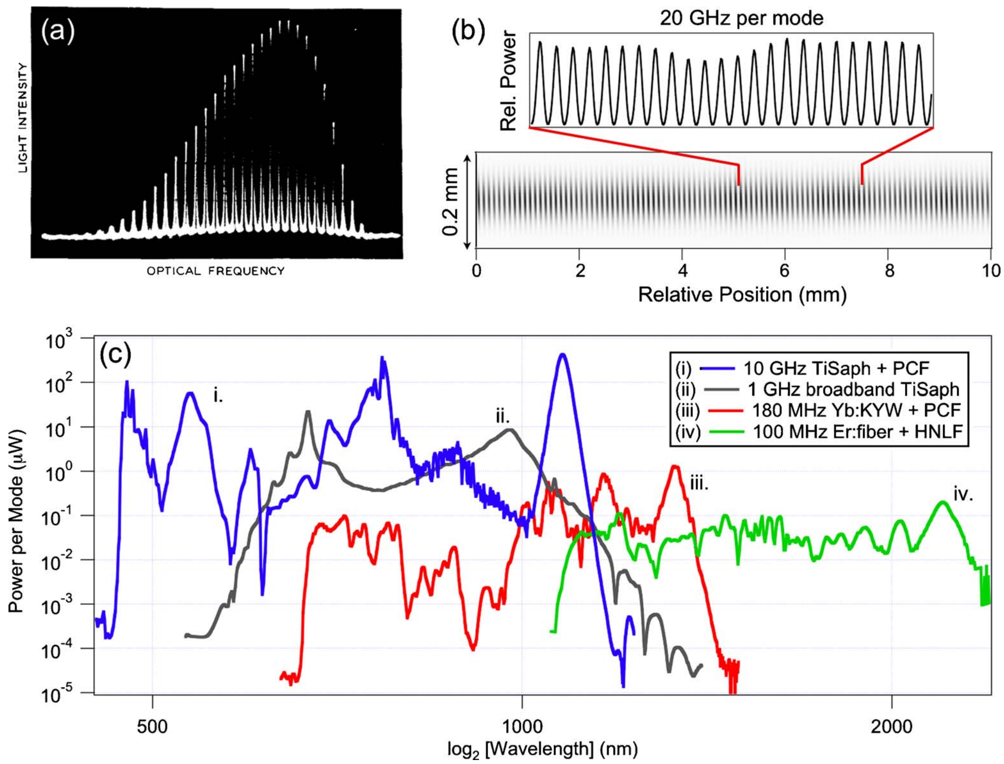

Fig. 3. (Color online) Spectra of early and more recent frequency combs. (a) Spectrum of first mode-locked laser operating near 633 nm, as measured with a scanning Fabry-Perot etalon. The mode spacing is ~56 MHz. Reprinted with permission from L. E. Hargrove, R. L. Fork, and M. A. Pollack, Appl. Phys. Lett. **5**, p. 4-5. Copyright 1964, American Institute of Physics. (b) A portion of the 20 GHz frequency comb generated by mode-filtering the output of a frequency-stabilized 1 GHz frequency comb [38]. The lower part of the figure is the grey-scale "fringes" of the comb as recorded with a grating spectrometer and CCD camera. The upper part of the figure shows a line out of a portion of the CCD image. The modes are spectrally separated by 20 GHz and spatially dispersed by about 100 microns per mode. Central wavelength is 960 nm. (c) Spectral envelope of some common optical frequency combs generated with different laser sources. (i) 10 GHz Ti:Saph [39], (ii) 1 GHz broadband Ti:Saph [37], (iii) 180 MHz Yb:KYW [26], (iv) 100 MHz Er:fiber [20].

each of the different lasers presented in Table 1 has its own benefits and disadvantages. However, in choosing the appropriate frequency comb for a given application, there are several key properties that need to be considered.

Repetition Rate: The best repetition rate is truly application dependent. For most traditional frequency metrology experiments, it is desirable to have the highest practical repetition rate at which an octave spectrum is obtained. This is typically in the range of a few hundred megahertz up to a few gigahertz. Given a fixed average power, a higher repetition rate provides more power per frequency mode [see Fig. 3(c)]. Some tunability (1–5%) of the repetition rate is desirable in the determination of the mode index. As will be discussed below, frequency synthesis, waveform generation, and the calibration of astronomical spectrographs require repetition rates in the range of a few gigahertz up to many tens of gigahertz. Broad spectral coverage at such high repetition rates is a significant and ongoing challenge. For applications that employ the comb directly for spectroscopy, the best repetition rate is more difficult to define. Lower rates ($\leq$100 MHz) with higher energy per pulse are desirable for high spectral resolution and when nonlinear frequency conversion is needed to get to mid-infrared or ultraviolet wavelengths. On the other hand, gigahertz repetition rates enable the direct resolution of individual modes and can permit nonlinear spectroscopy with an individual mode [28]. As seen in Table 1, with the exception of the gigahertz Ti:sapphire and Yb:fiber lasers, most frequency combs operate at repetition rates of a few hundred megahertz.

Frequency and Amplitude Noise: In the ideal case, the frequency comb should add no noise in excess of the reference oscillator that controls f_r and f_o (see Fig. 2). With reasonable control electronics, this is typically not an issue on time scales greater than ~0.01 s, as the comb faithfully reproduces the reference in accordance with $\nu_n = n \cdot f_r + f_o$. However, on time scales from $2/f_r$ up to ~1 ms, the noise properties of different frequency combs can vary substantially. Frequency noise that arises from temperature and acoustically driven fluctuations can usually be overcome with good mechanical design and well-designed control servos; however, amplitude and frequency noise coming from a noisy pump laser or from fundamental noise within the laser [amplified spontaneous emission, (ASE)] can be more challenging to eliminate [29,30]. Generally speaking, noise on the pump laser will be transferred to both fluctuations in the amplitude and frequency of the comb modes up to Fourier frequencies corresponding to the characteristic gain dynamics (typically ~5–10 kHz in Er:fiber [30], ~0.5–1 MHz in Ti:sapphire [31]). Femtosecond lasers with high intracavity power and short pulses will have less ASE-induced frequency noise on the comb. In this regard, Ti:sapphire and other solid-state lasers with cavity losses of a few percent should have an advantage over fiber lasers where losses can exceed 50%. External to the mode-locked laser itself, the main source of amplitude and phase noise come from spectral broadening in nonlinear fibers. Both technical and fundamental noise—from ASE and photon shot noise—can be amplified in the nonlinear fiber such as microstructured fiber and highly nonlinear fiber (HNLF) [32–34]. In some cases, this can lead to optical spectra nearly devoid of the original comb structure, although such decoherence is generally less of a factor when short pulses (e.g., <50 fs) are used to pump a short nonlinear fiber having zero dispersion near the central wavelength of the femtosecond laser [33].

Spectral Bandwidth and Coverage: Figure 3 shows that the wavelength range from 400 nm to nearly 2200 nm is well covered by a combination of Ti:sapphire, Er:fiber, and Yb-based lasers. To date, Ti:sapphire is the only laser capable of directly generating octave-spanning spectra without the use of microstructured fiber [35–37]. In all other cases, nonlinear optical fibers are employed to obtain the necessary spectrum for self-referencing.

