Optical Frequency Combs: From Frequency Metrology to Optical Phase Control

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Invited Paper

Abstract—The merging of continuous wave laser-based precision optical-frequency metrology with mode-locked ultrafast lasers has led to precision control of the visible and near-infrared frequency spectrum produced by mode-locked lasers. Such a phase-controlled mode-locked laser forms the foundation of a "femtosecond optical-frequency comb generator" with a regular comb of sharp lines with well-defined frequencies. For a comb with sufficiently broad bandwidth, it is now straightforward to determine the absolute frequencies of all of the comb lines. This ability has revolutionized optical-frequency metrology, synthesis, and optical atomic clocks. Precision femtosecond optical-frequency combs also have a major impact on time-domain applications, including carrier-envelope phase stabilization, synthesis of a single pulse from two independent lasers, nonlinear spectroscopy, and passive amplifiers based on empty external optical cavities. The authors first review the frequency-domain description of a mode-locked laser and the connection between the carrier-envelope phase and the frequency spectrum to provide a basis for understanding how the absolute frequencies can be determined and controlled. Using this understanding, applications in optical-frequency metrology and synthesis and optical atomic clocks are discussed. This is followed by discussions of time-domain experiments.

Index Terms—Atomic clocks, carrier-envelope phase, femtosecond lasers, frequency control, frequency synthesizers, metrology, nonlinear spectroscopy, optical frequency comb, optical frequency measurement, phase-locking, precision measurement, stabilized lasers, synchronization, ultrafast science.

I. INTRODUCTION TO MEASUREMENT OF OPTICAL FREQUENCIES

STATE-OF-THE-ART optical-frequency standards based on cold atoms, ions, and molecules exhibit excellent frequency stability and have the potential for achieving high reproducibility and accuracy. Some of them promise an instability down to $4 \cdot 10^{-17}$ at 1 s [1], [2] and uncertainties at a 10^{-18} level [3]. Such frequency references find applications in precise tests of fundamental physics [4], measurement of fundamental constants, and their possible variation with time [5]–[8], more accurate determination of atomic transitions [9] in spectroscopy, such as the value for the Rydberg constant and the 1S-Lamb shift [10], [11], tests of special [12] and general [13] theories of relativity, and quantum electrodynamics (QED) [14]. Fundamental physical constants such as the fine-structure constant, the ratio of Planck's constant to electron mass h/m_e, and the electron-to-proton mass ratio m_e/m_p [15] are also being determined with increasing precision using improved precision laser tools [16]. Optical frequency standards play an important role for applications in navigation or very long baseline interferometry, gravitational wave detection [17], optical communications [18], and modern length and frequency metrology [19]. Eventually, next-generation optical clocks are expected to be superior to existing microwave standards and may become future national standards of time and frequency.

However, the use of optical frequency standards for the realization of basic units, e.g., of time and length, was hampered by the difficulties of measuring their high frequencies because an absolute measurement of frequency must be based on the time unit "second," which is defined in terms of the microwave frequency of a hyperfine transition of the cesium atom. The precise knowledge of these frequencies or wavelengths requires complex "clockworks" to connect optical frequencies to those in the microwave region. The basic difficulty of optical frequency measurements is caused by the fact that the frequency of visible radiation is approximately a factor of 50 000 higher than that of the Cs clock and no means of direct electronic frequency counting exist for visible radiation.

For about 25 years, the method for measuring optical frequencies with a frequency multiplication chain has been the same as is routinely used for measuring microwave frequencies: an oscillator of precisely known frequency is used in conjunction with a nonlinear element to generate harmonics of its frequency. A suitable harmonic is then compared with an unknown higher frequency source by measuring the beat of the latter with the nearest harmonic. In a step-wise frequency multiplication, this procedure continues until the desired optical frequency is reached. A detailed description of the key elements of such a harmonic frequency multiplication chain can be found in [20].

Frequency chains are large scale and expensive research instruments that require resources that can be provided by only

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TABLE I
HARMONIC FREQUENCY CHAINS OPERATED BY NATIONAL LABORATORIES

Laboratory	reference frequency	target frequency	ref.
NIST	Cs clock	CH_4- HeNe	[39]
National Institute of Standards and Technology	CH_4- HeNe	I_2-HeNe	[43]
INMS/ NRC	Cs clock	CH_4-HeNe	[45]
Institute for National Measurement Standards of the National Research Council Canada		I_2-HeNe,	[142]
		Sr^+	[143]
PTB	Cs clock	CH_4- HeNe, OsO_4, Ca	[20]
Physikalisch Technische Bundesanstalt			
LPTF	Cs clock	$OsO_4 \ CO_2$	[46]
Laboratoire Primaire du Temps et des Frequences	OsO_4	CH_4- HeNe	[46]
	$OsO_4 \ CO_2$	I_2-HeNe	[47]
NPL National Physical Laboratory	Rb clock	CH_4- HeNe	[144]
ILP Institute of Laser Physics of SB RAS, Novosibirsk	Rb clock	CH_4- HeNe	[99, 145]
VNIIFTRI National Research Institute for Physical-Technical and Radiotechnical Measurements	Cs clock	CH_4- HeNe	[146]
NRLM National Research Laboratory of Metrology	Cs clock	CH_4- HeNe	[147]

a few national laboratories. Furthermore, the frequency chain can cover only some discrete frequency marks in the optical spectrum. Basically, each existing chain has been built for one single target frequency. Difference frequencies of many terahertz could still remain between a target frequency and a known reference so that sophisticated setups are needed to bridge these gaps.

These issues have represented major obstacles to making optical frequency metrology a general laboratory tool. As a result, the most precise frequency values recommended by the Comité International des Poids et Measures (CIPM) were based on very few absolute frequency measurements.

Since 1990, alternative approaches to overcome these limitations have been proposed and tested as simple and reliable solutions for bridging optical frequency gaps up to several terahertz. Some popular schemes include frequency interval bisection [21], optical-parametric oscillators (OPO) [22], optical comb generators [23]–[26], sum- and difference-frequency generation in the near-infrared [27], frequency-division-by-three [28], [29], and four-wave mixing in laser diodes [30]. All of these techniques rely on the principle of difference-frequency synthesis, in contrast to the frequency harmonic generation method normally used in traditional frequency chains. They offered the possibility to bridge frequency gaps of a few terahertz without fully solving the problem of measuring any desired frequency from the microwave to the visible spectrum.

In the year 1999, the group of T. W. Hänsch at the Max Planck Institute for Quantum Optics (MPQ), Garching, introduced ultrafast Kerr-lens mode-locked Titanium-Sapphire lasers into the field of optical frequency metrology [31], [32]. These lasers emit a periodic train of short pulses. The spectrum of this emission corresponds to a comb of distinct lines with well-defined spacing if the mode-coupling mechanism is sufficiently strong and fast. Together with the advent of microstructure optical

fibers [33]–[35] optical frequency measurements have been substantially facilitated, which has had a revolutionary impact.

A. Outline of Article

In Section II, we focus on optical frequency metrology, past, present, and prospects for the future. We describe the historical development of the frequency measurement of optical frequencies using harmonic frequency chains, and as a concrete example we describe the setup used at Physikalisch-Technische Bundesanstalt (PTB) to measure the frequency of a Ca-stabilized laser. Frequency divider chains as well as the recent advances in optical frequency synthesis and metrology triggered by the use of femtosecond lasers are then presented. Section III covers the aspect of optical atomic frequency standards controlling a femtosecond comb. Thus, a stable clock signal in the radio frequency domain is generated, leading to a so-called "optical atomic clock." Section IV deals with the control of the key laser parameters of a femtosecond comb in the frequency domain for time-domain applications such as stabilizing the carrier envelope offset phase of consecutive pulses, synchronization of independent lasers by means of the femtosecond comb, and consequences in nonlinear optics.

