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The measurement of optical frequencies*

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Abstract

Surprising as it might seem, it is possible to phase-coherently track, synthesize, count and divide optical frequencies of visible laser sources. In essence, the technologies described here now allow direct connection of basically any frequency from DC to 1000 THz. Modern 'self-referenced' femtosecond mode-locked lasers have enormously simplified the required technology. These revolutionary new systems build on a long history of optical frequency metrology that spans from the early days of the laser. The latest systems rely heavily on technologies previously developed for laser frequency stabilization, optical phase-locked-loops, nonlinear mixing, ultra-fast optics and precision opto-electronic metrology. Using examples we summarize some of the heroic efforts that led to the successful development of harmonic optical frequency chains. Those systems played critical roles in defining the speed of light and in redefining the metre. We then describe the present state-of-the-art technology in femtosecond laser frequency combs, their extraordinary performance capabilities and some of the latest results.

(Some figures in this article are in colour only in the electronic version)

Acronyms

BIPM	Bureau International des Poids et Mesures, Sèvres, France
CGPM	Conférence Générale des Poids et Mesures
CIPM	Comité International des Poids et Mesures
BNM-SYRTE	Bureau National de Métrologie - Systèmes de Référence Temps-Espace, Paris, France
FIR	Far-infrared
LMR	Laser magnetic resonance
LPTF	Laboratoire Première du Temps et des Fréquences (now BNM-SYRTE), Paris, France
MIM	Metal–insulator–metal diode
NBS	National Bureau of Standards (now NIST), USA
NIST	National Institute of Standards and Technology, Boulder, USA
Novosibirsk	Russian Academy of Sciences, Novosibirsk Institute of Thermophysics, Novosibirsk Institute of Laser Physics, Novosibirsk
NPL	National Physical Laboratory, Teddington, UK
NRC	Institute for National Measurement Standards, National Research Council, Ottawa, Canada
NRLM	National Research Laboratory of Metrology, now NMIJ-AIST (National Metrology Institute of Japan—Advanced Industry Science and Technology), Tsukuba, Japan
PTB	Physikalisch-Technische Bundesanstalt, Braunschweig, Germany
SI	Système International
Tu-FIR	Tunable far-infrared spectroscopy
VNIIFTRI	Institute of Time and Space Metrology, All-Russia Scientific-Research Institute of Physicotechnical and Radio Engineering Measurements (VNIIFTRI), Mendeleevo, Russia

1. Optical frequencies, why bother?

For more than 100 years optical atomic frequency references have played critical roles in basic science, in precision measurements and in a few technical applications [1–10]. Absorption and emission spectra of atoms and molecules

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provide necessary information to understand atomic structure and dynamics, and precision spectroscopy has been central to the development of quantum mechanics and modern atomic/molecular science. Select atomic and molecular transitions have provided, and continue to provide, convenient and highly accurate optical and infrared (IR) references for length metrology and, more recently, for optical frequency standards. With the SI unit of 'time' defined (since 1967) in terms of the frequency of the caesium clock transition at $\sim$9.2 GHz, and the unit of length defined (since 1960) in terms of the ^{86}Kr wavelength of 605.7 nm, it was natural to look towards the possibility of connecting the two base units of length and time through the simple relationship for the speed of light, $c = \lambda\nu$, where λ is the wavelength and ν is the frequency. This could be achieved with straightforward methods at microwave frequencies [11], but that technique did not provide a direct connection to the definition of the metre in the visible spectrum. Some interferometric optical methods were explored to make this connection. For example, it was possible to use frequency-modulated lasers with high-finesse Fabry–Perot cavities to infer optical wavelengths from RF sidebands imposed on an optical carrier, as demonstrated by Bay *et al* and DeVoe *et al* [12, 13]. Those advanced interferometric methods gave precise relative wavelengths between the optical carrier and its RF sidebands, but they were not actual optical frequency measurements. High-accuracy optical interferometry was critical for dimensional metrology, establishment of optical wavelength references and the ultimate redefinition of the metre [14–16]. The highest performance optical interferometers achieved fractional uncertainties in wavelength ratios of about 3×10^{-11} at the NPL under Rowley and collaborators [5, 6, 17–19]. (Note, acronyms are defined earlier.) But, unfortunately, optical frequencies could not be measured directly during that era. In the 1970s several national laboratories began research programmes to develop the technologies to measure optical frequencies and make the connection between microwave atomic frequency standards and optical atomic frequencies/wavelengths.

Even the suggestion of counting optical frequencies in the visible range ($\sim$500 THz) must have seemed ludicrous to many in the past, because traditional methods of frequency metrology failed to work above about 100 GHz. Nonetheless, with some vision and new ideas a few researchers had the will to try. This feat was finally accomplished in the 1970s and 1980s and resulted in a defined speed of light and a redefinition of the unit of length, the metre. Of particular importance was the goal of making a coherent connection of optical frequencies to microwaves sources and electronics. An even more powerful and useful tool is the recently developed capability to connect any arbitrary frequency to any other frequency from DC to 1000 THz ('DC to daylight' as is commonly heard). With technologies developed over the past 40 years this is now possible and is the subject of this paper.

As technology advances, information must move at higher data rates, demanding faster electronics, more precise timing and synchronization over longer physical distances. This in turn demands higher frequency oscillators and clocks. The natural extension of those ideas led to optical frequency references and optical atomic clocks [20–29]. (See also the paper by Gill in this issue.) Optical atomic clocks have significant advantages over their microwave counterparts and are now the major driving force behind the present explosive growth in optical frequency measurements and technology. Of course, if an optical atomic clock 'ticks' with a period of a femtosecond it requires a frequency counter that is fast enough to follow the femtosecond oscillations and to synchronously record, divide and distribute the timing information.

The field of optical frequency metrology is now a hot research topic and has seen tremendous progress in the last five years because of the development of new ideas based on mode-locked femtosecond laser technology, nonlinear optics and precision control. Not surprisingly, with all the research activity and exciting new results, the field has garnered considerable attention in the scientific literature. We encourage all to explore some excellent reviews that provide different perspectives ranging from historical time-lines [8, 10, 30–35] to summaries of the latest results [9, 25, 36–40]. There are even two books devoted to the technology and applications of optical frequency metrology [41, 42]. Equally interesting are key early papers on optical frequency measurements that contain a wealth of information along with the unique perspective of their time [5, 8, 10, 18, 21, 30, 31, 43–50].

2. The development of optical frequency measuring systems

Not long after the laser was invented, its coherence properties were explored and heterodyne beatnotes were detected between independent gas lasers. Those experiments showed that lasers [34, 51, 52] could have excellent spectral and spatial coherence with relatively narrow linewidths, $\Delta\nu$, and correspondingly high quality factors, $Q = \nu/\Delta\nu$.

Starting down the path, or rather upwards, towards measuring optical frequencies in terms of the caesium primary frequency standard, it was natural to start with the highest-frequency oscillators that could be phase-coherently controlled and then multiply those sources up to higher frequencies using nonlinear devices. For this purpose, new frequency multiplication methods were developed that could operate at millimetre-wave and FIR frequencies. From the 1960s through 1990s the approach was to start with high-frequency vacuum tube oscillators such as klystrons and backward wave oscillators (BWOs) operating in the millimetre-wave range (about 90 GHz to 300 GHz). With some effort, these sources could be phase-locked to lower-frequency microwave sources by means of microwave harmonic mixing methods. In turn, the lower-frequency microwave oscillators could be referenced to stable quartz-crystal oscillators, which could be steered to caesium atomic frequency standards on longer time scales. Going upwards in frequency from the 100 GHz to 200 GHz range required new custom nonlinear mixing and harmonic generating methods to reach fixed frequencies of FIR lasers in the sub-millimetre and FIR domain. Perhaps the first actual measurement of a laser frequency using frequency metrology rather than interferometry (and knowledge of the speed of light) was when Hocker *et al* [53] measured the frequencies of HCN laser lines that have very low laser frequencies of 890 GHz and 964 GHz (337 μm and 311 μm, respectively). This was accomplished by using a silicon ‘cat-whisker’ harmonic mixer in a waveguide to detect the beatnote between the laser and harmonics of a 75 GHz klystron. They achieved a fractional frequency uncertainty of a few parts in 10^7.

With hopes of reaching higher frequencies, and the visible range in particular, without requiring too many stable oscillators to span the electromagnetic spectrum, it was necessary to use the maximum frequency multiplication factors that were feasible. The search was on to find the fastest nonlinear mixers that could operate at millimetre and FIR frequencies, and this led to three interesting and important devices: whisker-contacted Schottky diodes, point-contact MIM diodes and superconducting Josephson mixers [24, 31, 34, 54]. Particularly useful were the special commercial Schottky diodes consisting of small gold islands on GaAs wafers that could be contacted with tiny metal whiskers and could generate harmonics of input microwave frequencies (e.g. 5 GHz to 90 GHz) up to about 5 THz [55–57]. Some work done by Prevedelli *et al* [58] showed that microwave harmonic orders up to ∼200 could be generated using a microwave-driven Schottky diode, and the resulting harmonic signals could be detected by using a methanol based FIR laser as a local oscillator at 2.5 THz. Nonlinear mixers such as Schottky diodes, MIM diodes and superconducting Josephson devices also proved useful in measuring large difference frequencies between lasers in the FIR, IR and visible ranges. Blaney and Knight [59] even detected the 825th harmonic of a 1 GHz signal using a Josephson junction mixer. In some cases the diodes could serve simultaneously as the optical detector/mixer and as a microwave harmonic generator. Thus, the difference between two laser frequencies ν_{L1}, ν_{L2} can be mixed with the mth harmonic of the microwave frequency, $\nu_{\mu\mathrm{wave}}$, producing a low-frequency IF output signal at $f_{\mathrm{IF}} = (\nu_{\mathrm{L1}} - \nu_{\mathrm{L2}}) \pm m\nu_{\mu\mathrm{wave}}$. With these techniques, using MIM and Schottky diodes, it was possible to detect laser difference frequencies in the visible range as high as 2.5 THz [60, 61] and also to phase-lock diode lasers with offset frequencies as large as 360 GHz [62, 63].