Size, Weight, and Power: A definite and important trend for frequency comb sources is towards smaller, more efficient, more robust and less expensive sources. Compelling applications in spectroscopy, length measurement [40], waveform synthesis, and optical atomic clocks will ultimately require frequency combs that can operate in real-world conditions outside the research lab. Such environments require not only robustness, but overall power usage is an important factor. Table 1 also provides approximate numbers for the present electrical-to-optical (i.e., "wall-plug") efficiency of existing frequency combs. It is clear that the most significant reduction in power usage comes with the direct diode- pumped Er and Yb systems, which are about 10× more efficient than Ti:sapphire. These are conservative values for just the laser part of the frequency comb, and one could realistically project that, with careful engineering, a wall-plug efficiency greater than 5% might be possible for a system producing an octave-spanning spectrum. This would imply a power usage of only 5–10 W. Considering the additional power for control electronics and temperature control, it is nonetheless plausible that a complete frequency comb could consume less than 50 W, weigh 10 kg, and fit a volume of roughly 10–15 liters.

The development of broad bandwidth combs with mode spacings on the order of 10–50 GHz is very challenging but valuable for emerging applications in microwave photonics as well as in the calibration of astronomical spectrographs. Because the pulse energy scales inversely with repetition rate, the nonlinear spectral broadening at rates of many tens of gigahertz is much less effective than at 100 MHz repetition rate. To date, the highest repetition rate for which octave-spanning spectra and self-referencing has been achieved is 10 GHz with a ring Ti:sapphire laser [39]. Along the lines of higher repetition rates, a fascinating development of the past few years was the parametric generation of frequency combs in highly nonlinear microresonators [41]. Pumped by a single cw laser, such parametric combs have been demonstrated in micro-toroids [41], crystalline resonators [42], and integrated waveguide resonators [43–45]. Octave bandwidths have recently been achieved for mode spacings of 850 GHz [46], and detection and stabilization of the carrier-envelope offset frequency seems possible.

3. FREQUENCY COMBS FOR IMPROVED FREQUENCY MEASUREMENTS AND OPTICAL CLOCKS

Tracing the history of time-keeping through astronomical, mechanical, and atomic eras, one is struck by the trend of exponential improvement [47]. In the past decade there has been no deviation from this trend, and in fact we have witnessed accelerated improvements arising from the transformational laser-based technologies which form the main components of the next-generation atomic clocks. The femtosecond laser frequency comb plays the role of an "optical clockwork" in this decade of accelerated progress. While focusing our attention on the frequency comb, here we also briefly discuss the advances in the areas of laser stabilization, laser-based cooling and trapping, and precision spectroscopy.

Figure 4 attempts to track the history of the published improvements in the uncertainty with which frequencies in the optical and terahertz domain have been measured

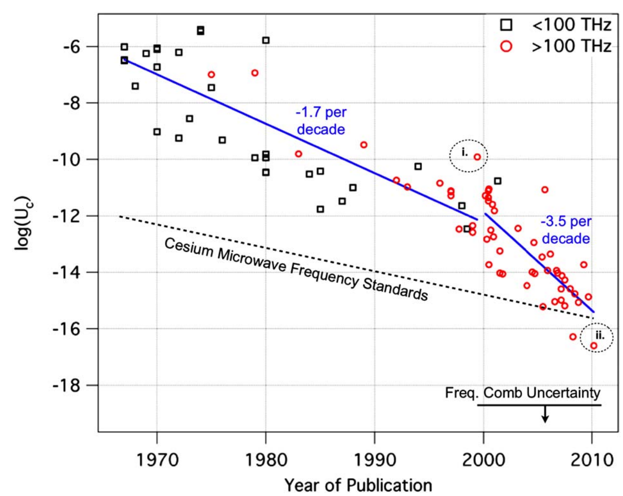

Fig. 4. (Color online) Record of improvement of precise frequency measurements in the optical and terahertz frequency domains over time. The y-axis is the base-ten logarithm of the fractional uncertainty $Uc = \sigma/\nu_0$, where σ is the standard uncertainty and ν_0 is the carrier frequency. A distinction is made between frequencies less than and greater than 100 THz, which is somewhat arbitrarily defined as the beginning of the "optical" domain. Note that there were just a few measurements at frequencies greater than 100 THz prior to 1990, and it was not until the introduction of the femtosecond laser frequency comb around 2000 that the rate at which optical frequencies were measured truly accelerated and became commonplace. The circled points are a few noteworthy measurements: (i) The first measurement of an optical frequency involving a femtosecond laser frequency comb [55]. (ii) The most accurate measurement of an optical frequency standard, which is presently the ratio of the frequencies of two Al^+ quantum logic clocks with combined uncertainty of 2.5×10^{-17}. The evaluated inaccuracy of one of the Al^+ clocks is a factor of ~3 smaller at 8.6×10^{-18} [63]. The residual fractional uncertainty of the frequency comb in the comparison of optical frequency standards and the construction of an optical clock is at or below 1×10^{-19}.

over the past ~40 years. This is a revised and updated version of similar data originally presented by Hollberg *et al.* [48]. Significant to this update is an attempt to include most of the accurate frequency measurements made in the past decade. That being stated, the rapid progress in this field makes it challenging to ensure that all frequency measurements have been included. Indeed, at NIST over the past decade, there were active periods when optical frequencies were measured almost daily. Many such measurements were used for internal evaluations and were therefore never published. The same situation could be expected from other metrology institutes and research labs.

Given the wide variety of frequencies and measurement techniques, it is impossible to expect that one plot could adequately (or fairly) compare all such measurements. In some of the measurements, the spectroscopy might have been the limitation, while in others it was the frequency reference or measurement technique. Nonetheless, the progress over the past 40 years is evident by eye, and a few linear fits have been included in attempt to quantify the progress. On average, the rate of improvement from 1967 through 2010 is an impressive factor of 80 per decade. As a reference, this is compared to the more mature technology of cesium microwave atomic clocks, where the historical rate of improvement over the same epoch is approximately a factor of 10 per decade. Linear fits to the periods pre and post 2000 yield rates of improvements of 50× and nearly 3200× per decade, respectively. Clearly, the confluence around the turn of the century of the frequency comb technology with advances in cold atom techniques and laser stabilization have made the past decade one of exceptional activity, progress, and excitement for developing optical clocks. Additionally, one clearly notes that there were just a few measurements at frequencies greater than 100 THz prior to 1990, and it was not until the introduction of the femtosecond laser frequency comb around 2000 that the rate at which optical frequencies were measured truly accelerated and became commonplace.

Even in the early 1960's, scientists working with the first lasers recognized the potential of lasers as superior tools for precision spectroscopy and measurement [49,50]. Yet, while significant breakthroughs in laser stabilization and precision spectroscopy provided the means for the oscillator and atomic reference of an optical clock, the critical clockwork required to make a practical device was largely missing. Interestingly, between the late 1970's and early 1990's there were a few publications that explored the concept of using the comb from a mode-locked laser to translate across large gaps in the optical domain [6,51], or to even coherently relate an optical frequency to a microwave one [9,52]. However, these ideas were largely placed on hold, while the multiplicative [53] and interval division [54] approaches were pursued for the connection of the optical and microwave domains. Although these approaches were complex, expensive, and not really practical outside large research facilities, Fig. 4 bears testimony to the significant results and consistent improvements in measurement uncertainty that were achieved from the mid 1960's through the end of the 20th century.