II. OPTICAL FREQUENCY METROLOGY

A. Historical Review of Frequency Measurements Using Harmonic Frequency Chains

Harmonic frequency chains have been developed and operated for many years in several national laboratories. However, in the past each laboratory focused on different target frequencies, hence, different frequency standards. Table I gives an overview of harmonic frequency chains built by various national laboratories. The last column lists references in which the individual frequency chains are described in more detail. A tabulation of

infrared (IR) frequency measurements performed up to 1981 has been given by Knight [9].

The first frequency measurement of a laser was performed as early as 1967 by Javan *et al.* at the Massachusetts Institute of Technology (MIT) [36]. In those experiments, far-infrared laser transitions of a CN gas laser around 900 GHz were measured with an uncertainty of a few parts in 10^7 by mixing the laser frequencies with higher harmonics of a microwave signal in a silicon diode. Three years later, Evenson and his coworkers at the National Institute of Standards and Technology (NIST), Boulder, reached the IR range at 28 THz [37]. In 1973, the NIST team measured the frequency of the CH_4 stabilized He–Ne laser at 88 THz [38] by the use of harmonic mixing and finally improved the uncertainty of the frequency measurement to a level of $6 \cdot 10^{-10}$ [39]. Barger and Hall [40] measured the wavelength of the CH_4 standard with a relative uncertainty of $3.5 \cdot 10^{-9}$ by an interferometric comparison with a Krypton standard, at that time the adopted representation of the length unit. This, in turn, resulted in a dramatically improved value for the speed of light [41]. The measurements were confirmed by a group at the National Physical Laboratory (NPL), Teddington, U.K. [42]. The 3.39-μm He–Ne laser is a convenient source with enormous gain which can be used with CH_4 (a symmetric molecule) to provide good accuracy. Since that time, this laser has played an important role as precise reference frequency in the mid infrared. In the following years, the accuracy and the range of frequency measurements have been further extended, leading to more precise frequency measurements of the iodine-stabilized He–Ne laser [43]. Finally, these activities led to the redefinition of the meter by means of the speed of light in 1983 by the Conférence Générale des Poids et Measures (CGPM).

The 10-μm region covered by CO_2 lasers has been addressed by several frequency chains. Whitford *et al.*, at the Institute for National Measurements of the National Research Council (NRC), Ottawa, used a technique of multiplication of difference frequencies produced by beating appropriate pairs of CO_2 frequencies, which were harmonically related to determine the frequencies of CO_2 lasers [44] and that of the CH_4-stabilized He–Ne laser [45] with a relative standard uncertainty of $8.3 \cdot 10^{-13}$. Precise frequency measurements of an OsO_4-stabilized CO_2 laser were performed by Clairon and coworkers at the Laboratoire Primaire du Temps et des Frequences (LPTF), Paris, with a relative standard uncertainty of $1.7 \cdot 10^{-12}$ [46]. The frequency of this laser was later used as a reference for an improved measurement of the I_2-stabilized He–Ne at 473 THz [47]. By that time, the CH_4-stabilized He–Ne laser at 3.39 μm had evolved into the most precise optical reference in the mid infrared. Using this reference frequency and the I_2-stabilized He–Ne at 633 nm, a French collaboration [48] measured a new value for the Rydberg constant with a relative standard uncertainty of $2.9 \cdot 10^{-11}$.

Extensions of frequency measurements from the IR to the visible were mostly performed by interferometric wavelength comparisons [49]–[51], which in some cases were combined with optical frequency mixing [52]. However, the uncertainties of the results were limited by the contributions due to diffraction corrections, imperfections of the optics, wavefronts distortions, and inhomogeneity of the index of refraction of the beam splitters.

In the end, this limits the obtainable uncertainty to a few parts in 10^{11}.

Some of the earlier measurements, as can be seen in Table I, were partially based on frequencies of secondary frequency standards that, in turn, were linked in separate measurements to the Cs clock. In these cases, the secondary standards themselves contributed to the uncertainty of the measured frequency value. The full benefit of an optical frequency standard can be gained only if its frequency is related to the primary standard of time and frequency without loss of accuracy due to intermediate oscillators.

In response to the continuous improvement of the reproducibility of optical frequency standards, and the increasing demand on optical frequency measurements with improved uncertainty, the PTB started activities in the field of optical frequency metrology in 1973. The initial motivation for the frequency chain was to provide an independent measurement of the frequency of the CH_4-optical frequency standard. Using additional CO_2 lasers the chain had been extended to the OsO_4-stabilized CO_2 laser. A detailed description of the IR chain can be found in [20]. Later, this chain was further extended to the Ca-stabilized frequency standard at 657 nm, bridging a frequency gap from the primary Cs clock at 9.2 GHz up to 456 THz [53], thus realizing the first fully phase-coherent frequency chain from the Cs clock up to the visible part of the electromagnetic spectrum.

Presently, 13 reference frequencies/wavelengths covering the visible and IR regions of the electromagnetic spectrum are recommended by the CIPM for the realization of the meter [19]. (See Fig. 1.)

B. Description of PTB's Harmonic Frequency Chain

For any phase-coherent frequency measurement the maintenance of the coherence or "phase-trackability" of the harmonic mixing signals is the essential requirement for a frequency measurement. Simply stated: if a frequency is multiplied by a factor N, its period duration is divided by N. If the low starting frequency is not ideally coherent but shows phase noise (i.e., the zero crossings of the field jitter by an amount Δt), there is a certain factor N_{crit} above which Δt exceeds the period duration of the multiplied frequency. This means that, on average, the period to which a zero crossing belongs is no longer defined (this phenomenon is often called "coherence collapse" which occurs at a certain multiplication factor). To perform phase-coherent frequency measurement the collapse frequency must be well above the (optical) frequency that is to be measured by the frequency multiplication process. These issues have been studied in detail by Telle [54].

The direct approach for solving this problem is to start from an RF oscillator with exceedingly low phase noise and to distribute the load of the coherence among the various oscillators along the multiplication path of a frequency measurement chain. Since gas lasers, which have a resonator of very high quality factor, can act as excellent "fly-wheels," the coherence of the lower stages in our frequency multiplication chain has only to be good enough to yield a coherence collapse well above the
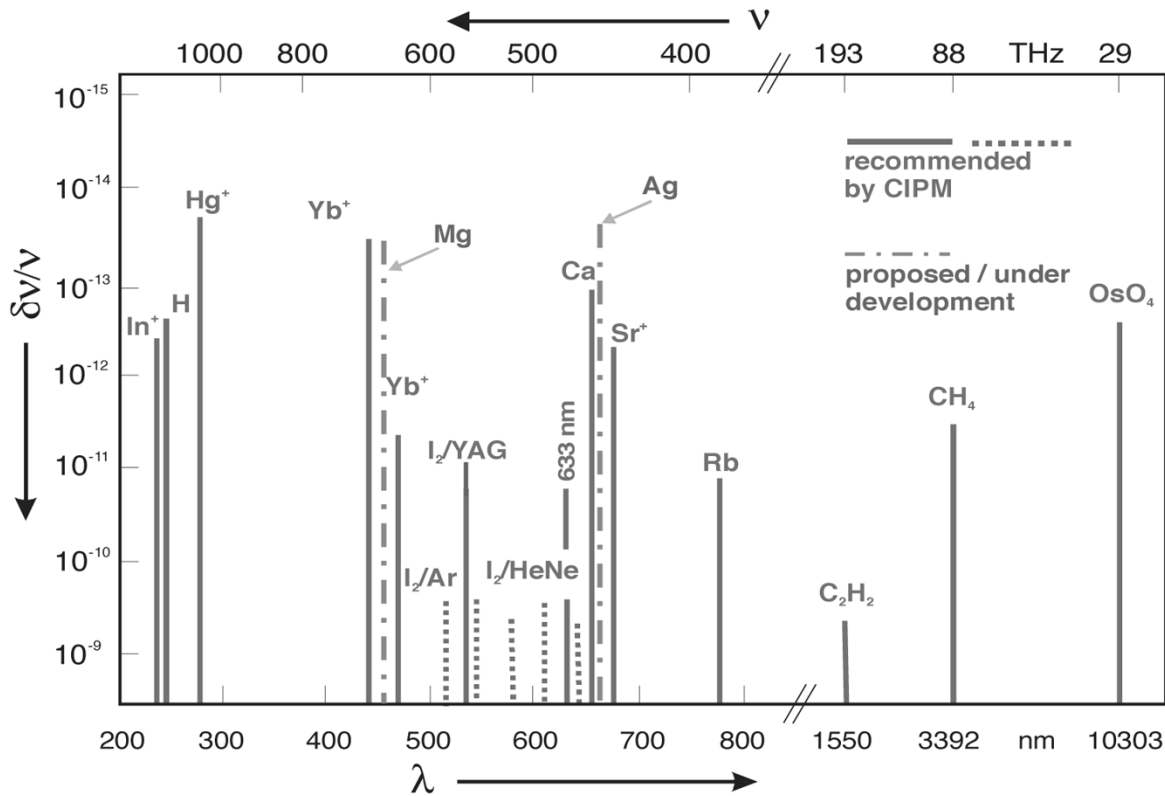
Fig. 1. Precision laser frequency references locked to atoms and molecules, showing fractional frequency uncertainties as accepted by the CIPM. Some examples of other promising frequency references and their attainable uncertainties are indicated by dotted–dashed lines.