Above the range of ∼2 THz to 5 THz that was accessible with harmonics of microwave sources, the optical frequency chains were forced away from electronic oscillators and into the then-uncharted domain of FIR lasers based on simulated emission from molecules. Most optical frequency chains relied on both directly excited and optically pumped FIR lasers to cover the frequency gap from a few terahertz up to the strong CO_2 lasers at 30 THz. More details on FIR-laser technologies can be found in [34, 56, 64]. At higher frequencies Schottky diodes were not the best mixers, and it was more common to shift to MIM diodes for optical mixing and optical harmonic generation. The elegant point-contact MIM diodes are truly remarkable devices, and were constructed simply by contacting the sharpened point of a tiny (25 μm diameter) tungsten wire on a smooth nickel surface (figure 1). The tungsten and nickel are separated by a very thin oxide layer that naturally forms on the nickel surface and acts like a nonlinear tunnel barrier.

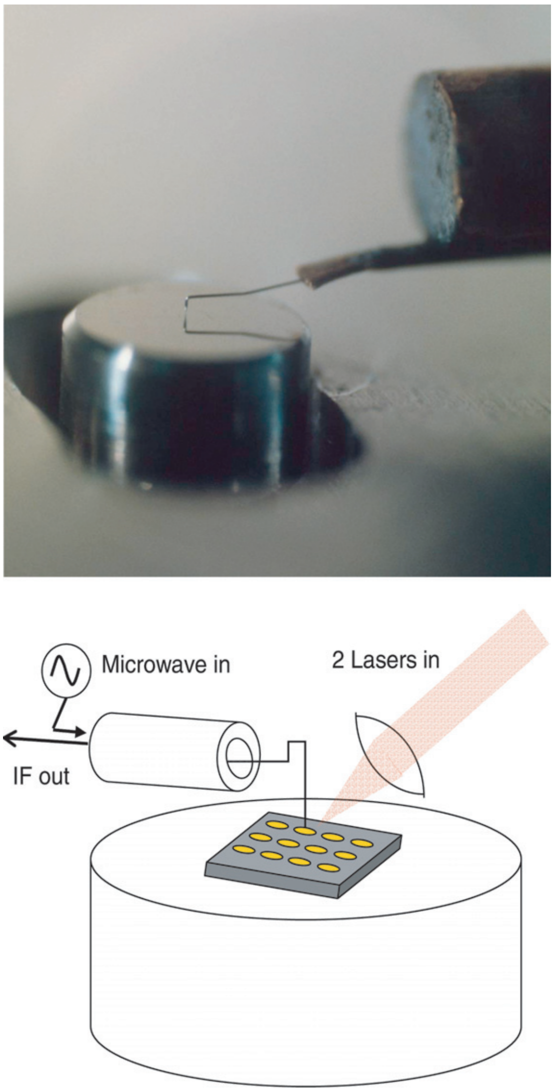

Figure 1. The photo shows a MIM diode constructed from a simple nickel post (near bottom centre, ∼3 mm diameter) that is contacted by the sharpened point of a ∼25 μm diameter tungsten wire. Laser light will be focused onto the point of contact. The sketch represents a simplified diagram of a whisker-contacted Schottky diode consisting of many tiny (typically a few micrometres in diameter) gold islands on a GaAs substrate mounted on a metal post. A tungsten whisker contacts a single diode element at a time and carries the IF signal out through a coaxial line that also brings microwaves onto the diode. Two laser beams are combined and focused on the tiny Schottky diode.

Unfortunately, at least in their common forms, MIM diodes are somewhat finicky devices and require considerable experimental tweaking for use at the highest frequencies. The nonlinear signals that they produce at the highest frequencies are small in amplitude; perhaps this is not so unreasonable, given the fact that they are microscopic devices that are driven with high-intensity laser fields to generate nonlinear mixing products at the optical frequencies. The theoretical understanding of MIM diodes is, at best, incomplete. Nonetheless, they are amazing nonlinear devices and probably represent the world’s fastest nonlinear mixers with the broadest bandwidths, having demonstrated usable mixing signals from DC to almost to 200 THz [31, 34, 65].

To coherently generate a visible laser frequency near 500 THz starting from a 1 THz source and relying only on successive stages of second-harmonic generation would require $2^N = 500$, or $N \approx 9$, stages. With judicious choices of

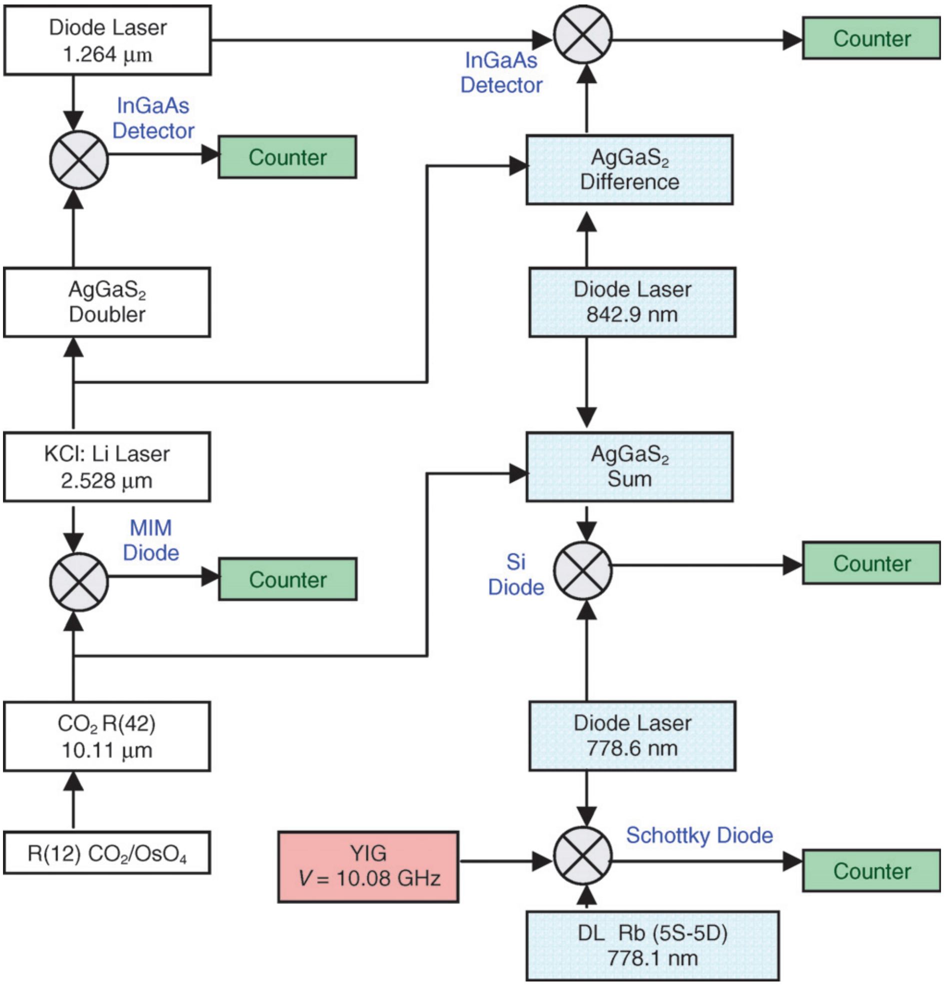

Figure 2. Simplified diagram of the optical frequency chain used at LPTF-Paris (now BNM-SYRTE) to measure the frequency of the two-photon transition in rubidium at 385 THz (778 nm). This chain did not span all the way to the Cs frequency standard but was referenced to the high-performance OsO_4 frequency standard at 29 THz that was previously measured relative to the Cs standard. (This figure adapted with permission from [35, 180].)

FIR lasers and mixers it was possible to achieve higher-order mixing (e.g. ×10 or ×12) in the terahertz range. This meant that is was possible to jump in a single step from a FIR laser at ~1 THz to another FIR laser at ~10 THz. With two additional mixing orders, the 10 THz signal can be multiplied up to the 30 THz frequency of the convenient, powerful and stable CO_2 lasers [66].

At frequencies above 30 THz, MIM diode mixers are suitable for some mixing and harmonic generating applications (e.g. $3 \times 30\,\mathrm{THz} = 90\,\mathrm{THz}$), but nonlinear optical crystals can be very effective alternatives because they can be phase-matched for efficient mixing of specific desired frequencies/wavelengths. Figure 2 shows a frequency chain from LPTF that used a variety of lasers, a combination of nonlinear crystals, semiconductor diodes and MIM diodes for mixing frequencies between 30 THz and 385 THz.

Regrettably, at higher frequencies no convenient nonlinear mixing elements were found that could generate continuous-wave (CW) high harmonics. Lacking that, the mixing orders were necessarily low, and optical frequency chains required several stable laser sources spanning the frequency range and all locked together with phase-locked-loops—hence the analogy of a ‘chain’ with interlocking loops. Figures 3–5 show three harmonic optical frequency chains that used these approaches to reach important optical frequency references directly from microwave frequency sources. In 1983, Jennings and collaborators at NIST were the first to successfully measure an optical frequency in the visible range (near 520 THz) using a chain similar to the one shown in figure 3. Unfortunately, the NIST frequency chains were not maintained in an operating mode much beyond that date. Nonetheless, other laboratories, including NRC, NPL, PTB, LPTF (now BNM-SYRTE), VNIIFTRI, Novosibirsk and NRLM continued to develop and refine optical frequency measurement methods and synthesis chains that were used to measure and evaluate optical frequency standards [21, 34, 44–46, 51, 52, 67–72].