1999 brought with it the transformational introduction of the femtosecond laser frequency comb as we presently know it [55,56]. This was a singular event that changed the direction of precision frequency metrology and the development of optical clocks [57]. The frequency comb, at last, provided the critical clockwork to complete the next-generation atomic clock. However, the facility with which the comb functioned to connect the optical and microwave domains was also accompanied by healthy metrological skepticism, leading to several measurements that confirmed the basic comb equation ($\nu_n = n \cdot f_r + f_o$) and cemented the comb's position as an accurate and reliable gear with residual uncertainty below 1×10^{-19} [56,58,59]

The timing of the arrival of the femtosecond frequency comb was fortuitous, as 1999 also brought with it the first cw laser having sub-Hertz linewidth, serving the purpose of the "pendulum" of the optical clock [60]. The remarkable laser stabilization results of Bergquist and coworkers remain the very best published to date, although sub-Hertz optical linewidths have now been achieved regularly and repeatedly in a variety of different laser systems. The technology associated with the laser cooling and trapping has also reached maturity, thereby providing various isolated and nearly motionless quantum references for the new clocks. Critical advances in this area over the past ten years include the proposal and demonstration of the dual-ion quantum logic clock [61–63], as well as the so-called "optical lattice" clock employing state-insensitive dipole trapping of neutral atoms [64]. Operating together, the three components of ultrastable lasers, cold atoms and ions and the clockwork of the optical frequency comb have provided a decade of unparalleled improvements in atomic timekeeping.

4. FREQUENCY COMB SPECTROSCOPY: FROM TRACE GAS DETECTION TO EXOPLANET SEARCHES

Beyond enabling advances in optical frequency metrology, a stabilized optical frequency comb can be a versatile spectroscopic tool, providing excellent frequency accuracy, high spectral purity and, at the same time, broad spectral coverage. Traditionally, ultrashort pulses have been used like a fast photographic flash to probe physical processes with temporal resolution on the sub-picosecond time scale. Thus, it might initially seem surprising to consider using the output of the mode-locked laser for high-spectral-resolution spectroscopy. The spectral resolution provided by a single ultrafast pulse is given by the inverse of the pulse duration (~1/100 fs=10 THz), but the coherent accumulation of many pulses is what leads to the formation of the discrete frequency comb. With good frequency control, absolute comb linewidths, and frequency uncertainty at the 1 Hz level are achievable over the hundreds of terahertz of the comb bandwidth. As depicted in Fig. 2, each of the $\sim10^6$ modes of the frequency comb can then be thought of as a frequency-stabilized cw laser available for precision spectroscopy.

As the frequency comb has developed over this decade, two general spectroscopic approaches have emerged. In the first case, the comb serves simply as a frequency ruler against which a cw laser is calibrated and measured. It is the cw laser that then performs the spectroscopy. One of the best-known examples of this approach is the spectroscopy and improved measurements of the 1s–2s transition in atomic hydrogen [65]. A variation on this approach is the exciting possibility to use a broad bandwidth, high-repetition-rate frequency comb as a precision wavelength scale for the calibration of astronomical spectrographs. Among other measurements of significance, such work could provide the required precision to detect the tiny Doppler shifts associated with an earthlike planet orbiting a star beyond our own sun. The second general approach employs the frequency comb to directly probe atomic and molecular samples. Interestingly, this is the route that was launched in the late 1970's to explore pairs of comb modes and multiphoton transitions in atomic systems [6,66]. In the following, this latter class of spectroscopy experiments is first described, and then we return to the former topic, highlighting recent work in astronomical spectrograph calibration.

Direct Frequency Comb Spectroscopy: Just as in cw laser spectroscopy experiments, generating the frequency comb light, controlling its frequency, and illuminating the sample is one part of the spectroscopy problem. However, retrieving the relevant (sometimes weak) signal often entails additional challenges, particularly in the case of frequency comb spectroscopy where one is now dealing with the equivalent of 10^5 or 10^6 cw lasers. Some of the most interesting advances in this regard have arisen with techniques that take full advantage of the multiplexed nature of high-resolution spectroscopy with a large number of spectral modes. Figure 5 shows three experimental approaches that have yielded promising results.

The first case, employing fluorescence detection, is the most straightforward to implement. A frequency comb illuminates the sample and the fluorescence from an excited state is detected while either f_r, and f_o is scanned. This basic approach has been implemented with alkali atoms in a magneto-optical trap (MOT), beam, and cell [67–70]. With cold atoms, or propagation orthogonal to an atomic beam, Doppler broadening can be greatly reduced, yielding uncertainties in the determination of transition frequencies that begin to rival the best measurements presented with cw laser spectroscopy. Counter-propagating beams can also provide reduced Doppler broadening when multiphoton transitions are being investigated, although the cancellation of the Doppler broadening is not complete in such a situation [70,71]. One disadvantage of fluorescence detection is that multiple comb elements can simultaneously interact with different transitions for the same value of f_r and f_o. Thus, the detected fluorescence may arise from two (or many) levels which share the same excited state, rendering the individual transitions indistinguishable.

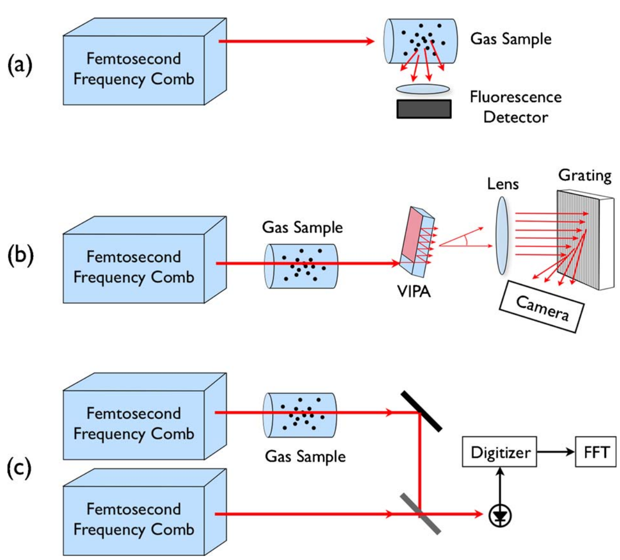

Fig. 5. (Color online) Three different spectroscopy approaches using femtosecond laser frequency combs. (a) Fluorescence detection. (b) Spectrally dispersed detection with a high resolution imaging spectrometer based on a VIPA (virtually-imaged phased array) disperser. (c) Dual comb multi-heterodyne spectroscopy with a point detector and high-speed data acquisition. In all cases, what is depicted as a vapor cell could in fact be atoms or molecules in a beam, trap, or other gas phase sample. In principle, liquids and solids could also be studied in a similar fashion.

The techniques shown in Fig. 5(b) and 5(c) provide alternative approaches that circumvent this problem by directly measuring the power and/or phase of individual comb teeth that have interacted with the atomic or molecular gas. In the case of Fig. 5(b), a novel high-resolution crossed spectral disperser is employed to project the various frequency comb modes onto a two-dimensional digital camera. While crossed spectral dispersers have been used for many years in spectroscopy, a distinguishing feature of the present approach is the use of a side-entrance etalon called a virtually imaged phased array (VIPA) disperser that provides ~1 GHz resolution in the visible spectral range (resolution of ~500 MHz at 1550 nm) [72–75]. When combined with a lower dispersion grating in the orthogonal spatial direction, 5–10 THz of bandwidth can be captured in a single measurement taking a few milliseconds. This technique has been successfully employed for molecular fingerprinting and trace gas detection in the visible, near-infrared, and mid-infrared spectral regions. When combined with a multi-pass cell or enhancement cavity, minimum detectable absorption below 1×10^{-9} cm^{-1} has been achieved along with sensitivities to concentration below 10 ppb for some common gases [72,74,75].