lowest frequency gas laser (and not necessarily above the high frequency which is to be finally measured).

In the harmonic frequency chains, the large frequency ratios between the Cs atomic clock and optical frequencies are bridged in several steps. In each step, the frequencies of two oscillators are compared. The lower frequency of one oscillator is multiplied by a nonlinear device and the corresponding harmonic is compared to the higher frequency of the second oscillator which is operating close to the harmonic. The corresponding beat frequency is detected and one of the two oscillators is servo-controlled by locking the phase of the beat frequency to that of a fixed frequency. Utilizing this method, the frequencies of the two oscillators are phase-coherently related to each other. The nonlinear devices used in these chains are Schottky diodes ($\nu \leq 5$ THz), metal-insulator-metal (MIM) diodes ($\nu \leq 120$ THz), and nonlinear crystals ($\nu \geq 120$ THz).

As a specific example, we consider some details of the PTB harmonic frequency chain spanning from the Cs clock to the Ca optical standard, as shown in Fig. 2. The lower part is locked from a 100-MHz standard frequency of a hydrogen maser up to a methanol laser at 4.2 THz. As the piezo-controlled-mirror of the CH$_3$–OH laser is heavy, its motion is slow; therefore, only the frequency of the far infrared response (FIR) laser is coarsely controlled from the harmonic mixing signal, ensuring that the emission is at the center of the gain line and thus the output power is maintained at maximum. The phase coherence is transferred to the 10-μm $P^*(28)$ CO$_2$ laser by a common synthesizer technique [54].

The intermediate part of the chain, consisting of CO$_2$ lasers (tuned to $P^*(28)$ of $^{13}C^{16}O_2$ and $P^*(20)$ of $^{12}C^{18}O_2$), is

locked to a methane stabilized He–Ne laser to improve the frequency stability. Summing four photons from two CO$_2$ lasers tuned to $P^*(20)$ of $^{12}C^{18}O_2$ and P(14) on a metal-insulator-metal point-contact diode results in a frequency very close to that of the color-center laser (CCL), which is phase-locked to the fourth subharmonic of the Ca stabilized laser. This beat note between the CCL and both CO$_2$ lasers represents the frequency ratio between the Ca stabilized laser and the Cs clock at 114 THz.

The high-frequency part is locked from the Ca optical frequency standard down to a color-center laser. A fraction of a prestabilized diode laser operating at about 1314 nm is converted into radiation with exactly twice the frequency in a nonlinear crystal by means of the second harmonic generation. The doubled frequency is very close to the frequency of the Ca stabilized laser and both radiations lead to a beat note with the difference frequency when directed onto a photo detector. The frequency of the diode laser is controlled by a phase-locked loop in such a way that the beat note is kept constant. Utilizing this method, the frequencies of the two oscillators, i.e., the Ca stabilized laser and the diode laser at 1314 nm, are phase coherently related to each other. A similar stage is used to connect the frequency of a color-center laser to that of the second harmonic of the diode laser.

In order to maintain the short time stability of the chain, in addition to the phase locks all beats along the chain are counted, simultaneously, by totalizing counters. This method allows one to track the phase of all intermediate oscillators and therefore leads to a truly phase-coherent measurement. Combining these beat signals in real time yields a frequency
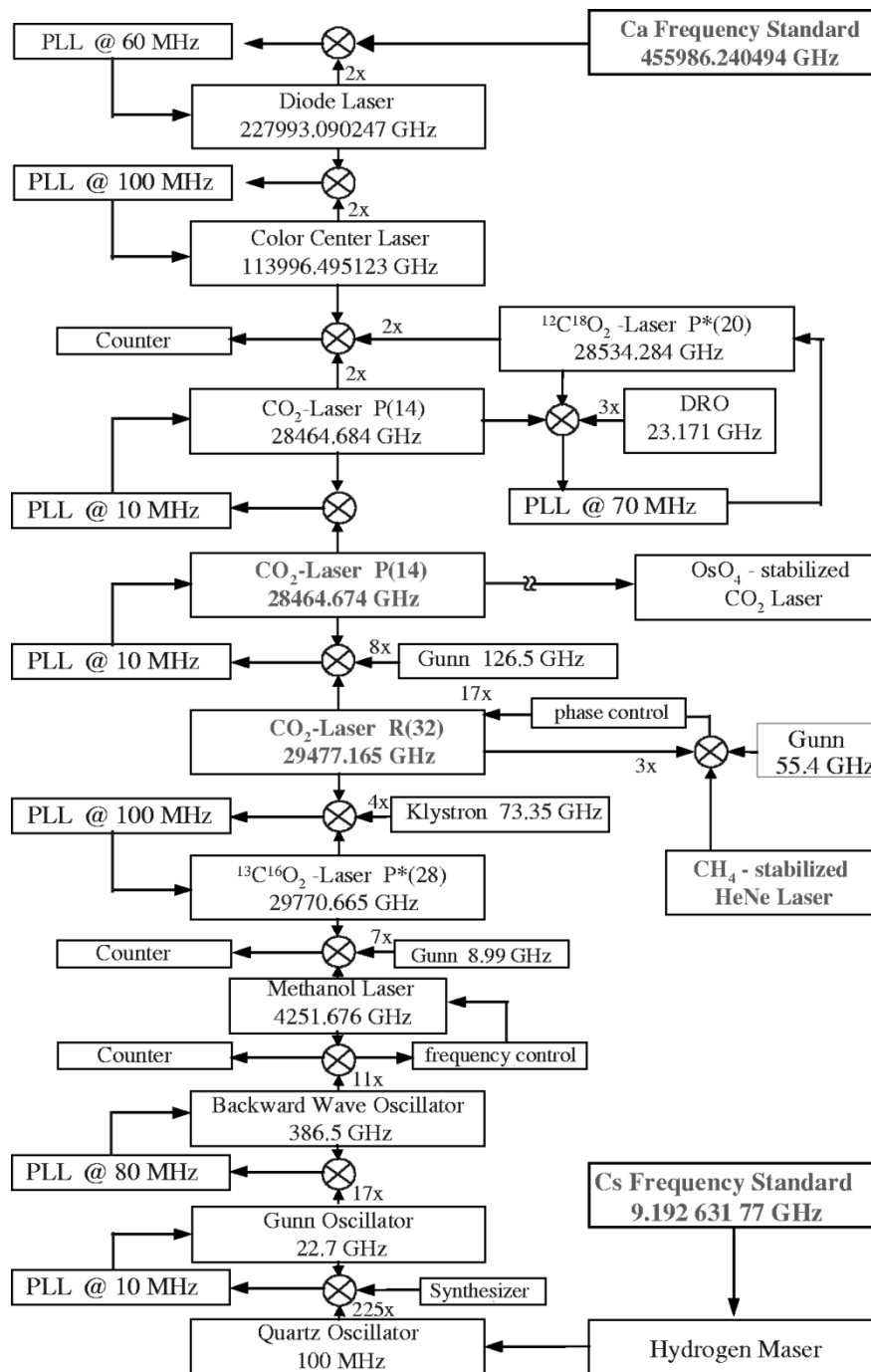
Fig. 2.   Harmonic frequency chain from the 9.2-GHz Cs frequency standard to the 456-THz Ca optical frequency standard as realized at PTB.

ratio independent of fluctuations of the intermediate transfer oscillators.