Whitford at NRC came up with a very different approach to optical frequency synthesis [73–75]. His optical frequency synthesizer relied solely on a combination of five different CO_2 lasers whose frequencies could be related to each other by rational fractions. The original versions of that IR frequency synthesizer could be used to establish connections between microwave frequencies sources and eventually reached the 88 THz CH_4 frequency standard. More recently, a team at NRC

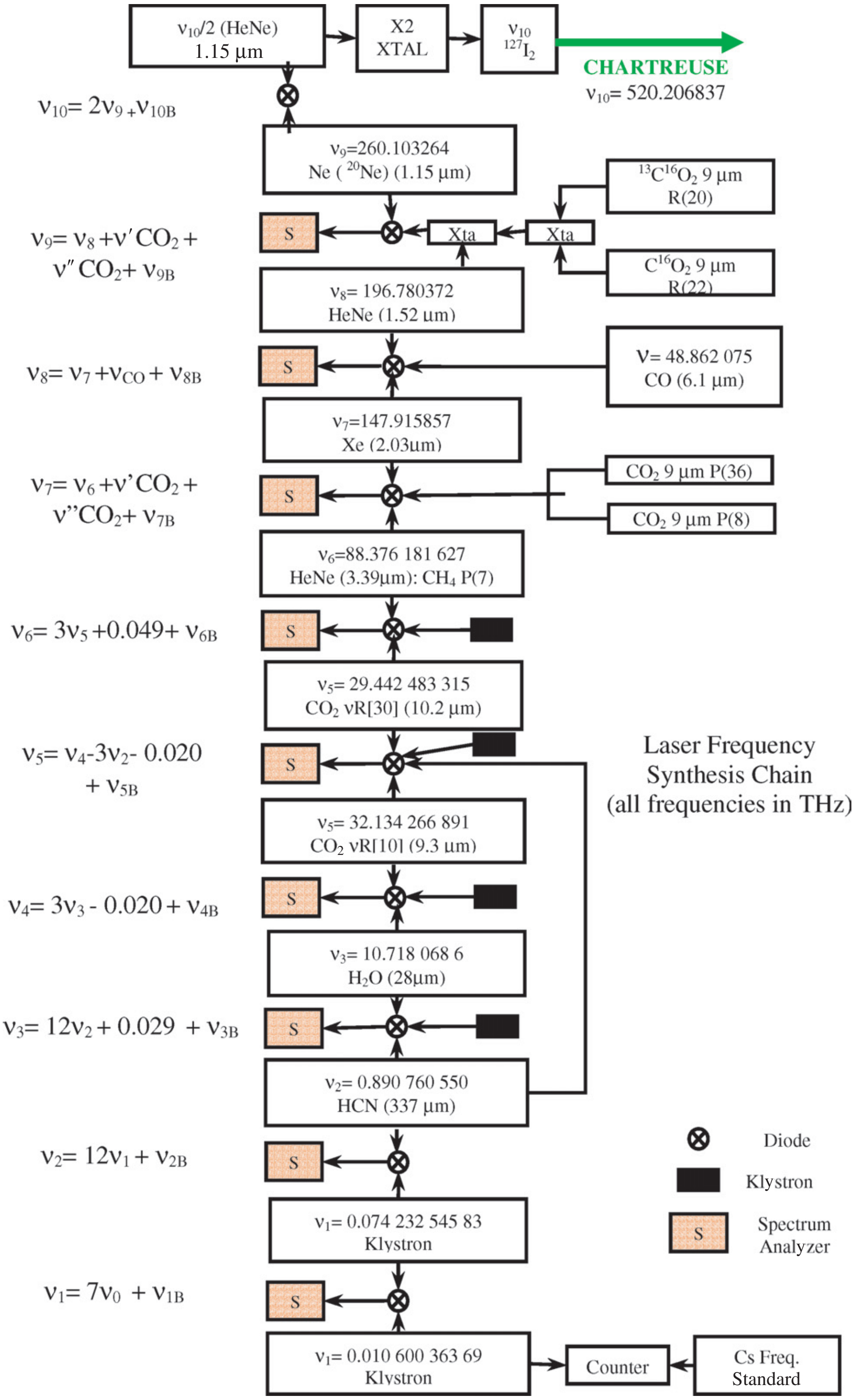

Figure 3. This harmonic frequency chain developed at NIST measured the frequency of an iodine-stabilized dye laser in the visible range (520 THz) relative to the caesium primary frequency standard. (Adapted with permission from NIST [181].)

extended the CO_2-laser-based optical frequency synthesizer to reach the visible Sr^+ frequency standard at 445 THz (674 nm) as shown in figure 6 [76–78]. Through the years the NRC chain was used to make several important optical frequency measurements [25, 79–83].

Because of their complexity and specialized applications, only a small number of optical frequency chains were ever constructed. Those systems existed primarily at national standards laboratories and were used to make high-accuracy frequency measurements of lasers stabilized to atomic and/or

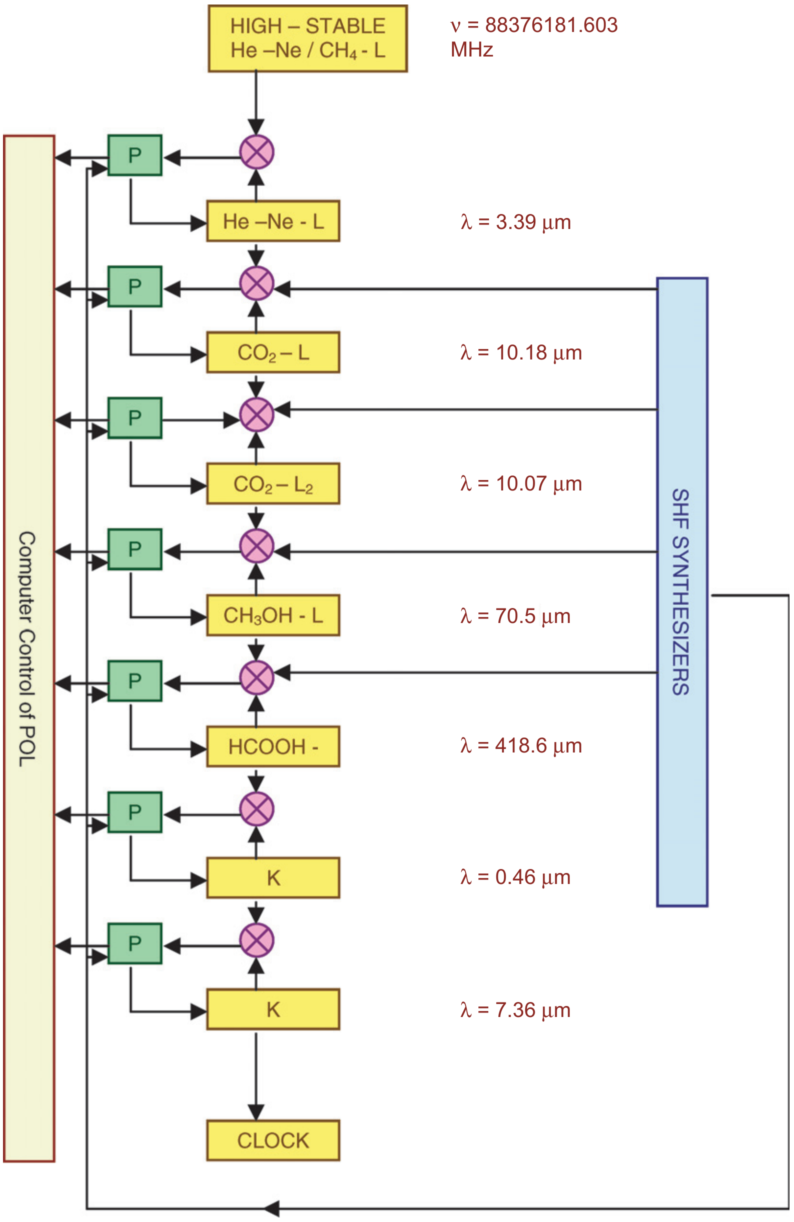

Figure 4. Harmonic optical frequency chain developed by Chebotayev's group at Novosibirsk and configured to operate as an optical time scale, i.e. an 'optical clock'. The frequency chain was controlled from a CH_4 frequency standard at 88 THz (3.39 μm) at the top and delivered an output at a RF/microwave frequency. (Adapted with permission from [20].)

molecular resonances. A plot of the relative frequency uncertainty achieved in optical frequency measurements through the years is shown in figure 7.

Noteworthy among these were measurements of the metrologically and scientifically important optical frequency references based on OsO_4 and SF_6 at ∼30 THz (10 μm), CH_4 at ∼88 THz (3.39 μm) and I_2 at 473 THz (633 nm) [5, 6, 21, 25, 34, 44–46, 51, 52, 67–71, 73–83]. These molecular resonances were selected because they overlapped the convenient CO_2 and HeNe gas lasers, and they have sharp resonance transitions that provided good stability and frequency reproducibility. Considerable effort focused on the CH_4- and I_2-referenced HeNe lasers because of their metrological importance and widespread use. Measurements of those two standards provided the basis for the highest-accuracy determinations of the speed of light and ultimately resulted in the definition of the speed of light by the CGPM in 1983 [7, 84]. Careful measurements of both the frequency and wavelength of the I_2-stabilized 633 HeNe laser then resulted in the redefinition of the SI unit of length, the metre, in terms of the defined speed of light. The CGPM also authorized (through the CIPM) three recommended realizations of the new unit of length (in vacuum), which are approximately as follows:

(1) the length travel by light in a measured time interval,
(2) through the wavelength of an electromagnetic wave of a measured frequency traceable to the Cs frequency, and
(3) from the known wavelengths of a few specific approved optical frequency references [7, 84, 85].

Optical frequency measurements are obviously a critical technology that is central to our system of units.