The final approach, which is illustrated in Fig. 5(c), employs two frequency combs with their repetition rates slightly detuned from each other (e.g., by ~1 kHz). One of the combs serves the role of a reference, while the second acts as a probe, which passes through the sample [76–78]. Data is acquired from the heterodyne beat between the two combs using a high-speed detector and digitizer. The basic operation can be understood in both the time and frequency domains. In the former, the slight detuning of the combs results in multiple heterodyne beats between individual pairs of modes from each comb. This is the time domain equivalent to the pulse trains from the two combs walking through each other, similar to what could be achieved with a scanning delay line—although with no moving parts. The digitized data is similar to that acquired from a conventional scanning Michelson interferometer, and the complex spectrum (amplitude and phase) is obtained via Fourier transformation. With mutual stabilization of the frequency combs, sequential interferograms can be obtained and averaged, resulting in high signal-to-noise over broad spectral bandwidths with high spectral resolution [78]. While two frequency combs are required, this approach has the advantage of employing a single point detector. It is also compatible with cavity enhancement, in which case a minimum detectable absorption $\sim 1\times 10^{-8}$ cm^{-1} has been demonstrated [79].

One interesting advantage of frequency comb spectroscopy over more conventional cw laser techniques is that the high peak powers associated with femtosecond lasers permit efficient generation of light at wavelengths that are challenging, if not impossible, to reach using cw lasers. Examples include the generation of light and its use in spectroscopy experiments at ultraviolet wavelengths in the range of 40–200 nm [80–82] as well as the infrared region (2.5–15 μm) [75,77,83]. Such broad-bandwidth frequency-comb spectroscopy is in its infancy, and its full impact remains to be determined. Nonetheless, the next decade promises new uses and applications of frequency combs ranging from fundamental spectroscopy, quantum control [84,85] and chemical analysis to trace gas detection for medical, environmental, and security applications.

Astronomical Spectrograph Calibration: Beginning more than 200 years ago with the discoveries of Wollaston and Fraunhofer, spectroscopy has been one of the most important tools for learning about the universe beyond our earth. Now, in much the same way as the frequency-stabilized laser has replaced the krypton lamp for the realization of the meter, broad-bandwidth frequency combs with large mode spacing may begin to replace more conventional discharge lamps and absorption cells for astronomical spectrographs that require the highest level of calibration. Precise measurements of small spectral shifts from astronomical sources are key to the discovery of earthlike exoplanets, measuring fundamental constants in the early universe, and perhaps even directly measuring the changing rate of cosmic expansion [86].

As illustrated by Fig. 6, the frequency comb has the desirable properties of atomically traceable comb modes

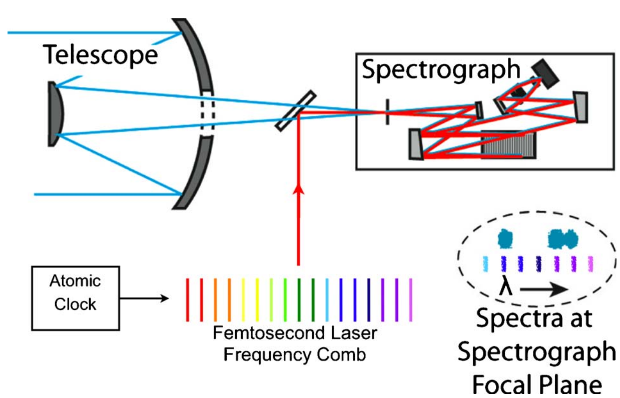

Fig. 6. (Color online) Concept of an astronomical spectrograph calibrated by a femtosecond laser frequency comb. The atomically-referenced comb spectrum enters the spectrograph offset from the star light such that it appears below the dispersed stellar spectrum. In reality a fiber-fed spectrograph is preferable to reduce pointing errors. The optimum spacing of the comb lines is one every 3–4 resolution elements [86], amounting to ~30 GHz for a visible spectrograph with resolving power of 60,000.

with uniform spacing and broad spectral coverage that appear to make it an ideal spectral calibrator for high-resolution astronomical spectrographs [86–90]. The level of required precision for the most demanding astronomical spectroscopy is $\sim 10^{-11}$ to 10^{-9}. While this may be many orders of magnitude less precise than present optical frequency standards and clocks (see Fig. 4), there are other factors involving both the astronomical instrumentation and the frequency comb that make such precision challenging. To begin, the required precision amounts to line centering at the level of a part in 10^4 or even 10^5 using very faint sources with significantly "messier" lines than those obtained from isolated atoms or molecules in the lab. This challenge certainly requires a stable reference, but for the comb to be useful for this application its modes must be easily resolved by the spectrograph. In a typical echelle spectrograph, this amounts to a mode spacing of several tens of gigahertz in the visible or near infrared. Moreover, such large mode spacing is required over bandwidths of hundreds of nanometers, which is an extremely difficult laser physics problem—particularly in the visible portion of the spectrum.

Some promising avenues towards this goal include the direct generation of high-repetition-rate self-referenced frequency combs, such as a recently demonstrated 10 GHz Ti:sapphire [see spectra (i) of Fig. 3(c)] [39], mode filtering of lower repetition rate sources [see Fig. 3(b)] [38,87–90], or the previously mentioned generation of broadband combs via parametric means in microresonators [41–46]. At present, no ideal source in all regards exists for this application, which makes it a challenging but exciting field of research. In the meantime, several groups are moving forward with calibration efforts using narrower bandwidth combs, and preliminary results appear promising [91–93].

5. FREQUENCY COMBS FOR MICROWAVE PHOTONICS

In this final section, the role of optical frequency combs as an emerging tool in applications in microwave photonics will be examined briefly. For the purposes of this article, microwave photonics will be broadly defined as the field that utilizes tools in the optical domain for the generation, processing, control, and distribution of microwave, millimeter-wave, and terahertz signals. Within this context, the broad bandwidths and precise frequency control of the frequency comb appear to be some unique and potentially interesting benefits when employed as an optical and microwave frequency synthesizer. Here we discuss two emerging applications along these lines: (1) the generation of microwave signals with low phase noise, and (2) line-by-line optical pulse shaping.

As already mentioned a cw laser oscillator stabilized to a high-finesse optical cavity (resonant Q approaching 10^{11}) is one of the lowest phase noise electromagnetic oscillators available in any frequency range [60]. One can then use this ultrastable cw oscillator as the reference for a frequency comb. In the manner described in Fig. 2, the low phase noise properties of the cw laser oscillator can be transferred to all the elements of the optical frequency comb, including the repetition rate and its harmonics.