In this way, the described frequency chain allows us to measure simultaneously the frequencies of the CH_4-stabilized He–Ne laser at 3.39 μm, the OsO_4-stabilized CO_2 laser at 10.6 μm and the Ca-stabilized dye laser at 0.657 μm.

C. Optical Synthesis by Divider Approach

1) Optical Synthesis Using Parametric Oscillators: Another approach to optical synthesis is to use nonlinear optics and take advantage of the natural complementary frequencies ($f_{out1} + f_{out2} = f_{in}$) that are generated in optical parametric

down conversion. Wong [22], [55] has proposed such systems that use multiple interconnected parametric oscillators that could divide optical frequencies down to low enough frequencies that they could be counted electronically. Some experiments with optical parametric oscillators spanning large frequency intervals have demonstrated the concepts, but none of those systems has yet successfully spanned the gap from optical to microwave [56], [57].

2) Optical Synthesis by Bisection: A promising alternative to harmonic frequency chains for synthesizing and measuring optical frequencies is the optical "interval bisection" (IB) technique, introduced by Telle *et al.* [21]. In contrast to the har-
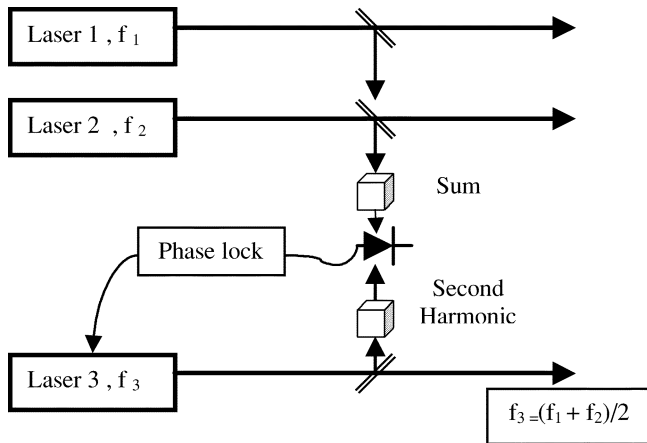
Fig. 3. Basic optical frequency interval bisection method. Sum frequency $(f_1 + f_2)$ is phase locked to the second harmonic of f_3.
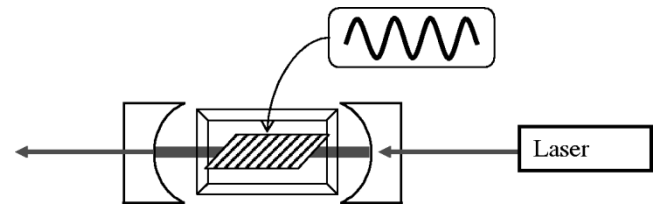

Fig. 4. Doubly resonant EOM and cavity for optical comb generation. Driving frequency to the EOM matches to a harmonic integer of the cavity-free spectral-range frequency.

monic frequency chains that start at low frequencies and then multiply up to optical frequencies, the IB method starts with two lasers at optical frequencies and then locks a third laser at the midpoint in frequency between the original two lasers. This bisection is accomplished (as shown in Fig. 3) by detecting the beat note between the second harmonic of the midpoint laser and the sum frequency of the two original lasers. A phase-lock loop is then used to force the frequency and phase of the beat note to zero and, hence, the third laser to exactly the frequency $f_3 = (f_1 + f_2)/2$.

The IB prescription accurately divides the frequency interval between the original pair of lasers in half, thus bisecting that interval. Successive bisections of each resulting 1/2-interval continue to divide the original interval by $1/2^N$, where N is the number of bisections. A particularly interesting case is when the two original frequencies are harmonically related to each other, as $f_2 = 2f_1$, then successive applications of the IB would produce a frequency interval of $\Delta f_N = f_1/2^N$ and relates the interval itself to the original frequency [58], [59]. In principle, the technique could be used to divide optical frequencies all the way down to countable microwave frequencies, and thus provide knowledge of all the lasers within the bisection chain, as well as the original laser frequency in terms of the final microwave interval. IB is based on a beautifully simple concept and is fairly straightforward to apply, but dividing by factors of two still requires lots of steps, and also components, to reach the microwaves from the visible. For example, starting at 600 THz in the visible it would take 12 bisection stages to reach 146 GHz. A microwave harmonic synthesizer could then be constructed to reach the 146-GHz frequency so that the difference frequency between the microwave source and the bisector output could be counted with sufficient precision to determine the optical frequency with high accuracy. Given that each of the IB stages requires an additional phase-locked laser along with two nonlinear mixings, a system that reached from the visible to microwaves would be complex. There are important differences between the outputs of IB chains and harmonic frequency chains that result from the distinction of division versus multiplication. An IB system consists of many stable references in the visible, whereas the harmonic chains end up with numerous stable os-

cillators at low frequencies (millimeter wave, far-IR, IR) and typically only one or two in the visible. In addition, interval bisector chains are not as frequency specific, nor are they as complex, but still no interval bisector has been made that spans from optical to microwave frequencies. Nonetheless, the idea is a powerful one that has been used to bisect optical frequency intervals of 500 THz and even divide frequency intervals of about a terahertz down to an electronically countable frequency and has proved useful in measurements of hydrogen energy levels [60], [61]. At this time, not much work is being done with the IB method, because high-accuracy optical frequency combs based on mode-locked lasers provide a much simpler solution.

3) Optical Frequency Combs: Obviously, a precisely spaced array of optical frequencies could serve as a type of "measuring stick" for optical frequencies and for the synthesis of arbitrary frequencies. A few different methods have been developed to generate an optical frequency comb with equal spacing between adjacent comb teeth. For instance, it is possible to lock a series of lasers together with a fixed frequency offset between them. This approached may be useful in making short optical pulses from continuous wave (cw) sources [62]. It is also possible to use active modulators on a cw laser beam, or to use broad-band mode-locked lasers.

a) EOM-Based Optical Combs: A natural approach to generate a comb of optical frequencies is to use phase modulation of an optical beam to generate sidebands on the optical carrier. The resulting spectrum can be quite broad if the modulation frequency and modulation index are both very large. Unfortunately, it is difficult to obtain a very large spectral coverage with a simple electro-optic modulator (EOM). The process can be enhanced by making the EOM resonant and additionally by putting the modulator inside an optical cavity that is simultaneously resonant with the optical carrier and the generated sidebands (see Fig. 4).