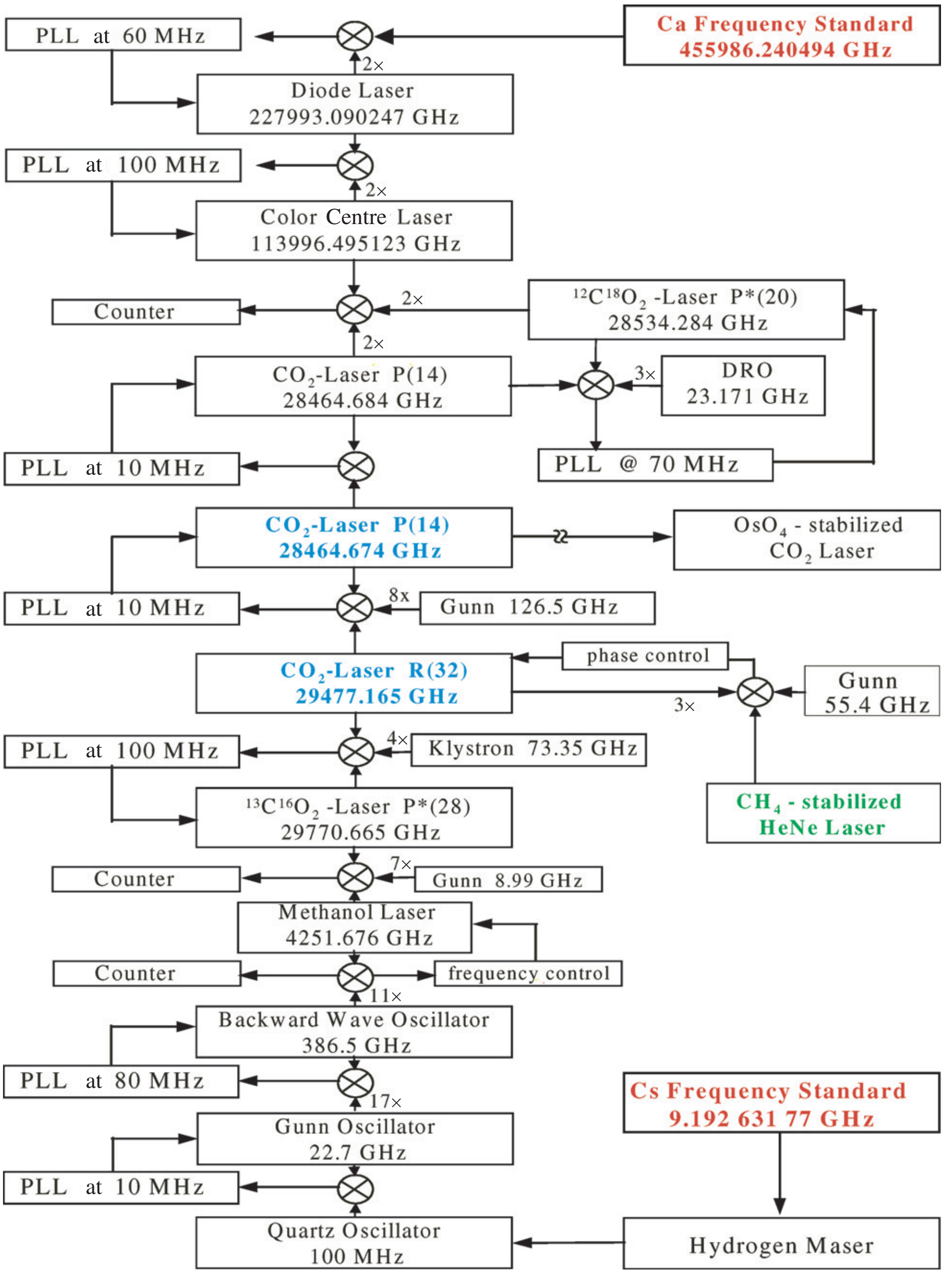

Figure 5. The PTB harmonic frequency chain was the first to achieve a phase-coherent connection between the caesium primary standard and an optical-frequency reference in the visible range. In this case, the target was the calcium optical frequency standard at 457 THz (657 nm). (Adapted with permission from [96].)

A novel way around the problem of not having any efficient nonlinear mixers for low-power CW lasers was the concept from Klement'ev *et al* [86], who proposed using resonant interactions in atoms to efficiently sum three optical frequencies to generate a fourth. Specifically, they proposed using Ne atomic resonances to sum the well-known HeNe laser lines at 3.39 µm, 1.15 µm and 1.5 µm in order to generate the 633 nm laser line as shown in figure 8. The advantage is that resonant atomic nonlinear mixing can be 8 to 10 orders of magnitude stronger than that found in bulk nonlinear optical materials. The obvious disadvantage is that the optical mixing is not broadband, and so only very specific frequencies can be generated. The four-photon optical mixing approach of figure 8 was demonstrated in Russia [86] and was also used at NIST for the measurement of the 633 nm HeNe frequency [31].

To take advantage of the higher stability and accuracy promised by optical references it was envisioned that optical frequency chains would be referenced and controlled from the high-frequency optical end of the chain. This would provide an output at the low-frequency end that is coherent with the optical reference but which can be connected to electronic sources and counted. The long-term vision to develop high-accuracy, high-stability optical clocks continues today. (Also, see the paper by Gill in this issue.) Proof-of-principle demonstrations of optical molecular clocks were first done in Novosibirsk [87] and at PTB [21]; both used the 88 THz

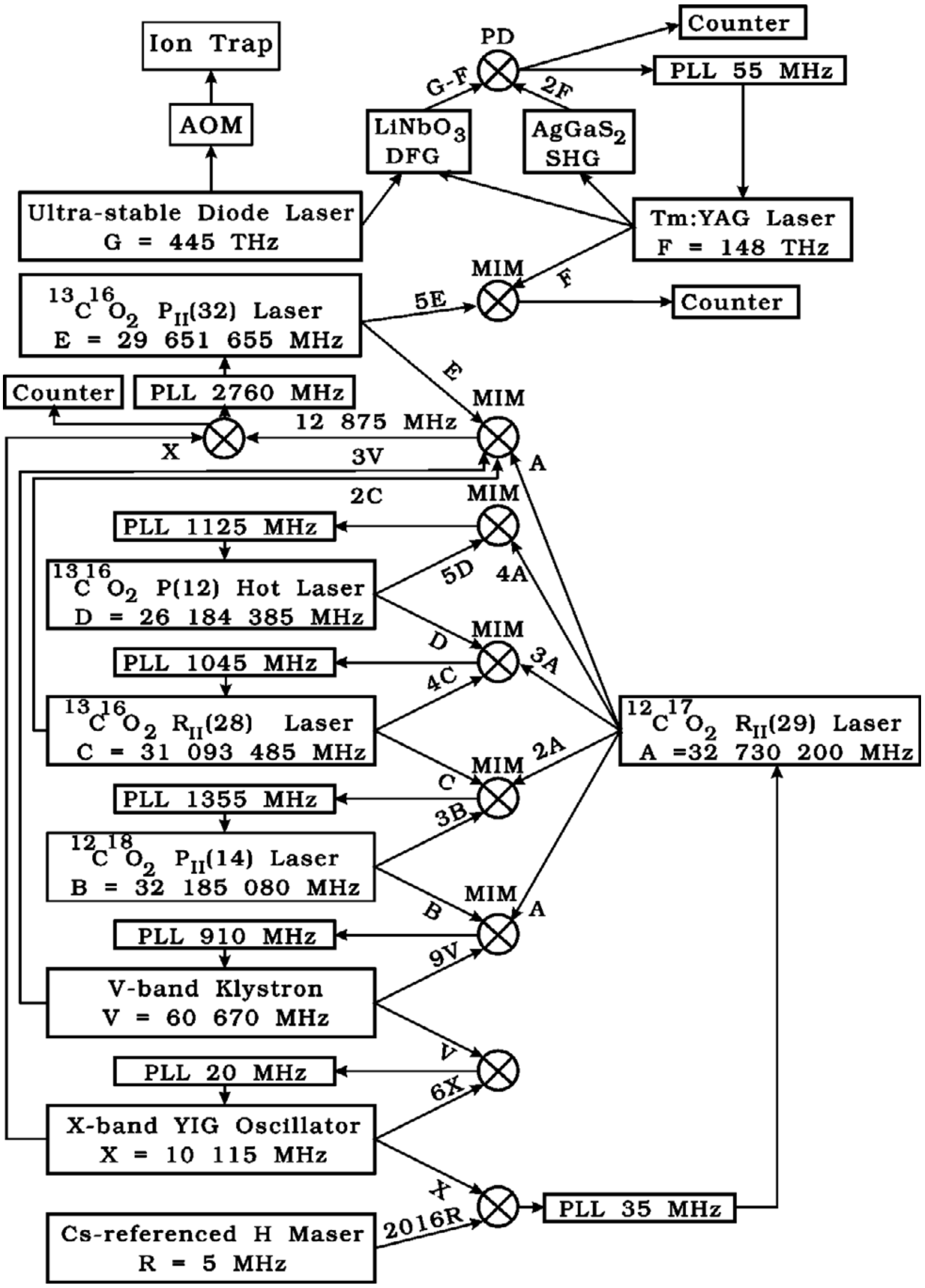

Figure 6. The NRC optical frequency chain as it was configured to measure the Sr^+ optical frequency reference at 445 THz (674 nm) relative to the caesium standard. (Figure used with permission from [78].)

CH_4–HeNe optical frequency standard. More recently, optical atomic clocks have been demonstrated using I_2 [28] and cold atoms/ions [23, 88–90]. The dramatic improvement in fractional frequency uncertainty of optical frequency standards and measurements beginning in about 2000 (see figure 7) was the result of the higher performance achieved with optical-frequency references based on laser-cooled atoms/ions, and the development of optical-frequency comb technology (section 5). Cold atom/ion optical-frequency standards are now being developed in many laboratories around the world, and the femtosecond optical frequency combs provide practical optical synthesizers and frequency dividers.

The other major force pushing the development of optical-frequency measurements has been precision atomic spectroscopy and fundamental physics. Particularly important is the spectroscopy of hydrogen, which provides data that constrain quantum electrodynamic theory [45, 91–94].