Once stabilized, the various modes of the frequency comb can be used to synthesize other frequencies or waveforms with low phase noise in the microwave, millimeter-wave, or terahertz domains. In the simplest case, a fast photodetector at the output of the stabilized frequency comb will generate photocurrent at frequencies equal to the spacing of the comb modes [97,98]. For example, in the case of a 1 GHz frequency comb, the photocurrent output from the photodiode is made up of tones at 1,2,3,4.... GHz, up to the cutoff frequency of the diode. Figure 7(a) shows what should be achievable for the phase noise on the 10 GHz harmonic, based on predictions and measurements of the phase noise associated with a stable optical reference cavity [60]. Close to carrier, the phase noise is fundamentally limited by thermal noise of the high-finesse optical reference cavity at about −110 dBc/Hz at 1 Hz, decreasing as $1/f^3$. Residual noise as low as −120 dBc/Hz at 1 Hz offset frequency from an 11.55 GHz carrier was recently achieved using fiber laser frequency combs [99]. The white noise floor in such a system is provided by the photodetector shot noise, relative to the power in the generated 10 GHz harmonic. With a 1 GHz frequency comb, a white noise floor near −155 dBc/Hz was measured [97], limited by the saturation effects in the photodiode. With higher repetition rates, saturation in the photodiodes is less severe and higher 10 GHz powers have been achieved, indicating that a noise floor near −165 dBc/Hz might be achievable for average photocurrents approaching 10 mA [100]. For

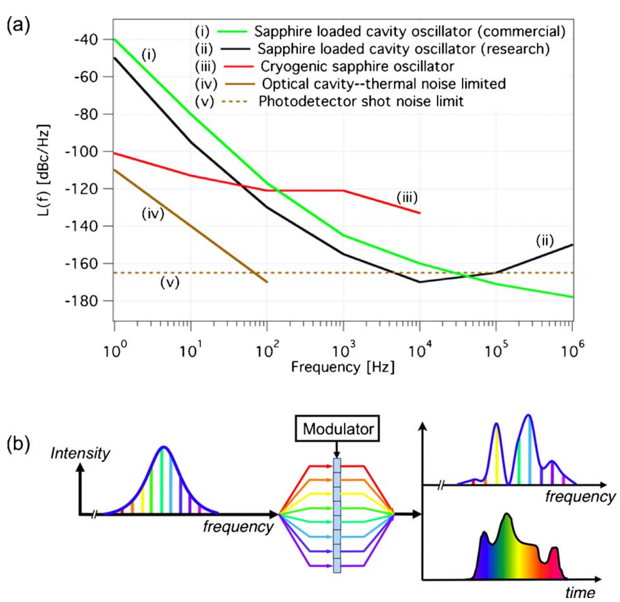

Fig. 7. (Color online) (a) Comparison of measured and projected phase noise on the 10 GHz signal generated from a few sources. (i) measured commercial sapphire oscillator [94], (ii) measured research sapphire oscillator [95], (iii) measured cryogenic sapphire oscillator [96], (iv) projected phase noise for a frequency comb locked to a cw laser that is stabilized to a high-finesse reference cavity [97,98,100]. (v) dashed line is the projected white noise floor given by photodetector shot noise. (b) General approach to line-by-line pulse shaping with an optical frequency comb. The different comb elements are spatially/spectrally dispersed and directed to a modulator array, where individual comb lines are modified in amplitude and phase. The modes are then re-combined to form an optical field with user-defined spectral and temporal characteristics.

comparison, the phase noise measurements from a few other 10 GHz microwave oscillators based on sapphire dielectric resonators are shown in Fig. 7(a).

Should such a level of stability be achieved with high-repetition-rate frequency combs, then one could also envision using line-by-line amplitude and phase control of the individual modes for the generation of synthesized optical and microwave waveforms with correspondingly low phase noise and timing jitter [Fig. 7(b)]. Demonstrations of different pulse shaping architectures using unstabilized frequency combs have been carried out [101,102], and the additional aspect of line-by-line pulse shaping with a low-phase-noise comb could add enhanced capabilities for applications in signal processing, secure communications, radar, and imaging, to name a few.

6. SUMMARY AND OUTLOOK

This past decade has witnessed the opening of many new research directions associated with femtosecond laser frequency combs. Some of the rapidly changing developments include the realization of optical clocks that now keep time on the femtosecond time scale, new broadband laser sources, novel spectroscopic techniques, and frequency synthesis with ultralow phase noise. Indeed, if there is a common theme in the story of the frequency comb it is that new developments have emerged often in unexpected areas of research at the borders of existing fields. Analogous to its role in optical clocks, the frequency comb is at its best as a connector of new applications and ideas, and it will be interesting to see what new branches and associated connections evolve in the coming decade.

ACKNOWLEDGMENTS

Much of this review is drawn from the contributions and collaborative discussions over the past decade with A. Bartels, S. Cundiff, J. Hall, T. Hänsch, L. Hollberg, R. Holzwarth, N. Newbury, Th. Udem, and J. Ye. Special thanks go to T. Johnson, and C. Oates for their additional thoughtful comments on this manuscript. I am grateful to T. Fortier, D. Heinecke, M. Kirchner and S. Meyer for their contributions to Figs. 3(b) and 3(c), and S. Osterman for providing Fig. 6. Finally, thanks to L. Hargrove and R. Fork for providing Fig. 3(a) as well as illuminating information about the first mode-locked He–Ne laser. NIST is an agency of the U.S. government, and this work is not subject to copyright in the U.S.

REFERENCES

1. T. H. Maiman, "Stimulated optical radiation in ruby," Nature **187**, 493 (1960).
2. A. Javan, W. R. Bennett, Jr., and D. R. Herriott, "Population inversion and continuous optical maser oscillation in a gas discharge containing a He–Ne mixture," Phys. Rev. Lett. **6**, 106–110 (1961).
3. L. E. Hargrove, R. L. Fork, and M. A. Pollack, "Locking of He–Ne laser modes induced by synchronous intracavity modulation," Appl. Phys. Lett. **5**, 4–5 (1964).