This multiply resonant EOM approach has been studied by a number of groups for application to optical frequency metrology [23]–[26], [63], and typical systems might have comb spacing in the 0.5 to 20 GHz range and spectral coverage of from 1 to 10 THz. EOM-based optical comb generators have some attractive features; they are compact, relatively low cost, can be rugged, and applicable at most wavelengths. Another significant advantage of the EOM combs is that the frequency spacing between comb teeth can be relatively high (e.g., 10 GHz), which makes efficient use of the photons and provides a convenient frequency grid for measurements and for calibration of other systems and components, such as those used in the frequency grid for fiber communication systems.
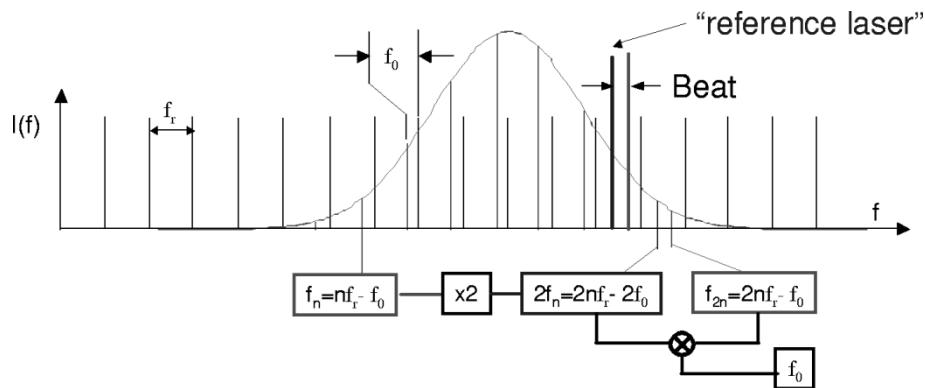
Fig. 5. Self-referenced optical frequency comb. Solid lines represent comb spectral components, while dashed lines represent fictitious frequency markers equal to exact integer multiples of the comb repetition frequency.

b) Optical Frequency Combs Based on Mode-Locked Lasers: As suggested and first demonstrated by Hänsch and collaborators in the 1970s, the regular spacing of modes in a mode-locked laser can be used to measure optical frequency intervals [64]. Early studies of this idea used the mode-locked dye lasers and ion lasers available at the time, with relatively long pulses by today's standards. Only a small amount of work in the 1980s and 1990s focused on the questions of applying mode-locked lasers for frequency measurements, with some effort invested in trying to develop FM mode-locked systems using modulators in the laser cavities [65]. Even with these initial suggestions and demonstrations by Hänsch, Ferguson, and a similar discussion of the concepts by Chebotayev [66], the scientific community that actually needed high-precision optical frequency measurements was reluctant to pursue this approach. The whole landscape changed in 1999 when Udem *et al.* first convincingly demonstrated that the optical frequency comb from a commercial Kerr-lens mode-locked laser has extremely regular mode spacing, that is equal to the repetition rate, with fractional frequency errors $<6 \times 10^{-16}$ over a frequency interval of 20 THz [31], [32]. More recent tests of the fidelity of optical frequency combs have verified that the combs can have fractional frequency instabilities of $6.3 \times 10 \leq^{-16}$ in 1 s [67] and produce frequency ratios with precisions of 7×10^{-19} [68]. This performance is somewhat better than what is required for the present cold-atom-based optical frequency standards, but the performance must continue to improve to keep pace with the promises for next generation of optical standards.

In retrospect, these ideas could have been applied effectively much sooner than they were. We can speculate on a few reasons for this slow acceptance. One reason was that there were prominent experiments (notably frequency measurements of positronium and hydrogen 1S-2S transitions) that were using pulsed lasers for precision spectroscopy, and these measurements were plagued by serious problems with distortion of absorption lineshapes and frequency metrology due to the distribution of frequencies within the pulse. Variations in the optical pulses with time and frequency chirp made it a challenge to find the center of the atomic line with high accuracy. Secondly, until the IB method was established it was difficult to prove the fidelity of the frequency combs that were produced by mode-locked lasers.

(We note that the accuracy achieved with state-of-the-art atomic frequency standards is now about 15 digits.) Thirdly, there were certainly historical biases that blinded the research community to exploring very different approaches.

Following the proof that mode-locked lasers do produce frequency combs with a mode spacing accurately given by the repetition frequency, it was clear that this method was the simplest route to optical frequency synthesis. Nearly immediately, most research groups in the field began building systems based on Kerr-lens mode-locked lasers for optical frequency measurements, synthesis, and optical atomic clocks. The field continues to advance rapidly, with significant new results coming every few months. One of the really important contributions came from another field, that being the discovery that microstructure optical fibers with air holes could broaden the spectrum of the ultrashort pulses from Ti:Sapphire lasers and generate a white-light continuum with well over an octave of bandwidth [35]. Fortunately, within some range of operating parameters a spectral comb with discrete lines persists throughout the spectrum [69]. The octave spanning spectrum was the key ingredient to "self-reference" the optical frequency comb as first demonstrated at JILA [70] and soon thereafter at MPQ [71] (see Fig. 5).

In the "self-referenced" case, the optical frequency of any mode of the comb can be written as a simple function of two measured radio frequencies, the pulse repetition frequency f_r, and the offset frequency f_o; thus $f_n = nf_r + f_o$. Here, n (a large integer, ranging from a few hundred thousand to a few million) represents the nth mode of the comb, and the offset frequency is simply offset of the comb from zero frequency as the mode number n is extrapolated to zero. Careful experimental tests of this simple mode formula, and a physical interpretation of f_0 as due to the difference between the phase and group velocities within the laser cavity, can be found in a number of papers [72], [73]. The technology of optical frequency combs continues to evolve, with new systems improving upon various aspects of the performance, including broader spectra directly from the lasers [74]–[76], self-referencing without fibers, and broad spectra at different wavelengths and from lasing materials other than Ti:sapphire (e.g., Cr:forsterite [77], fiber lasers [78]–[80], and semiconductor lasers [81], [82]). Nonetheless, to date, most high-accuracy optical frequency measurements are still done using the basic self-referenced system shown in Fig. 5.
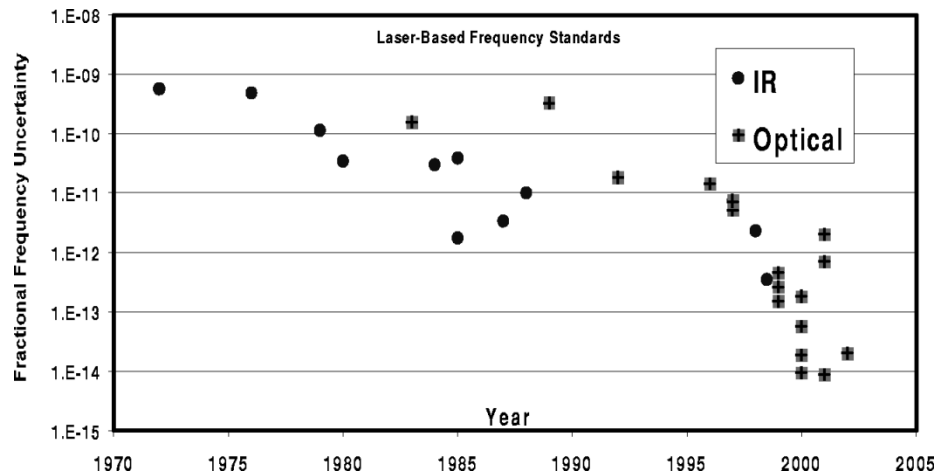
Fig. 6. Plot of the fractional frequency uncertainty of some high-accuracy optical frequency measurements versus date.

Excellent summaries of the application of mode-locked lasers to optical frequency measurement and experimental details can be found in some recent works [1], [83]–[87]. Hall has also provided a very nice historical perspective on optical frequency measurements [88].

In Fig. 6, we plot some of the state-of-the-art optical frequency measurements as a function of date, and the data show some interesting facts. Some details of the plotted data are provided in Table II. In the last three years, there has been a dramatic improvement in the accuracy, in the number of measurements, and in the prevalence of visible and UV frequency standards. These changes are primarily the result of three factors: the advent of convenient optical frequency combs based on mode-locked lasers, a dramatic improvement in accuracy of optical standards resulting from laser cooling and trapping of neutral atoms and ions that have appropriately narrow optical transitions, and the development of highly stabilized cw lasers that have the performance needed to serve as local-oscillators for the narrow optical transitions.