3. Alternative optical frequency synthesis schemes

In the 1970s to 1990s, optical frequency chains produced landmark results in optical science and technology, including proof-of-principle demonstrations that visible frequencies could indeed be connected coherently to microwave

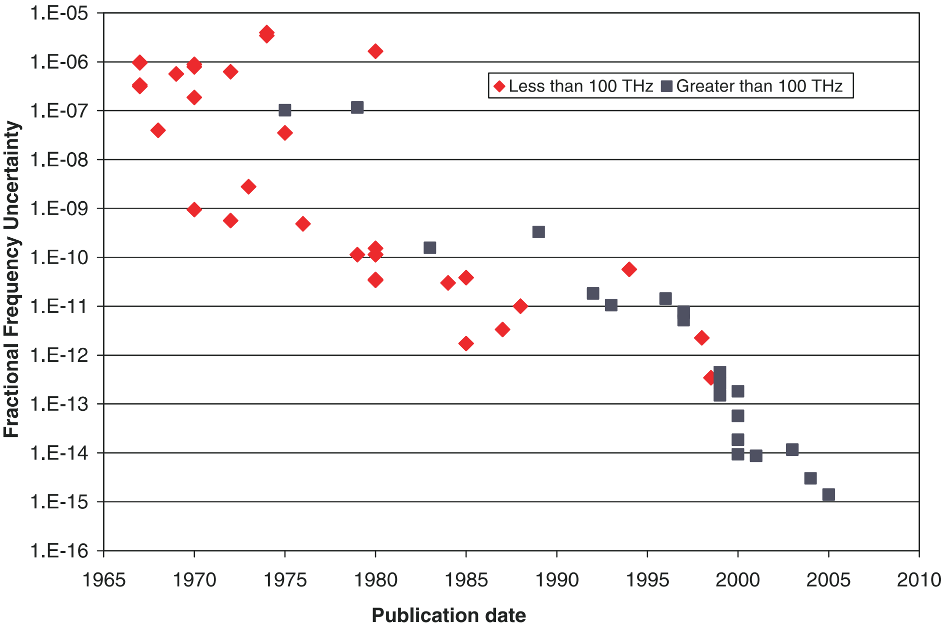

Figure 7. The fractional frequency uncertainty of laser-based frequency references is plotted as a function of the measurement date. The first data point in 1967 was the measurement by Hocker *et al* [53] of two FIR-laser lines near 900 GHz. The first visible measurements were the 1983 measurements of the 520 THz and 473 THz frequencies by Jennings and collaborators [95, 182]. The measurements plotted here are absolute frequency measurements, and most were referenced to caesium frequency standards, although a few were referenced to other better-known optical standards. The data points up to 1999 represent nearly the entire list of high-accuracy optical frequency measurements that were made. After 1999 the optical frequency combs arrived and many new optical-frequency measurements were made, and so we have selected data that are representative of the state-of-the-art from that date forwards.

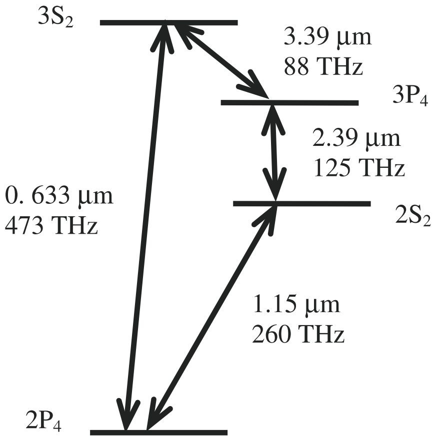

Figure 8. Using atomic resonances for optical mixing was proposed by Klement'ev *et al* [86], and first demonstrated in Russia using the Ne transitions shown here. This scheme was also implemented by a team at NBS using 8 m long HeNe gain tubes in a measurement of the frequency of the 473 THz (633 nm) laser [31, 95]. (A photo of those very long gain tubes can be found in a review paper by Hall [10].)

sources [95, 96], the highest precision measurements of the speed of light and redefinition of the metre, and serve as the measuring instruments that calibrated optical frequency standards for precision physics experiments. Unfortunately, optical frequency chains were large, complex and difficult to operate.

Motivated by a growing need for high-accuracy optical-frequency measurements for precision spectroscopy of hydrogen and the vision of optical atomic clocks based on laser-cooled atoms, alternative approaches for optical frequency synthesis were required. Several ideas were proposed to get around the severe limitations of the traditional optical frequency chains. It was desirable to eliminate the complex mixing stages, troublesome nonlinear elements and the large non-tunable gas lasers in the IR and FIR frequency chains. Some of the proposed methods included Hänsch's idea of using broad bandwidth mode-locked lasers to span large optical frequency intervals (more on this below, as it turned out to be the winner), and the electro-optic modulator-based optical comb generators developed by Kourogi *et al* [97–101] (see figure 9).

The other very promising method was the optical frequency-bisector concept proposed by Telle *et al* [102], where optical frequency intervals are divided successively in half, so that the system could eventually divide an optical frequency down to the countable microwave range. A diagram of a single-stage optical bisector is shown in figure 10.

Some alternative approaches were also explored that would connect optical frequencies with known fractional integer ratios (e.g. $\frac{1}{4}, \frac{1}{2}, \frac{1}{3}, \frac{2}{3}$) [103–106]. Most of the recent efforts tried to operate in the visible or near visible region where convenient tunable lasers and low-cost diode lasers are readily available. Some of these systems were reminiscent of the NRC CO_2 laser frequency synthesizer. A very promising approach was to use optical parametric oscillators interlocked with fractional ratios as advocated by Wong and co-workers [107–110]. Initial experiments showed that these basic principles were all viable at some level. A few of the alternative methods were able to demonstrate a connection between laser harmonics, and some were even used in experiments that measured relatively large optical frequency intervals [111–114]. However, none

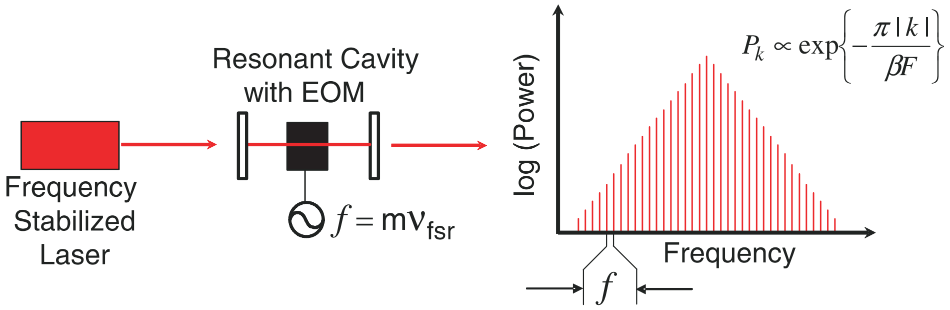

Figure 9. Electro-optic modulator-based optical frequency comb generator. An electro-optic crystal is contained within a Fabry–Perot cavity and simultaneously in a microwave resonator. The laser field is resonated inside the Fabry–Perot cavity, whose free-spectral range, ν_{fsr}, is set to match the microwave frequency, f (or a subharmonic thereof). These systems have been used to generate optical-frequency combs with spectral widths up to ~40 THz [98, 183].

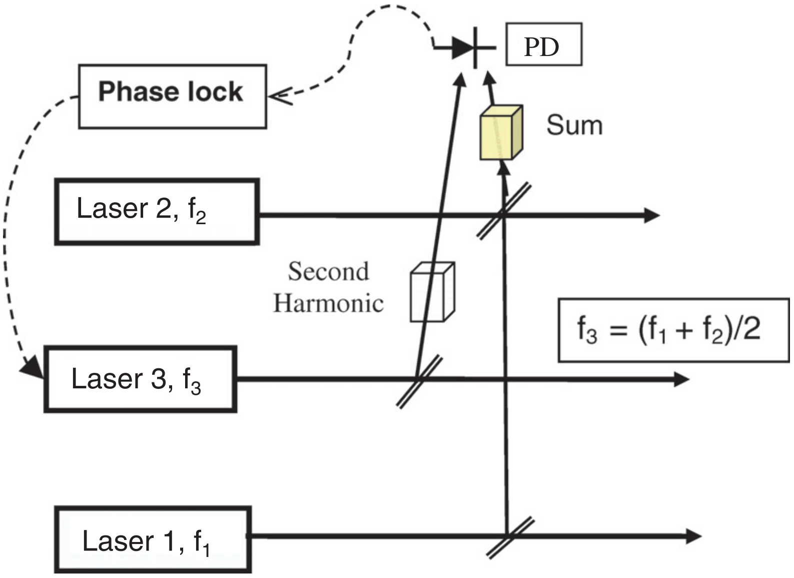

Figure 10. The optical-frequency bisector approach proposed by Telle *et al* uses a phase-locked-loop to force the frequency of laser 3 to be exactly at the frequency mid-point between lasers 1 and 2.

of the alternatives to the harmonic frequency chains were ever used to construct a frequency synthesizer that spanned from microwave to optical frequencies.

4. Some spin-offs

Along the way to the development of optical frequency-measuring systems, important discoveries were made and several technologies developed that had an impact on other fields of research. These include ultra-fast MIM diodes, Schottky diode optical mixers, FIR-laser technologies and nonlinear optics. In fact, one of the major spin-offs of this endeavour was that the development of stable laser sources throughout the electromagnetic spectrum provided high-quality sources that were used for atomic and molecular spectroscopy in the millimetre-wave, FIR, IR, near-IR and visible ranges. Results from precision IR spectroscopy have proved invaluable in providing an understanding of molecular structure and chemistry of radicals, pollutants and atmospheric constituents. An excellent example of a technological spin-off from optical frequency metrology is the research done by Wells and Maki, who used a handful of frequency-stabilized IR lasers with accurately known frequencies to measure molecular absorption spectra of several molecules. From precise experimental measurements and molecular modelling, they were able to determine molecular constants and transition energies for more then 30 000 IR molecular transitions in the spectral region from 488 cm^{-1} to 4400 cm^{-1} (14.6 THz to 132 THz) with precisions of ~8 digits (data are now accessible online at http://physics.nist.gov/PhysRefData/wavenum/html/contents.html). Similarly, CO_2 and FIR lasers were used at NBS/NIST by Evenson and his many collaborators to determine precise molecular frequencies and structures for a very large number of molecular species and radicals. (Most references are accessible at http://tf.nist.gov/general/publications.htm.) Two spectroscopic methods from Evenson's lab derived from optical frequency chains are Tu-FIR laser spectroscopy and LMR. Tu-FIR uses an MIM diode as a mixer to generate the difference frequency between two CO_2 lasers, plus a tunable microwave synthesizer to generate and radiate precisely tunable FIR beams into free space. The Tu-FIR beam proved extremely useful for molecular spectroscopy in the FIR. On the other hand, LMR uses large magnetic fields to tune molecular transitions into resonance with the not very tunable IR and FIR gas lasers. Perhaps the most important spin-offs have been the plethora of laser frequency stabilization methods that have been developed, including stabilization to Fabry–Perot reference cavities, the Pound–Drever–Hall method, modulation locks, optical phase-locked-loops, stabilization of lasers to atomic and molecular transitions and the general techniques of precision optical metrology. These technologies are now in widespread use beyond the field of optical frequency measurements, such as in gravity-wave interferometers.