4. A. J. DeMaria, D. A. Stetser, and H. Heynau, "Self mode-locking of lasers with saturable absorbers," Appl. Phys. Lett. **8**, 174–176 (1966).
5. E. P. Ippen, C. V. Shank, and A. Dienes, "Passive mode locking of the cw dye laser," Appl. Phys. Lett. **21**, 348–350 (1972).
6. J. N. Eckstein, A. I. Ferguson, and T. W. Hänsch, "High-resolution 2-photon spectroscopy with picosecond light-pulses," Phys. Rev. Lett. **40**, 847 (1978).
7. J. L. Hall, "Optical frequency measurement: 40 years of technology revolutions," IEEE J. Sel. Top. Quantum Electron. **6**, 1136 (2000).
8. Th. Udem, R. Holzwarth, and T. W. Hänsch, "Optical frequency metrology," Nature **416**, 233 (2002).
9. T. W. Hänsch, "Nobel lecture: Passion for precision," Rev. Mod. Phys. **78**, 1297 (2006).
10. J. L. Hall, "Nobel lecture: Defining and measuring optical frequencies," Rev. Mod. Phys. **78**, 1279 (2006).
11. J. K. Ranka, R. S. Windeler, and A. J. Stentz, "Visible continuum generation in air—silica microstructure optical fibers with anomalous dispersion at 800 nm," Opt. Lett. **25**, 25–27 (2000).
12. S. A. Diddams, D. J. Jones, J. Ye, S. T. Cundiff, J. L. Hall, J. K. Ranka, R. S. Windeler, R. Holzwarth, T. Udem, and T. W. Hänsch, "Direct link between microwave and optical frequencies with a 300 THz femtosecond laser comb," Phys. Rev. Lett. **84**, 5102 (2000).
13. H. R. Telle, G. Steinmeyer, A. E. Dunlop, J. Stenger, D. H. Sutter, and U. Keller, "Carrier-envelope offset phase control: A novel concept for absolute optical frequency measurement and ultrashort pulse generation," Appl. Phys. B **69**, 327 (1999).
14. D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall, and S. T. Cundiff, "Carrier-envelope phase control of femtosecond modelocked lasers and direct optical frequency synthesis," Science **288**, 635 (2000).
15. T. M. Ramond, S. A. Diddams, L. Hollberg, and A. Bartels, "Phase coherent link from optical to microwave frequencies via the broadband continuum from a 1 GHz Ti:sapphire femtosecond oscillator," Opt. Lett. **27**, 1842–1844 (2002).
16. Z. Chang and P. Corkum, "Attosecond photon sources: the first decade and beyond," J. Opt. Soc. Am. B **27**, B9–B17 (2010).
17. A. Baltuška, Th. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, Ch. Gohle, R. Holzwarth, V. S. Yakovlev, A. Scrinzi, T. W. Hänsch, and F. Krausz, "Attosecond control of electronic processes by intense light fields," Nature **421**, 611 (2003).
18. A. Apolonski, A. Poppe, G. Tempea, Ch. Spielmann, Th. Udem, R. Holzwarth, T. W. Hänsch, and F. Krausz, "Controlling the phase evolution of few-cycle light pulses," Phys. Rev. Lett. **85**, 740–743 (2000).
19. R. Holzwarth, M. Zimmermann, Th. Udem, T. W. Hänsch, P. Russbüldt, K. Gäbel, R. Poprawe, J. C. Knight, W. J. Wadsworth, and P. St. J. Russell, "White-light frequency comb generation with a diode-pumped Cr:LiSAF laser," Opt. Lett. **26**, 1376–1378 (2001).
20. B. Washburn, S. Diddams, N. Newbury, J. W. Nicholson, M. F. Yan, and C. G. Jørgensen, "A self-referenced, erbium fiber laser-based frequency comb in the near infrared," Opt. Lett. **29**, 252–254 (2004).
21. F. Tauser, A. Leitenstorfer, and W. Zinth, "Amplified femtosecond pulses from an Er:fiber system: Nonlinear pulse shortening and self-referencing detection of the carrier-envelope-phase evolution," Opt. Express **11**, 594–600 (2003).
22. K. Kim, B. R. Washburn, G. Wilpers, C. W. Oates, L. Hollberg, N. R. Newbury, S. A. Diddams, J. W. Nicholson, and M. F. Yan, "Stabilized frequency comb with a self-referenced femtosecond Cr:forsterite laser," Opt. Lett. **30**, 932–934 (2005).
23. K. Kim, S. A. Diddams, P. Westbrook, J. W. Nicholson, and K. S. Feder, "Improved stabilization of a 1.3 μm femtosecond optical frequency comb using spectrally tailored continuum from a nonlinear fiber grating," Opt. Lett. **31**, 277–279 (2006).
24. I. Hartl, M. E. Fermann, P. Pal, and W. H. Knox, "Self-referenced Yb-fiber-laser frequency comb using a dispersion micromanaged tapered holey fiber," in Conference on Lasers and Electro-Optics/Quantum Electronics and Laser Science Conference and Photonic Applications Systems Technologies, OSA Technical Digest Series (CD) (Optical Society of America, 2007), paper CMU2.
25. I. Hartl, L. B. Fu, B. K. Thomas, L. Dong, M. E. Fermann, J. Kim, F. X. Kärtner, and C. Menyuk, "Self-referenced f_{CEO} stabilization of a low noise femtosecond fiber oscillator," in Conference on Lasers and Electro-Optics/Quantum Electronics and Laser Science Conference and Photonic Applications Systems Technologies, OSA Technical Digest (CD) (Optical Society of America, 2008), paper CTuC4.
26. S. A. Meyer, J. A. Squier, and S. A. Diddams, "Diode-pumped Yb:KYW femtosecond laser frequency comb with stabilized carrier-envelope offset frequency," European Physics Journal D **48**, 19 (2008).
27. M. C. Stumpf, S. Pekarek, A. E. H. Oehler, T. Südmeyer, J. M. Dudley, and U. Keller, "Self-referencable frequency comb from a 170-fs, 1.5-μm solid-state laser oscillator," Appl. Phys. B **99**, 401–408 (2009).
28. D. C. Heinecke, A. Bartels, T. M. Fortier, D. A. Braje, L. Hollberg, and S. A. Diddams, "Optical frequency stabilization of a 10 GHz Ti:sapphire frequency comb by saturated absorption spectroscopy in 87Rubidium," Phys. Rev. A **80**, 053806 (2009).
29. H. A. Haus and A. Mecozzi, "Noise of mode-locked lasers," IEEE J. Quantum Electron. **29**, 983–996 (1993).
30. N. R. Newbury and W. C. Swann, "Low-noise fiber-laser frequency combs," J. Opt. Soc. Am. B **24**, 1756–1770 (2007).
31. R. P. Scott, T. D. Mulder, K. A. Baker, and B. H. Kolner, "Amplitude and phase noise sensitivity of modelocked Ti-:sapphire lasers in terms of a complex noise transfer function," Opt. Express **15**, 9090–9095 (2007).
32. N. R. Newbury, B. R. Washburn, K. L. Corwin, and R. S. Windeler, "Noise amplification during supercontinuum generation in microstructure fiber," Opt. Lett. **28**, 944– 946 (2003).
33. K. L. Corwin, N. R. Newbury, J. M. Dudley, S. Coen, S. A. Diddams, K. Weber, and R. S. Windeler, "Fundamental noise limitations to supercontinuum generation in microstructure fiber," Phys. Rev. Lett. **90**, 113904 (2003).
34. J. N. Ames, S. Ghosh, R. S. Windeler, A. L. Gaeta, and S. T. Cundiff, "Excess noise generation during spectral broadening in microstructured fiber," Appl. Phys. B **77**, 279 (2003).
35. R. Ell, U. Morgner, F. X. Kärtner, J. G. Fujimoto, E. P. Ippen, V. Scheuer, G. Angelow, and T. Tschudi, "Generation of 5 fs pulses and octave-spanning spectra directly from a Ti-:sapphire laser," Opt. Lett. **26**, 373–375 (2001).
36. T. M. Fortier, D. J. Jones, and S. T. Cundiff, "Phase stabilization of an octave-spanning Ti:sapphire laser," Opt. Lett. **28**, 2198–2200 (2003).
37. T. Fortier, A. Bartels, and S. A. Diddams, "Octave-spanning Ti:sapphire laser with a repetition rate >1 GHz for optical frequency measurements and comparisons," Opt. Lett. **31**, 1011–1013(2006).
38. M. S. Kirchner, D. A. Braje, T. M. Fortier, A. M. Weiner, L. Hollberg, and S. A. Diddams, "Generation of 20 GHz, sub-40 fs pulses at 960 nm via repetition rate multiplication," Opt. Lett. **34**, 872–874 (2009).
39. A. Bartels, D. Heinecke, and S. A. Diddams, "10 GHz self-referenced optical frequency comb," Science **326**, 681 (2009).
40. I. Coddington, W. C. Swann, L. Nenadovic, and N. R. Newbury, "Rapid and precise absolute distance measurements at long range," Nat. Photonics **3**, 351 (2009).
41. P. Del'Haye, A. Schliesser, O. Arcizet, T. Wilken, R. Holzwarth, and T. J. Kippenberg, "Optical frequency comb generation from a monolithic microresonator," Nature **450**, 1214–1217 (2007).
42. A. A. Savchenkov, A. B. Matsko, V. S. Ilchenko, I. Solomatine, D. Seidel, and L. Maleki, "Tunable optical frequency

comb with a crystalline whispering gallery mode resonator," Phys. Rev. Lett. **101**, 093902 (2008).