It is interesting to consider what absolute atomic and molecular frequency measurements actually represent. The unit of time in the SI system of units, the second, is defined by international agreement in terms of the frequency of the Cs ground-state hyperfine splitting ($F = 3, m_f = 0$ to $F = 4, m_f = 0$), and the splitting is 9 192 631 770 Hz, exactly. An absolute frequency measurement of an atomic frequency is necessarily a comparison of the unknown atomic transition with the Cs transition. These are, in essence, measurements of frequency ratios (i.e., energy ratios), and the results have some implications in terms of fundamental physics and any time variation of fundamental constants [8]. In this same light, it has been common practice (either by harmonic chains or more recently via optical frequency combs) to measure one atomically stabilized laser frequency relative to another, for example CO_2 to CH_4, I_2 stabilized He–Ne to CH_4 stabilized He–Ne, I_2 stabilized YAG to rubidium two-photon transtions, and Calcium to Hg^+, and literally thousands of molecular lines measured relative to CO_2 and CO reference lasers [89], [90].

c) Frequency-Transfer Method: Recently, Telle and collaborators introduced a clever method of using the comb from a mode-locked laser to measure the ratio of two frequencies [91]. By a judicious choice of mixing frequencies, they show how it is possible to transfer the frequency stability of one optical source to a very different optical frequency without adding additional noise from the comb. The advantage of their frequency-transfer (FT) method is that the frequency noise introduced by the mode-locked laser drops out of the measurement process and it is possible to compare two narrow optical atomic lines without requiring the optical frequency comb to be highly stabilized. In fact, with some constraints, in ratio measurements the mode-locked laser can be left to run freely, or only partially controlled as was done in a frequency measurement between Hg^+ and Ca standards where f_0 was uncontrolled [92]. In a demonstration experiment the group at PTB showed narrow beat signals in the terahertz range that were actually the transferred beat notes between optical standards (a Yb^+ stabilized laser, an iodine stabilized YAG, and a Ca stabilized dye laser) [91]. The FT methods have limitations, but can be a useful tool for measuring frequency ratios from microwaves to the visible.

III. OPTICAL CLOCKS

The dramatic simplification of a complex optical frequency chain to that of a single mode-locked laser has greatly facilitated optical frequency measurement. Another important aspect of this new technology is its high degree of reliability and precision and absence of systematic errors. Consequently, there has been an explosion of absolute frequency measurements using fs comb methods in the last two years, as reported in Section II. Not surprisingly, the most accurate results come from optical standards that are based on dipole- or spin-forbidden transitions in cold samples with extraordinary quality factors [93]–[95]. Indeed, testing fundamental physical postulates or determining constants at the next decimal place is again attracting great interest, with this new increase in measurement precision.

Another direction is to explore the less complex systems where one can consider tradeoffs such as accuracy or stability versus power consumption, cost, or size of the apparatus. Cell-based optical frequency standards such as a solid-state laser stabilized on sub-Doppler transitions of molecular iodine already offer a competitive stability near or below 1×10^{-14} when averaged over 10 to 1000 s. We have measured the

TABLE II
SELECTED HIGH-ACCURACY OPTICAL AND IR FREQUENCY MEASUREMENTS THAT INDICATE THE ABSOLUTE FREQUENCIES AND FRACTIONAL FREQUENCY
UNCERTAINTIES THAT WERE ACHIEVED FOR VARIOUS REFERENCE FREQUENCY STANDARDS OVER THE LAST 30 YEARS

Atom/ Molecule /Ion	(nm)	Frequency (THz)	Uncertainty		Year	First Author & Institution
			±Hz	Fractional		
OsO_4	10318.	29.054 057 446 660 29.054 057 446 579	50 10	2×10^{-12} 3×10^{-13}	1985 1999	Clarion, LPTF Ducos, LPTF
CH_4	3392.2	88.376 181 627 88.373 149 028 553	50 000 200	6×10^{-10} 2×10^{-11}	1972 1998	Evenson, NBS Ering, PTB
Rb – 2 photon	778.11	385.285 142 367 385.285 142 374 8	8 000 3 000	2×10^{-11} 8×10^{-12}	1993 2000	Nez, ENS Diddams, JILA
Sr+	674.03	444.779 904 409 540 444.779 044 095 6	200 300	4×10^{-13} 7×10^{-13}	1999 2001	Bernard, NRC Lea, NPL
Ca	657.45	455.986 240 493 950 455.986 240 494 158 455.986 240 494 150	430 26 8	9×10^{-13} 6×10^{-14} 2×10^{-14}	1996 2000 2002	Schnatz, PTB Udem, NIST Helmcke, PTB
I_2 a_{15} of $R(127)$ 11-5	632.99	473.612 340 492	74 000	2×10^{-10}	1983	Jennings, NBS
I_2 a_{16} of $R(127)$ 11-5	632.99	473.612 353 604 8	1 200	3×10^{-12}	2000	Ye, JILA
I_2 a_{10} of $R(56)$ 32-0	532.24	563.260 223 480 563.260 223 514	70 000 5 000	1×10^{-10} 9×10^{-12}	1995 2000	Jungner, JILA Diddams, JILA
Yb+	435.51	688.358 979 230 931	6	9×10^{-15}	2001	Stenger, PTB
Yb+	466.89	642.121 496 772 6	1300	2×10^{-12}	2001	Lea, NPL
Hg+	281.57	1064.721 609 899 140	10	9×10^{-15}	2000	Udem, NIST
In+	236.54	1267.402 452 899 920	230	2×10^{-13}	2000	VonZanthier MPQ
H $1S$-$2S$	121.57	2466.061 413 187 100	46	2×10^{-14}	2000	Niering, MPQ
H $2S$-$8S$	389.01	770.649 561 581 1	5 900	8×10^{-12}	1997	de Beauvoir, ENS

absolute frequency of such a system over the past three years. At present, the long-term self-reproducibility (single cell) is limited to about 3×10^{-13}. Better stability and reproducibility are expected from future improvements. Cell-based optical standards also play an essential role in length metrology. The important practice of international comparisons of length-standard lasers could now be accomplished by local calibrations with GPS systems, leading to achievable precision below 10^{-13} with adequate averaging times.

As the measurement precision of optical frequencies is pushed to an ever-higher level, the stability limitation imposed by available radio frequency standards served as references for fs comb becomes an important issue. Instead of running the fs comb from microwave frequencies up to optical frequencies, it appears to be advantageous to operate the comb in the other direction [96]. In other words, an optical frequency standard is used to stabilize the fs comb which, in turn, produces stable clock signals in the radio frequency domain, leading to a

so-called "optical atomic clock" [97], [98]. (In fact, harmonic chains were first used for demonstrations of optical clocks based on the 3.39-μm He–Ne laser [99], [100].) Of course, there are highly stable RF signal sources available, such as hydrogen maser or Cs fountain clock, but with limited availability. Recent experimental demonstrations support the concept that, in the future, the most stable and accurate frequency standards will be based on optical transitions. Another strong argument favoring optical stabilization of an fs comb stems from the fact that a frequency division process is involved in comparison to frequency multiplication when the fs comb is stabilized by an RF source.