With the advent of femtosecond-laser-based optical frequency synthesis methods (described in the next section) there is an amazing new synergy between precision laser spectroscopy, metrology and ultra-fast science. This brings new perspectives, understandings and technologies that are beneficial to all. Stabilization of optical frequency combs also brings unprecedented precision and control to the time domain, resulting in subfemtosecond timing jitter, control of the carrier-envelope phase and low-phase-noise microwaves, just to name a few.

5. Mode-locked lasers for optical frequency measurements

The use of a mode-locked laser as a tool for optical frequency metrology was first demonstrated with picosecond lasers by Hänsch and co-workers in the late 1970s [115]. In this classic comb paper, a synchronously pumped mode-locked dye laser was employed to measure fine-structure splittings in sodium of the order of 1 GHz. The essence of these original idea was to use the comb of frequencies emitted from a mode-locked laser as a precise ‘optical frequency ruler’. The spacing of the tick marks of such an optical frequency ruler is given by the repetition rate, f_r, at which pulses are emitted from the mode-locked laser, and while the concept of an offset frequency common to all comb elements did not appear in [115], it does show up in the dissertation of James Eckstein, where it is loosely termed the ‘carrier frequency’ about which the comb of modes expands [116]. With the exception of related two-photon spectroscopy work [117, 118], the development of the FM laser by the group of Ferguson and co-workers [119], and the proposal by Wineland *et al* [120] of using a mode-locked laser as an optical-to-RF divider, the topic of mode-locked lasers for frequency metrology lay largely dormant for almost two decades.

It is perhaps not surprising that the mode-locked laser comb re-emerged in the laboratory of T Hänsch in Garching, Germany. As already mentioned above, the interval division approach [102] to measuring optical frequencies was seriously pursued in the mid-1990s and resulted in what was then the most accurate measurement of the hydrogen 1s–2s transition [91]. However, this approach still required an auxiliary optical standard as the reference (CH_4-stabilized He–Ne at 3.39 µm) in addition to five interval division stages to reduce a 2.1 THz frequency gap down to a countable microwave frequency. The entire system required an impressive 14 lasers controlled by 16 different servos [121]. Some manner of simplification was provided when an EOM-based frequency comb of Kourogi was used instead of the optical interval dividers [92], but still a broader bandwidth comb source was needed for significant simplification to occur.

In 1998–99, just such a broadband comb ($\sim$18 THz in the first demonstration) was found in the form of Kerr-lens mode-locked femtosecond lasers [122, 123], thus marking a significant shift in the experimental approach and thinking of those in the field of optical frequency metrology. Where in the past frequency metrologists liked to design and use carefully controlled, low-power (milliwatt level) CW oscillators, the new femtosecond laser approach started with a laser that generated few-cycle optical pulses via the nonlinear Kerr effect that required peak powers at the megawatt level. There was a certain level of scepticism in the frequency metrology community that such a highly nonlinear laser could be controlled in a way comparable with that of the best CW lasers. And as might be expected, this (mis)conception was exacerbated by the general lack of communication between optical frequency metrologists and those developing and using the femtosecond lasers. To their credit, the frequency metrologists did recognize a good idea when it was demonstrated in a convincing fashion, as was the case in the experiments of Udem *et al* [122, 123], which first introduced the now-familiar femtosecond laser comb concept shown in figure 11 and demonstrated its utility for optical frequency metrology.

Beyond the reasons given above, the absence of robust solid-state femtosecond lasers likely contributed to the 20 year delay between the first conceptual experiments and practical implementation of the frequency comb concept. Although the femtosecond dye laser was developed throughout the 1970s and 1980s, it was not until the 1990s that high-power Kerr-lens mode-locked femtosecond lasers based on titanium-doped sapphire (Ti : sapphire) were perfected [124–127]. Such lasers rely on the Kerr nonlinearity to provide a nonlinear intracavity lens that effectively acts as an ultra-fast time gate to tightly synchronize the broad array of frequencies that are resonant in the laser cavity. Besides the striking bandwidth that is achievable directly from a femtosecond laser (a Fourier-limited 10 fs pulse requires roughly 40 THz of bandwidth), perhaps the most important development in the (re)introduction of these lasers to the realm of frequency metrology was the clear connection between the pulse-to-pulse carrier-envelope phase slip, $\Delta\phi$, and the overall offset, $f_0 = f_\mathrm{r}(\Delta\phi/2\pi)$, of the resulting spectral comb from exact harmonics of the repetition rate, f_r [128, 129]. This offset arises from the difference between the group and phase velocities as the pulse traverses the various dispersive elements in the laser cavity. The relationship between f_r and f_0 and the nth element of the optical frequency comb is given by the simple expression

$$f_n = nf_\mathrm{r} + f_0. \tag{1}$$

In this equation we find the evenly spaced optical frequency ruler in addition to the route to a direct link between the microwave domain (f_0 and f_r) and the optical domain, f_n.

Assuming that the integer $n = N$ can be determined [130, 131], the microwave-to-optical connection requires measurement of the three other variables found in this equation. Generally speaking, the frequency of f_N can be determined via the heterodyne beat with an optical frequency standard [128]. Additionally, the microwave frequency, f_r, can be simply

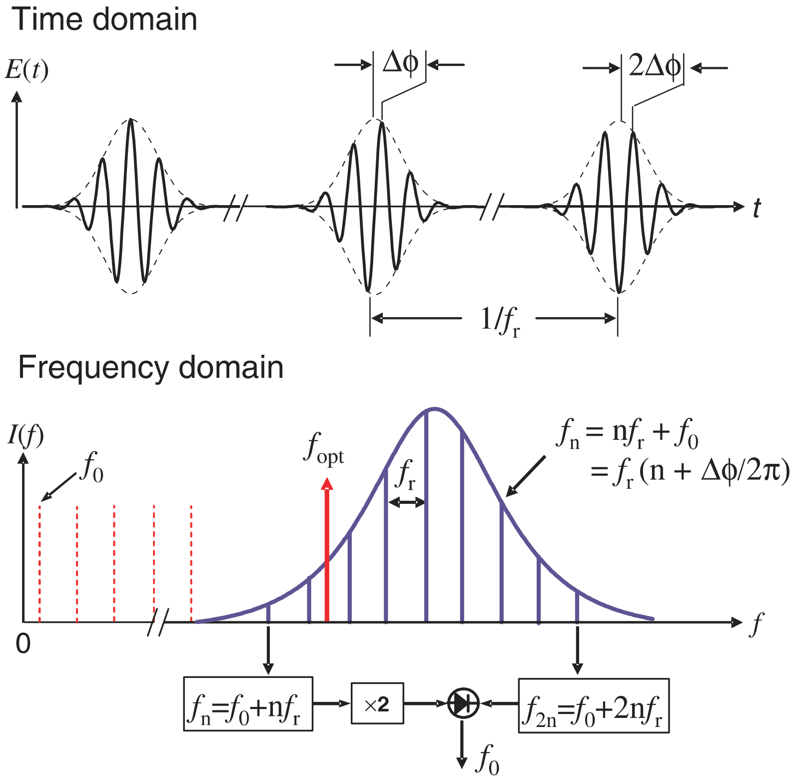

Figure 11. The basic time- and frequency-domain representations of the output of a mode-locked femtosecond laser. Pulses are emitted at the rate f_r, but because of dispersion in the laser cavity, the carrier advances with respect to the envelope by $\Delta\phi$ from one pulse to the next. In the frequency domain, the result of this phase slip is an offset common to all modes of $f_0 = f_r\Delta\phi/(2\pi)$. The lower half of this figure also shows the f–2f technique of self-referencing, whereby the offset frequency is measured. Heterodyne beats between elements of the comb and an optical frequency standard f_{opt} can be measured, provided f_{opt} falls within the comb bandwidth.

measured via photodetection of the output pulse train from the laser. However, because of the difficulty in calculating or measuring the differential group and phase delays, an accurate determination of f_0 is not as straightforward. Fortuitously, at about the same time as the first femtosecond frequency comb measurements were being performed in Garching, groups at Lucent Technologies and the University of Bath were developing highly nonlinear optical fibres that could spectrally broaden the output of an 800 nm unamplified femtosecond Ti : sapphire laser to more than 300 THz—or equivalently an octave spanning from at least 500 nm to 1000 nm [132, 133]. The availability of an octave-spanning spectrum provided a convenient means of exactly determining f_0 with a technique now referred to as self-referencing. The frequency of the comb elements at the low-frequency end of the spectrum can be doubled in a nonlinear crystal and subsequently heterodyned against the high-frequency components of the comb to yield $2(nf_r + f_0) - (mf_r + f_0) = f_0$ when $n = m/2$ (see figure 11). Such an approach was first demonstrated by Jones *et al* [134] and continues to be the most common manner in which to measure f_0. This so-called 'f–2f' self-referencing scheme has now been demonstrated with low-power Cr : LiSAF [135], Cr : forsterite [136] and Er-doped fibre lasers [137–141]. It has also been shown that 'f–2f' self-referencing can be accomplished with Ti : sapphire lasers that directly emit an octave-spanning spectrum [142–145]. As summarized by Telle *et al* [129], it is not necessary to have an octave-spanning spectrum from the femtosecond laser to measure f_0. The price to be paid, however, is that extra steps of nonlinear conversion must be employed. For example, with a spectrum spanning two-thirds of an octave, f_0 can be obtained through a comparison of the second harmonic and third harmonic of the separated portions of the optical spectrum, e.g. $3(nf_r+f_0)-2(mf_r+f_0) = f_0$ when $n = 2m/3$ [137, 142, 146].