43. J. S. Levy, A. Gondarenko, M. A. Foster, A. C. Turner-Foster, A. L. Gaeta, and M. Lipson, "CMOS-compatible multiple-wavelength oscillator for on-chip optical interconnects," Nat. Photonics **4**, 37–40 (2010).
44. L. Razzari, D. Duchesne, M. Ferrera, R. Morandotti, S. Chu, B. E. Little, and D. J. Moss, "CMOS-compatible integrated optical hyper-parametric oscillator," Nat. Photonics **4**, 41–45 (2010).
45. D. Braje, L. Hollberg, and S. A. Diddams, "Brillouin-enhanced hyperparametric generation of an optical frequency comb in a monolithic highly nonlinear fiber cavity pumped by a cw laser," Phys. Rev. Lett. **102**, 193902 (2009).
46. P. Del'Haye, T. Herr, E. Gavartin, R. Holzwarth, and T. J. Kippenberg, "Octave spanning frequency comb on a chip," arXiv:0912.4890v1 [physics.optics].
47. S. A. Diddams, J. C. Bergquist, S. R. Jefferts, and C. W. Oates, "Standards of time and frequency at the outset of the 21st century," Science **306**, 1318 (2004), and references therein.
48. L. Hollberg, S. Diddams, A. Bartels, T. Fortier, and K. Kim, "The measurement of optical frequencies," Metrologia **42**, S105 (2005).
49. A. Javan, E. A. Ballik, and W. L. Bond, "Frequency characteristics of a continuous-wave He–Ne optical maser," J. Opt. Soc. Am. **52**, 96–98 (1962).
50. R. L. Barger and J. L. Hall, "Pressure shift and broadening of methane line at 3.39 μ studied by laser-saturated molecular absorption," Phys. Rev. Lett. **22**, 4 (1969).
51. D. M. Kane, S. R. Bramwell, and A. I. Ferguson, "FM dye lasers," Appl. Phys. B **39**, 171 (1986).
52. D. J. Wineland, J. C. Bergquist, W. M. Itano, F. Diedrich, and C. S. Weimer, "Frequency standards in the optical spectrum," in *The Hydrogen Atom* G. F. Bassani *et al.*, eds. (Springer, 1989), pp. 123–133.
53. H. Schnatz, B. Lipphardt, J. Helmcke, F. Riehle, and G. Zinner, "First phase-coherent frequency measurement of visible radiation," Phys. Rev. Lett. **76**, 18–21 (1996).
54. H. R. Telle, D. Meschede, and T. W. Hänsch, "Realization of a new concept for visible frequency division: phase locking of harmonic and sum frequencies," Opt. Lett. **15**, 532–534 (1990).
55. T. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, "Absolute optical frequency measurement of the cesium D-1 line with a mode-locked laser," Phys. Rev. Lett. **82** 3568–3571 (1999).
56. Th. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, "Accurate measurement of large optical frequency differences with a mode-locked laser," Opt. Lett. **24**, 881–883 (1999).
57. S. A. Diddams, Th. Udem, J. C. Bergquist, E. A. Curtis, R. E. Drullinger, L. Hollberg, W. M. Itano, W. D. Lee, C. W. Oates, K. R. Vogel, and D. J. Wineland, "An optical clock based on a single trapped 199Hg+ ion," Science **293**, 825 (2001).
58. J. Stenger, H. Schnatz, C. Tamm, and H. R. Telle, "Ultraprecise measurement of optical frequency ratios," Phys. Rev. Lett. **88**, 073601 (2002).
59. L.-S. Ma, Z. Bi, A. Bartels, L. Robertsson, M. Zucco, R. S. Windeler, G. Wilpers, C. Oates, L. Hollberg, and S. A. Diddams, "Optical frequency synthesis and comparison with uncertainty at the 10–19 level," Science **303**, 1843 (2004).
60. B. C. Young, F. C. Cruz, W. M. Itano, and J. C. Bergquist, "Visible lasers with subhertz linewidths," Phys. Rev. Lett. **82**, 3799–3802 (1999).
61. D. J. Wineland, J. C. Bergquist, J. J. Bollinger, R. E. Drullinger, and W. M. Itano, "Quantum computers and atomic clocks," in*Proceedings of the 6th Symposium on Frequency Standards and Metrology*, P. Gill, ed. (World Scientific, 2002) pp. 361–368.
62. P. O. Schmidt, T. Rosenband, C. Langer, W. M. Itano, J. C. Bergquist, and D. J. Wineland, "Spectroscopy using quantum logic," Science **309**, 749–752 (2005).
63. C. W. Chou, D. B. Hume, J. C. J. Koelemeij, D. J. Wineland, and T. Rosenband, "Frequency comparison of two high-accuracy Al+ optical clocks," Phys. Rev. Lett. **104**, 070802 (2010).
64. J. Ye, H. J. Kimble, and H. Katori, "Quantum state engineering and precision metrology using state-insensitive light traps," Science **320**, 1734–1738 (2008).
65. M. Niering, R. Holzwarth, J. Reichert, P. Pokasov, Th. Udem, M. Weitz, T. W. Hänsch, P. Lemonde, G. Santarelli, M. Abgrall, P. Laurent, C. Salomon, and A. Clairon, "Measurement of the hydrogen 1S-2S transition frequency by phase coherent comparison with a microwave cesium fountain clock," Phys. Rev. Lett. **84**, 5496–5499 (2000).
66. Y. V. Baklanov and V. P. Chebotayev, "Narrow resonances of two-photon absorption of super-narrow pulses in a gas," Appl. Phys. **12** 97–99 (1977).
67. M. J. Snadden, A. S. Bell, E. Riis, and A. I. Ferguson, "Two-photon spectroscopy of laser-cooled Rb using a mode-locked laser," Opt. Commun. **125**, 70–76 (1996).
68. A. Marian, M. C. Stowe, J. R. Lawall, D. Felinto, and J. Ye, "United time-frequency spectroscopy for dynamics and global structure," Science **306**, 2063 (2004).
69. V. Gerginov, C. E. Tanner, S. A. Diddams, A. Bartels, and L. Hollberg, "High resolution spectroscopy with a femtosecond laser frequency comb," Opt. Lett. **30**, 1734–1736 (2005).
70. J. E. Stalnaker, V. Mbele, V. Gerginov, T. M. Fortier, S. A. Diddams, L. Hollberg, and C. E. Tanner, "Femtoseond frequency comb measurement of absolute frequencies and hyperfine coupling constants in cesium vapor," Phys. Rev. A **81**, 043840 (2010).
71. P. Fendel, S. D. Bergeson, Th. Udem, and T. W. Hänsch, "Two-photon frequency comb spectroscopy of the 6 s–8 s transition in cesium," Opt. Lett. **32**, 701–703 (2007).
72. M. J. Thorpe, K. D. Moll, R. J. Jones, B. Safdi, and J. Ye, "Broadband cavity ringdown spectroscopy for sensitive and rapid molecular detection," Science **311**, 1595–1599 (2006).
73. S. Diddams, L. Hollberg, and V. Mbele, "Molecular fingerprinting with spectrally-resolved modes of a femtosecond laser frequency comb," Nature **445**, 627 (2007).
74. M. J. Thorpe and J. Ye, "Cavity-enhanced direct frequency comb spectroscopy," Appl. Phys. B **91**, 397–414 (2008).
75. K. C. Cossel, F. Adler, K. A. Bertness, M. J. Thorpe, J. Feng, M. W. Raynor, and J. Ye, "Analysis of trace impurities in semiconductor gas via cavity-enhanced direct frequency comb spectroscopy," arXiv:1003.1314v1 [physics.optics].
76. S. Schiller, "Spectrometry with frequency combs," Opt. Lett. **27**, 766–768 (2002).
77. F. Keilmann, C. Gohle, and R. Holzwarth, "Time-domain mid-infrared frequency-comb spectrometer," Opt. Lett. **29**, 1542–1544 (2004).
78. I. Coddington, W. C. Swann, and N. R. Newbury, "Coherent multiheterodyne spectroscopy using stabilized optical frequency combs," Phys. Rev. Lett. **100**, 013902 (2008).
79. B. Bernhardt, A. Ozawa, P. Jacquet, M. Jacquey, Y. Kobayashi, T. Udem, R. Holzwarth, G. Guelachvili, T. W. Hansch, and N. Picque, "Cavity-enhanced dual-comb spectroscopy," Nat. Photonics **4**, 55 (2010).
80. R. J. Jones, K. D. Moll, M. J. Thorpe, and J. Ye, "Phase-coherent frequency combs in the vacuum ultraviolet via high-harmonic generation inside a femtosecond enhancement cavity," Phys. Rev. Lett. **94**, 193201 (2005).
81. Ch. Gohle, Th. Udem, J. Rauschenberger, R. Holzwarth, M. Herrmann, H. A. Schussler, F. Krausz, and T. W. Hänsch, "A frequency comb in the extreme ultraviolet," Nature **436**, 234–237 (2005).
82. D. Z. Kandula, C. Gohle, T. J. Pinkert, W. Ubachs, and K. S. E. Eikema, "Extreme ultraviolet frequency comb metrology," arXiv:1004.5110v2 [physics.atom-ph].
83. T. A. Johnson and S. A. Diddams, "Mid-IR frequency comb upconversion spectroscopy," in *Conference on Lasers and Electro-Optics and Quantum Electronics and Laser Science Conference*, OSA Technical Digest (Optical Society of America, 2010), paper CPDB11.
84. A. Pe'er, E. A. Shapiro, M. C. Stowe, M. Shapiro, and J. Ye, "Precise control of molecular dynamics with a femotosecond frequency comb," Phys. Rev. Lett. **98**, 113004 (2007).