The advantage of an optical frequency standard over a traditional microwave standard is apparent if we examine the frequency stability of an atomic clock. Resonance natural widths $\Delta \nu$, in the few kilohertz to the subhertz domain, are available by selection of an atomic transition with a natural decay time, τ_0, in the 100-μs to 1-s domain. In principle, one could obtain $\sim 1/(2\tau_0)$ interactions per second with approximately only two-fold broadening of the resonance linewidth by the interrogation process. So, if we collect all the available information-bearing photons, for a single measurement a signal-to-noise ratio (SNR) $\sim \sqrt{N}$ should be available, where N is the number of participating particles. Normalizing to a standard 1-s measurement time gives us SNR $\sim \sqrt{N} \times \sqrt{1/(2\tau_0)}$. An optimum frequency control system could find the center of the resonance with a precision $\sim(1/\text{SNR})$ in 1 s. Taking the resonance linewidth into account leads to a frequency uncertainty $\delta\nu$ (at 1 s) $\sim \Delta\nu/(\text{SNR}) = (2/N\tau_0)^{1/2}$. In case the interrogation time T_R (we assume the Ramsey separated-field method) is shorter than the actual lifetime of the transition under study, the fractional frequency (in)stability is then given by

$$\sigma_y(\tau) = \frac{\delta\nu}{\nu_0} = \frac{1}{\omega_0 \sqrt{N T_R \tau}}. \tag{1}$$

In this expression, $\omega_0 \ (= 2\pi\nu_0)$ is the clock transition frequency, and $\tau (\tau > T_R)$ is the total averaging time. Clearly, higher stability is most easily attained if we can increase ω_0, for example, by changing ω_0 from a microwave to an optical frequency. This simple formula gives an optimistic fundamental limit of projected stability, but even calculations that are more thorough indicate that instabilities of about $1 \times 10^{-16}/\tau^{-1/2}$ should be achievable with modern optical frequency standards [1], [2].

With the advent of wide-bandwidth optical comb technology, it is now possible to transfer the stability of the highest quality optical frequency standards across vast frequency gaps to other optical spectral regions. Furthermore, the comb technology has also established a possibility to transfer the optical stability down to the RF domain. One can now realize a network of microwave and optical frequencies at a level of stability and reproducibility that surpasses the properties of basically all commercially available frequency sources, but with a reasonable cost. Easy access to the resolution and stability offered by optical standards will greatly facilitate the application of frequency metrology both to precision experiments for fundamental physics and to practical devices.
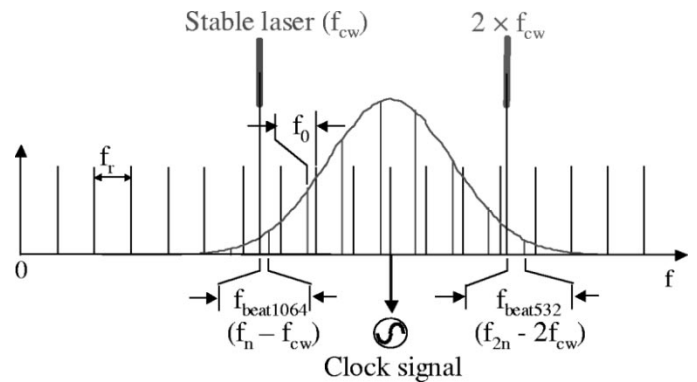

Fig. 7. Another scheme for the implementation of optical clocks using an octave-spanning optical frequency comb. Fundamental and second harmonic of the cw optical-frequency standard are used for error-signal derivation and comb stabilization.

As discussed in Section II, a comb system has two degrees of freedom, f_r and f_0, and both can be controlled effectively. We need to have two experimental observables to recover the information relating to f_r and f_0. This step can be accomplished with two different but fundamentally related approaches. The first approach (Fig. 5) uses the self-referencing technique to recover f_0, which can then be stabilized with respect to either f_r or an auxiliary stable RF source. It is worth noting that stabilization of f_0 to a few millihertz is more than adequate, as it yields fractional frequency noise of $<10^{-17}$ for an optical carrier. Once f_0 is stabilized, a heterodyne beat between one of the comb components and a cw laser (f_{cw}), which acts as the optical frequency standard, yields information about fluctuations in f_r. After appropriate processing, this error signal is used to stabilize the phase of f_r coherently to f_{cw}, thereby producing a clock signal output in the RF domain derived from f_{cw}. The second approach (Fig. 7) uses two beat signals between a cw stabilized laser (f_{cw}) and its second harmonic ($2f_{cw}$) and two respective comb components in the closest neighborhoods of these two cw frequencies. One immediately sees that the two approaches are intimately related and we are taking the same advantage of the octave bandwidth of an fs comb in both cases. (In the second case, frequency doubling happens to the cw laser instead of the comb.) Through appropriate electronic mixing of the two beat signals, one can derive the servo control error signals associated with f_0 and f_r.

The optical atomic clock that NIST recently demonstrated uses a single laser-cooled Hg^+ ion or a sample of cold Ca atoms as the frequency reference that controls the femtosecond optical "clockwork" [97]. Using self-referencing (Fig. 5), every element of the femtosecond comb, as well as their frequency separation f_r, is phase-coherently connected to the cold atom-based optical frequency standards. With no other frequency reference as an input, a high accuracy, high-stability optical atomic clock is realized in a stable local oscillator (the laser) locked to a narrow atomic reference and a pulsed microwave output that can be recorded with a counter.

One of the JILA optical frequency standards is a diode-pumped solid state Nd:YAG laser (f_{cw}) with its second harmonic ($2f_{cw}$) locked on a hyperfine component of an iodine transition (R(56) 32-0, a_{10}) near 532 nm; this
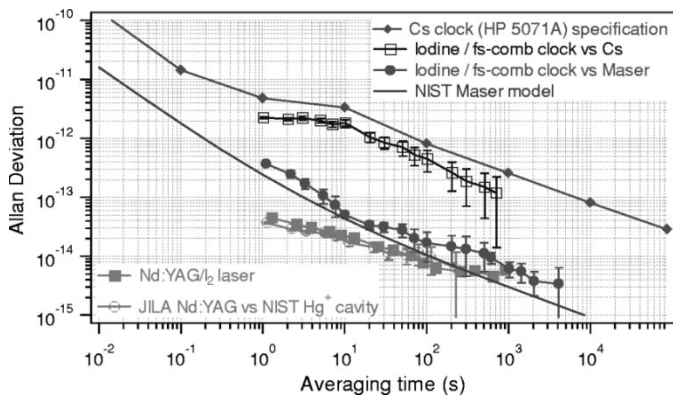
Fig. 8. Summary of various frequency measurements associated with the I_2 optical standard and the subsequently derived RF clock signal. All traces represent Allan deviation. Filled diamonds: Cs atomic clock; Open squares: beat between the I_2 clock and the Cs standard; Filled circles: beat between the I_2 clock and the maser standard; Line: model of the NIST maser standard (ST-22); Filled squares: I_2 standard measured in the optical domain in JILA; Open circles: I_2 standard measured in the optical domain in NIST. (Mention of particular commercial device is for technical clarity and does not imply endorsement.)

system offers an (in-)stability of 4×10^{-14} at 1 s [101]. Using the self-referencing approach, we have achieved not only a millihertz stability for the value of f_0 when measured by a frequency counter, but also a millihertz scale linewidth for f_0. The beat signal between a comb component near 1064 nm and the stabilized Nd:YAG laser is phase locked to an RF signal derived either in a self-consistent manner from f_r, which we want to stabilize, or from a moderately stable RF signal source. The fortuitous wavelength of the I_2/Nd:YAG optical standard is also well suited for realization of an optical clock using the second approach shown in Fig. 7 [98]. In this case, two beat signals, namely the fundamental and the second harmonic of the cw laser against the respective comb components, are appropriately mixed to produce control signals related to f_r and f_0. The frequency/phase variations arising in both f_r and f_0 are, therefore, directly manifested in the two control variables and are directly linked to the optical frequency standard f_{cw}. For clock signal generation, essentially we need to establish phase coherence between f_r and f_{cw}. The variable f_0 can be actually left free-running since it can be effectively taken out of the control equation.