In just a matter of years the new femtosecond comb technology has fully replaced laboratory efforts that existed for decades, and it is now widely accepted that mode-locked femtosecond lasers will play a critical role in the next generation of atomic clocks based on optical frequencies [28, 89]. A few representative Ti : sapphire laser configurations and resulting optical spectra are shown in figure 12. It is important to realize that the array of frequency modes given by equation (1) reside beneath the broad spectral envelopes shown in this figure (which were recorded using a low-resolution grating spectrometer). At present, such a femtosecond laser frequency comb connecting optical and microwave domains can occupy less than a square metre of lab space (see figure 12(*g*))—which is between a factor of 100 and 1000 less floor space than was required for a complete harmonic frequency chain. Moreover, due to its simplicity and relatively low cost, the techniques and tools described here can be implemented in university and industrial research labs, which has resulted in a variety of interesting new applications and avenues of research in the time domain [147–149] in addition to the frequency-domain applications on which we focus here.

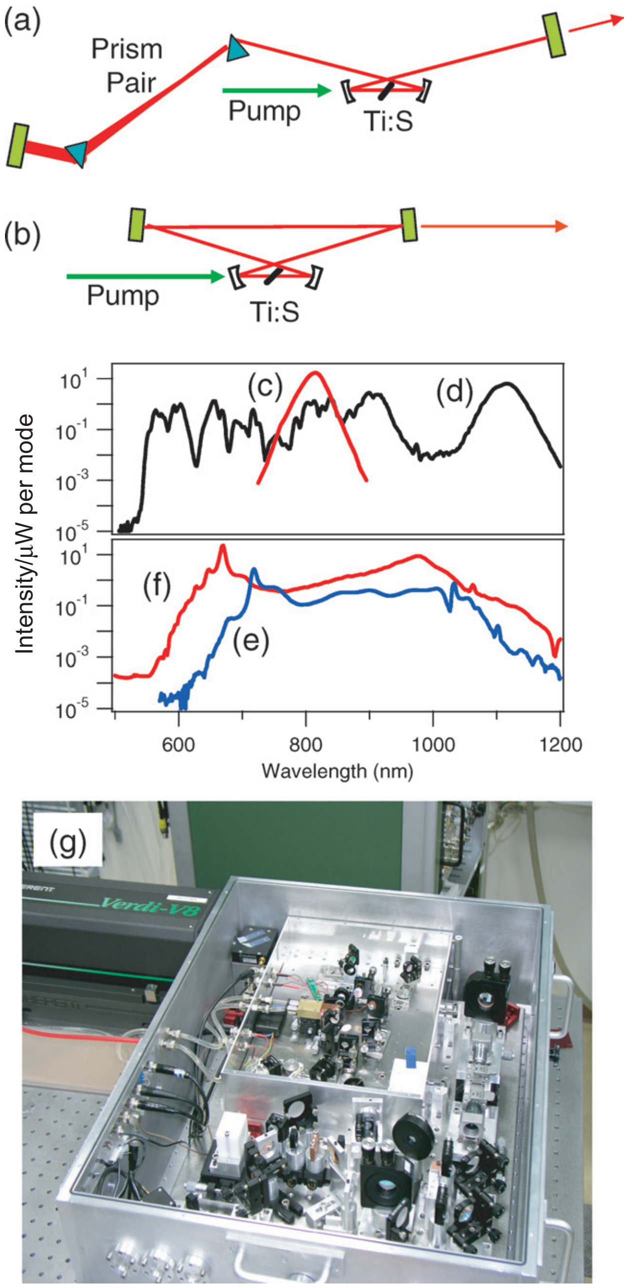

Figure 12. Common Ti : sapphire femtosecond lasers used for frequency metrology and their output spectra. (*a*) 100 MHz linear cavity design employing prisms and in some cases a combination of chirped mirrors [126, 143]. (*b*) 0.5–3 GHz ring laser utilizes only chirped mirrors [184]. (*c*) Typical narrow bandwidth spectrum obtained from (*a*) or (*b*). (*d*) Typical spectrum generated with (*c*) coupled into microstructure nonlinear fibre. With optimized dispersion control and cavity alignment lasers (*a*) and (*b*) can directly produce the very broad spectra (*e*) [143] and (*f*) [145, 185], respectively. (*g*) A photo of a complete 800 MHz femtosecond-laser frequency comb housed in a box with dimensions $69 \times 54 \times 23\,\mathrm{cm}^3$ [186]. Photo courtesy of BIPM.

The most immediate application of the femtosecond laser frequency comb has been in the comparison and measurement of emerging optical frequency standards in terms of the Cs microwave standard. The acceleration of progress with the advent of the femtosecond laser frequency comb in this area has already been discussed in the context of figure 7. Within weeks of combining a femtosecond laser with a nonlinear microstructure fibre, the first direct microwave-to-optical frequency measurements with octave-spanning frequency combs were made [134, 150]. Over the past few years, this technology has become commonplace in national metrology institutes and university labs, yielding frequency measurements of many excellent optical and IR frequency standards [37, 151–156].

Typically, such measurements make use of an octave-spanning frequency comb as either a microwave-to-optical multiplier or an optical-to-microwave divider. As will be discussed below, the most significant distinction between these two measurement modes is in regard to whether the reference for the comb is a microwave standard or an optical standard. In both cases, the offset frequency of the comb is measured (e.g. as shown in figure 11) and typically phase-locked to a stable microwave source. Modulation of the intracavity power of the femtosecond laser and the tilting of a mirror in a linear-cavity laser have both been shown to be useful actuators in controlling f_0. Since f_0 is an additive term in equation (1), its control is generally (but not always) less critical than the multiplicative f_r. Once f_0 is fixed we can measure f_r using a high-speed photodetector and then phase-lock the measured f_r (employing a PZT to change the length of the laser cavity and hence f_r) to a signal derived from a microwave frequency standard. In such an approach, the noise of the microwave reference is multiplied up to the optical domain. However, beyond Fourier frequencies of ∼1 kHz, the noise on f_r in a free-running femtosecond laser falls below that of typical laboratory microwave standards [157], such that a low bandwidth loop is all that is required to guide the average frequencies of the optical comb elements without adding significant multiplicative noise.

With both f_0 and f_r phase-locked to a caesium-referenced microwave source, every optical element f_n of the frequency comb can have the same fractional stability and uncertainty as the microwave reference. Thus, a heterodyne beat $f_\mathrm{b} = f_\mathrm{opt} - f_N$ between a CW optical frequency standard and element N of the comb allows one to absolutely determine the frequency of the optical standard as $f_\mathrm{opt} = Nf_\mathrm{r} + f_0 + f_\mathrm{b}$, where the signs associated with f_b and f_0 can assume either positive or negative values. In the case of f_b the sign can be determined by varying the frequency of f_r (while f_0 is phase-locked) and observing the associated change in f_b and, similarly, the sign of f_0 can be determined by varying f_0 (while f_r is phase-locked) and again observing the change in f_b. In practice, we do not filter the optical comb to the extent that only a single element is heterodyned with f_opt, but rather a group of comb elements are selected with a grating or optical bandpass filter and are then combined with f_opt on the heterodyne detector [128]. The photocurrent then contains the discrete beat signals between many comb elements and f_opt (corresponding to different values of N and/or different signs of f_b), and a specific beat is selected in the microwave domain. The integer N is typically determined from additional information provided by a low-resolution frequency measurement, such as that obtained with a wavemeter, although this requires that the mode-spacing of the comb be greater than the uncertainty of the wavemeter. Alternatively, in some cases N can be determined by multiple measurements with different values of f_r, as described in [130, 131].

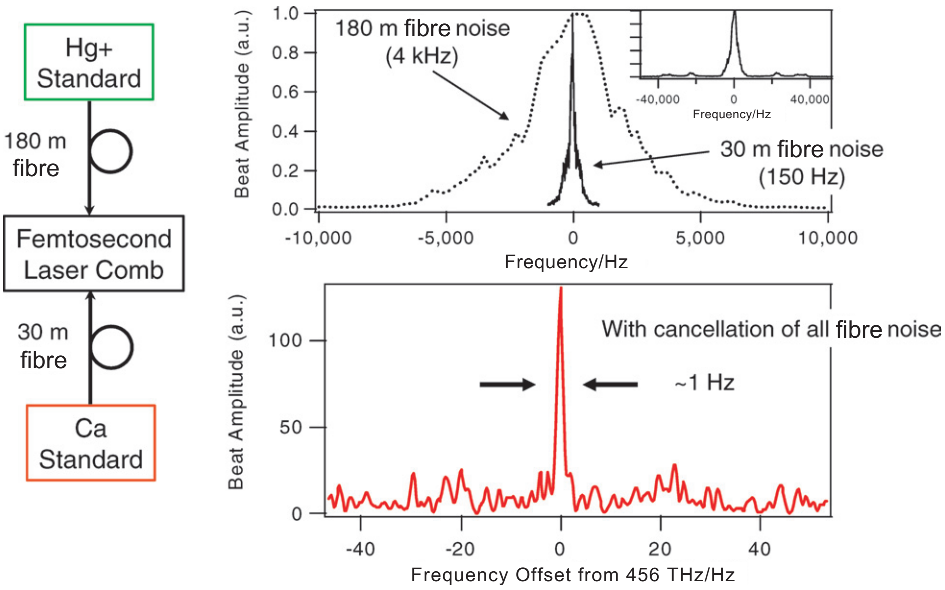

Figure 13. Comparison of the local oscillators of the Hg$^+$ (532 THz) and Ca (456 THz) frequency standards. The femtosecond laser frequency comb is phase-locked to the 532 THz reference. With no cancellation of the noise introduced by the 180 m optical fibre, the phase lock writes the noise onto the femtosecond comb, where it is manifest as a 4 kHz linewidth when measured against the 456 THz stable laser. Once the transmission noise on the 180 m fibre is actively cancelled, we see the 150 Hz linewidth added by the 30 m fibre. With the additional active cancellation of this noise, the 1 Hz wide effective beat between the two CW lasers is observed.