85. K. -K. Ni, S Ospelkaus, M. H. G. de Miranda, A. Pe'er, B. Neyenhuis, J. J. Zirbel, S. Kotochigova, P. S. Julienne, D. S. Jin, and J. Ye, "A high phase-space density gas of polar molecules," Science **322**, 231–235 (2008).
86. M. T. Murphy, Th. Udem, R. Holzwarth, A. Sizmann, L. Pasquini, C. Araujo-Hauck, H. Dekker, S. D'Odorico, M. Fischer, T. W. Hansch, and A. Manescau, "High-precision wavelength calibration of astronomical spectrographs with laser frequency combs," Mon. Not. R. Astron. Soc. **380**, 839 (2007).
87. S. Osterman, S. Diddams, M. Beasley, C. Froning, L. Hollberg, P. MacQueen, V. Mbele, and A. Weiner, "A proposed laser frequency comb-based wavelength reference for high-resolution spectroscopy," Proc. SPIE **6693**, 66931G (2007).
88. C.-H. Li, A. J. Benedick, P. Fendel, A. G. Glenday, F. X. Kartner, D. F. Phillips, D. Sasselov, A. Szentgyorgyi, and R. L. Walsworth, "A laser frequency comb that enables radial velocity measurements with a precision of 1 cm s^{-1}," Nature **452**, 610–612 (2008).
89. T. Steinmetz, T. Wilken, C. Araujo-Hauck, R. Holzwarth, T. W. Hänsch, L. Pasquini, A. Manescau, S. D'Odorico, M. T. Murphy, T. Kentischer, W. Schmidt, and T. Udem, "Laser frequency combs for astronomical observations," Science **23**, 1335 (2008).
90. D. Braje, M. Kirchner, S. Osterman, T. Fortier, and S. A. Diddams, "Astronomical spectrograph calibration with broad-spectrum frequency combs," Eur. Phys. J. D **48**, 57–66 (2008).
91. T. Wilken, C. Lovis, A. Manescau, T. Steinmetz, L. Pasquini, G. Lo Curto, T. W. Hänsch, R. Holzwarth, and Th. Udem, "High-precision calibration of spectrographs," Mon. Not. R. Astron. Soc. **405**, L16–L20 (2010).
92. A. J. Benedick, G. Chang, J. R. Birge, L. Chen, A. G. Glenday, C. Li, D. F. Phillips, A. Szentgyorgyi, S. Korzennik, G. Furesz, R. L. Walsworth, and F. X. Kärtner, "Visible wavelength astro-comb," Opt. Express **18**, 19175–19184 (2010).
93. F. Quinlan, G. Ycas, S. Osterman, and S. A. Diddams, "A 12.5 GHz-spaced optical frequency comb spanning >400 nm for infrared astronomical spectrograph calibration," Rev. Sci. Inst. **81**, 063105 (2010).
94. see, for example, http://www.psi.com.au/. Mention of specific products is for scientific communication only and does not constitute an endorsement by NIST.
95. E. Ivanov and M. E. Tobar, "Low phase-noise sapphire crystal microwave oscillators: Current status," IEEE Trans. Ultrason. Ferroelectr. Freq. Control **56**, 263 (2009).
96. S. Grop, P.-Y. Bourgeois, R. Boudot, Y. Kersale, E. Rubiola, and V. Giordano, "10 GHz cryocooled sapphire oscillator with extremely low phase noise," Electron. Lett. **46**, 420–421 (2010).
97. J. J. McFerran, E. N. Ivanov, G. Wilpers, C. W. Oates, S. A. Diddams, and L. Hollberg, "Low noise synthesis of microwave signals from an optical source," Electron. Lett. **41**, 36–37 (2005).
98. A. Bartels, S. A. Diddams, C. W. Oates, G. Wilpers, J. C. Bergquist, W. Oskay, and L. Hollberg, "Femtosecond laser based synthesis of ultrastable microwave signals from optical frequency references," Opt. Lett. **30**, 667–669 (2005).
99. W. Zhang, Z. Xu, M. Lours, R. Boudot, Y. Kersalé, G. Santarelli, and Y. Le Coq, "Sub-100 attoseconds stability optics-to-microwave synchronization," Appl. Phys. Lett. **96**, 211105 (2010).
100. S. A. Diddams, M. Kirchner, T. Fortier, D. Braje, A. M. Weiner, and L. Hollberg, "Improved signal-to-noise ratio of 10 GHz microwave signals generated with a mode-filtered femtosecond laser frequency comb," Opt. Express **17**, 3331–3340 (2009).
101. Z. Jiang, C.-B. Huang, D. E. Leaird, and A. M. Weiner, "Optical arbitrary waveform processing of more than 100 spectral comb lines," Nat. Photonics **1**, 463–467 (2007).
102. N. K. Fontaine, R. P. Scott, J. Cao, A. Karalar, K. Okamoto, J. P. Heritage, B. H. Kolner, and S. J. B. Yoo, "32 phase $\times$ 32 amplitude optical arbitrary waveform generation," Opt. Lett. **32**, 865–867 (2007).