If the femtosecond laser frequency comb is used as a divider, one would use f_b as the small microwave offset in a phase-lock of the Nth comb element to the CW laser. Since the femtosecond comb elements are generated phase-coherently, all the other elements in addition to f_r are fixed in this manner such that $f_r = (f_{opt} + f_b + f_0)/N$. Provided reasonably good phase-locks are employed for f_b and f_0 (uncertainty below the 0.1 Hz level on what is typically a 100 MHz signal), the dominant source of noise in f_r should be due to the optical frequency reference, f_{opt}. Optical references with 1 s fractional frequency instability near or below the 10^{-15} regime have been demonstrated [158, 159], which is a value significantly below what can be obtained with typical microwave references. Controlling the comb, as just described, with such an optical reference has the distinct advantage that the fractional instability and frequency noise of the optical reference can potentially be transferred to the comb elements and to f_r. Indeed, this technique can lead to the generation of optical pulse trains at f_r with subfemtosecond residual jitter [160]. Moreover, the optically referenced frequency comb is a convenient tool for comparing optical standards separated by frequency gaps that are far too large to bridge with standard heterodyne techniques. At NIST, we have employed such a technique for comparing the local oscillators of the Ca (456 THz) and Hg$^+$ (532 THz) optical frequency standards [89]. Figure 13 shows an example of the $\sim$1 Hz wide optical beatnote that has been observed between these two stable lasers via a femtosecond comb that is stabilized to the 532 THz light. As shown in this figure, the two stable CW lasers are sent to the femtosecond comb in optical fibres. Depending on the fibre length and its environment, the phase modulation within the fibre is sufficient to broaden the observed linewidth to several kilohertz. In addition to the two extremely narrow linewidth CW lasers [158, 159] and the well-stabilized frequency comb [161], active cancellation of the transmission noise [162] must be employed on both fibre links to reach the hertz level shown in this figure.

Within the context of controlling the frequency comb and using it for measurements, it is worth noting the 'transfer oscillator' concept of Telle *et al* [163] in which a judicious choice of frequency mixings effectively eliminates the noise properties of the *unstabilized* femtosecond laser when it is used to determine the ratio of widely separated optical frequencies, or even optical and microwave frequencies. An advantage of this technique is that it allows one to replace the more difficult and relatively slow ($<$100 kHz) control of the femtosecond laser with more straightforward and faster ($\sim$1 MHz bandwidth) phase-tracking oscillators on the various heterodyne beats.

As might be expected, the introduction of mode-locked femtosecond lasers into the field of optical frequency metrology was met with some initial questions of how well such a new technology could perform relative to what existed. The most obvious question one can ask about equation (1) is whether or not the comb of optical frequencies is indeed uniform or evenly spaced. The time-domain perspective of how the mode-locked laser functions leads one to the conclusion that this must be the case. An uneven spacing of the modes would imply that different portions of the spectrum experience different roundtrip delays in the cavity. Were this the case, the pulse would rapidly spread and break apart, which is not consistent with the soliton-like operation of mode-locked lasers. The first frequency-domain verification of this was offered by the group of T Hänsch, by comparing a 44 THz wide comb from a femtosecond laser with an optical interval divider, thus confirming the uniformity at the level of a few parts in 10^{18} [123, 164].

In order to minimize the possibility of unknown systematic effects, it is valuable to compare several independent mode-locked laser frequency combs. Along these lines, a few tests have been performed using microwave standards to reference multiple femtosecond laser frequency combs and/or more traditional frequency chains, which were subsequently compared in the optical domain [80, 165, 166]. The best result in this case is an uncertainty of $\sim 5 \times 10^{-16}$ [165], with the uncertainty limited predominantly by the required averaging time. As already noted, significantly improved short-term instability can be obtained when the femtosecond comb is referenced to an optical standard. This has now permitted tests of equation (1) with Ti : sapphire-based frequency combs at or below fractional uncertainty levels of 10^{-18} [167–169]. It is important to note that these most stringent tests are based on comparisons of the optical components of the frequency comb. A significant difficulty has been the attainment of the same level of uncertainty in an electronic signal obtained by photodetection of f_r, where the conversion of amplitude noise to phase noise has been identified as a significant obstacle to generating electronic signals of the lowest noise [170]. Only recently have electronic signals with instabilities in the low 10^{-15} range been generated with a frequency comb that is referenced to a stabilized CW laser [171]. The close-to-carrier phase noise on such 'optically generated' microwave signals is orders of magnitude better than what can be obtained from any conventional microwave oscillator [172, 173].

The reliability of the mature Ti : sapphire laser technology has made it the natural place to begin this exciting field, and it is likely that femtosecond laser-based frequency combs employing Ti : sapphire will continue to be used in many applications. However, the present size and cost of the pump laser for Ti : sapphire based systems have motivated the search for alternative systems. In the future, it would not be surprising to find that femtosecond laser-based synthesizers and optical clocks will be widely used in science and technology and will be commercially available in compact packages similar to today's microwave synthesizers and clocks. The availability of robust, low-priced femtosecond-laser synthesizers will be particularly important for applications in air- or space-borne platforms, or widespread applications (such as communications systems) for which cost and rugged packaging are of greatest importance.

To date, some of the promising femtosecond lasers that have produced octave-spanning spectra include diode-pumped Cr : LiSAF [135], a fibre-laser pumped Cr : forsterite [174] and an Er-doped fibre laser [137–140] (see figure 14). Each of these has some advantages and disadvantages relative to Ti : sapphire. For example, both Cr : LiSAF and Cr : forsterite have more convenient and compact pumping schemes either directly with diode lasers or with a Yb-doped fibre laser. One of the trade-offs here is that these laser hosts are not as broadband as Ti : sapphire and tend to have worse thermal properties. An Er-fibre laser-based comb generator has a number of advantages over Ti : sapphire. It can be much lighter and more compact, robust and power-efficient than a bulk optic solid state laser system. Additionally, it can be easily integrated into a telecommunication system in the important 1300 nm to 1600 nm regime. However, at this point, the Er-fibre based systems operate only at repetition rates of 50 MHz to 100 MHz and can have excess noise that is not fully understood [137, 139]. If wavelength coverage is a concern, one can take advantage of nonlinear frequency conversion external to the femtosecond laser itself. This has been demonstrated for SHG and THG with Cr : forsterite [175] and fibre laser sources [137]. Another interesting option is to use DFG between two extremes of a Ti : sapphire laser comb to generate a frequency comb further in the IR [176]. While this provides extra wavelength tunability for the femtosecond source, it is also a means of generating a frequency comb that is independent of the offset frequency, f_0. As also shown in figure 14, wide tunability between 7 µm and 10 µm can also be accomplished with independent Ti : sapphire lasers that are synchronized and potentially phase-locked [177].

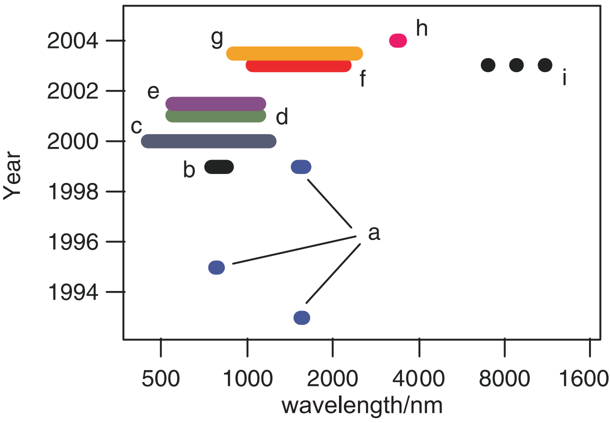

Figure 14. Spectral extent (plotted logarithmically) of some sources used as combs in frequency metrology. (a) Electro-optic modulator-based comb developed by Kourogi and co-workers [98, 183, 187]. (b) Ti : sapphire femtosecond laser comb [122, 123]. (c) Octave-spanning femtosecond laser comb generated using microstructure fibre [134, 150]. (d) Broadband spectrum generated directly from a Ti : sapphire laser [188]. (e) Cr : LiSAF femtosecond laser plus microstructure fibre [135]. (f) Octave-spanning comb generated with femtosecond Cr : forsterite and nonlinear optical fibre [174]. (g) Octave-spanning comb generated with Er-fibre laser and nonlinear optical fibre [137–140]. (h) Offset-frequency-free comb near 3400 nm generated via difference frequency generation [176]. (i) Tunable frequency comb generated via difference-frequency generation between repetition-rate-locked Ti : sapphire lasers [177].

6. Conclusion

With the introduction of new tools and technology, the field of optical frequency metrology is more vibrant today than at any point in its history. Femtosecond laser-based frequency combs have transformed the measurement of optical frequencies from a formidable task requiring the resources of large-scale national facilities to a routine undertaking accessible to virtually any researcher. Indeed, relatively compact femtosecond laser systems are now commercial products, and when linked to caesium-based time via the global positioning system (GPS) it is possible to make an 'absolute' optical frequency measurement at any place on earth [178, 179]. Moreover, this straightforward connection between the microwave and optical domains has invigorated the development of optical frequency standards, with the result that a new generation of clocks based on optical transitions are being developed in laboratories around the world. These research efforts already have many interesting scientific and technological spin-offs, which promise to keep the field of optical frequency metrology an exciting area of research for many years to come.

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