Highly stable microwave/RF frequency standards, such as NIST-maintained hydrogen masers, can be used to characterize these newly demonstrated optical clocks via heterodyne beat. The time record of the beat frequency can be used to determine the Allan variance [102] that displays the frequency noise versus its characteristic time scales. Fig. 8 summarizes the comparison results of an iodine-based optical clock against the Cs and H-maser references. The Allan deviation (square root of Allan variance) reaches 1×10^{-14} at 500 s, slightly worse than the maser itself, most likely due to phase noise in the fiber link used to transfer the maser-based signal from NIST to JILA. The data for the optical standard itself were obtained from heterodyne experiments between two similar laser stabilization systems. With the tracking of the comb system exceeding the stability of the current optical frequency standards, we expect the stability of the derived clock signal to be basically that of the op-

tical standard. However, while NIST experiments in the optical domain comparing two sets of frequency combs both stabilized to a common optical standard confirmed the tracking stability of fs comb to optical standard at the level of 6×10^{-16} at 1 s averaging time, tests performed in the microwave domain have revealed some added noise during the photo-detection process that is limiting the stability of transfer from optical to microwave signals at the $1 \times 10^{-14} \tau^{-1}$ level [103], [104]. Actively studied at present is how to avoid stability degradations when a clock signal is generated in the microwave domain from a phase-stabilized fs comb.

Many other laboratories are developing these optically based atomic clocks, including single-Sr^+-based systems in National Research Council, Canada, and National Physical Laboratory, U.K., Yb^+ at NPL and PTB, and CH_4-stabilized He–Ne lasers at LPTF, Paris, and at IFL and Lebedev, Russia, and Sr-based systems at several labs around the world. Certainly, we still face some technical challenges on the road to making an optical clock a reliable device rather than a scientific experiment. Further developments in technological areas necessary for an advanced optical atomic clock include both oscillators and clockwork, namely, optical frequency standards that are highly accurate and reproducible, along with reliable, stable, and compact ultrafast laser technology [75], [105] for practical implementation of optical clocks. Another important area that needs urgent care is development of highly stable and accurate transmission networks that are suitable for distribution and intercomparison of the next generation atomic frequency standards with unprecedented stabilities. Recent joint work between JILA and NIST has made an initial step toward this direction [106]. A city-wide multi-agency fiber communications network is used to connect optical and radio frequency standards located 3.45 km apart in JILA and NIST laboratories. Active phase compensation is implemented to eliminate fiber-induced propagation noise so that the phase coherence of an optical frequency standard is preserved during the transfer process. The instability of the transfer process is 3×10^{-15} at 1-s averaging time when the phase correction loop is activated. The advantage of direct optical signal transfer and measurement over traditional microwave modulation approaches has been clearly demonstrated.

With the development of an optical atomic clock and phase stabilized fs comb, we have now established an optical frequency grid with lines repeating every repetition frequency (100 MHz –1 GHz) over an octave optical bandwidth and with every line stable at the level of the optical standard. This is useful for a number of applications. However, we often desire a single-frequency optical-"delta"-function (of reasonable power) that can be tuned to any desired frequency on demand. Indeed, it has been a long-term goal in precision optical frequency metrology to construct an optical frequency synthesizer that would allow access in the frequency domain to any optical spectral feature of interest with a well-defined single-frequency optical carrier wave. Realization of such an optical frequency synthesizer (analogous to its radio-frequency counterpart) will add a tremendously useful tool for modern optical experiments. One could foresee an array of diode lasers, each covering a successive tuning range of ~10–20 nm that would collectively cover most of the visible spectrum. Each diode laser frequency

would be controlled by the stabilized optical comb and, therefore, be directly related to the absolute time/frequency standard in a phase coherent fashion, while the setting of the optical frequency will be accomplished via computer control. We have constructed a demonstration system that allows a laser diode (LD) to tune through a targeted spectral region with a desired frequency step size, while maintaining frequency reference to the stabilized optical comb. A self-adaptive search algorithm allows for tuning of the LD frequency to specific values of absolute frequencies along the comb structure. Two fundamental aspects of an optical frequency synthesizer have been demonstrated, namely, continuous, precise frequency tuning and arbitrary frequency setting on demand [107].

IV. TIME DOMAIN AND APPLICATIONS

Prior to the development of femtosecond comb technology, mode-locked lasers were used almost exclusively for time-domain experiments. Although the fs comb technology has primarily impacted the frequency-domain applications described Sections II and III, it is having a strong impact on time-domain experiments and promises to bring about just as dramatic advances in the time domain as it has in optical-frequency metrology and optical clocks. Indeed, it is fascinating to blur the boundary between traditional cw precision spectroscopy and ultrafast phenomena [108]. As discussed below, these applications put stringent requirements on the carrier-envelope phase coherence. Once long-term phase coherence is achieved, comb technology can be used to "stitch" together the output of multiple lasers into a single coherent pulse stream, given sufficiently accurate synchronization of the two lasers. A pulse train with good carrier-envelope phase coherence is also very promising for experiments that are sensitive to ϕ_{CE}, i.e., the "absolute" pulse phase, not just its pulse-to-pulse change. This can be manifest in "extreme" nonlinear optics experiments or coherent control.

A. Carrier-Envelope Phase Coherence—Time-Domain Consequences of Frequency-Domain Control

The carrier-envelope phase coherence is critical for all of the time-domain processes discussed in the remainder of this section. Physically, the carrier-envelope phase coherence simply reflects how well we can tell what the carrier-envelope phase is of a given pulse in the train if we know the phase of an earlier pulse. In order to understand the effect of frequency domain control on the time domain phase coherence, it is useful to first explore the connections between the disciplines of the ultrastable and the ultrafast. The connection between the ultrastable and the ultrafast arises from the fact that fs laser oscillators produce pulses in a periodic train via mode locking, with a corresponding rigorous periodicity in the spectral domain. Mode-locked lasers generate "ultrashort" optical pulses by establishing a fixed phase relationship across a broad spectrum of frequencies. In any dispersive material, the difference between group and phase velocities will cause the carrier-envelope phase, ϕ_{CE}, to evolve. The generation of ultrashort pulses requires that the group velocity (v_g) dispersion inside the laser cavity is minimized across the pulse's frequency spectrum. The

frequency comb spacing (or the repetition frequency) is $f_r = 1/\tau_{r.t.} = v_g/l_c$, where l_c is the laser cavity length and $\tau_{r.t.}$ the cavity round trip time. However, the individual mode frequencies correspond to cavity eigenmodes of the phase-velocity (v_p) of the light. In general, we have $v_g \neq v_p$ due to laser cavity dispersion. This results in a pulse envelope function that is not fixed with respect to the underlying optical oscillation frequencies—there is a phase slip (denoted by $\Delta\phi_{CE}$) between the "carrier" phase and the envelope peak for each of the successive pulses emitted by the laser, with $\Delta\phi_{CE} = (1/\nu_g - 1/\nu_p)l_c\omega_c \mod (2\pi)$ [109], [110]. Here, ω_c is the carrier frequency. In the frequency domain, $\Delta\phi_{CE}$ yields an offset of the comb from the exact harmonics of f_r by the amount $f_0 = \Delta\phi_{CE}f_r/2\pi$ [73], [111], [112]. Hence, each optical comb frequency is effectively given by $f_n = nf_r + f_0$, where n is the integer harmonic number of a particular given optical comb line.

Such an intimate relation between the time and frequency domain dynamics has allowed the extensive tools of frequency domain laser stabilization to be employed for ultrafast optics, with dramatic results. Frequency-domain control of both f_r and f_0 makes it possible to establish a high-precision fs-laser-based optical comb and its applications to optical frequency metrology have already been discussed in Sections II and III. For time-domain experiments, control of the frequency ratio f_0/f_r establishes the evolution of $\Delta\phi_{CE}$. However, establishing a long-term coherence of the carrier-envelope phase requires precise phase control of f_0. In fact, although the cross-correlation measurements presented in [70] demonstrate some degree of phase coherence, they are actually rather insensitive to phase fluctuation because they only measure the change between pulse i and pulse $i + 2$. Instead, frequency domain measurements of the frequency noise spectrum f_0 is a much more sensitive measurement because it uses much longer time intervals. Given a measurement of the frequency noise power spectral density, $S_\nu^{f_0}$, the accumulated root-mean-square fluctuations of ϕ_{CE}, are given by

$$\Delta\phi_{CE}^{RMS}|_{\tau_{obs}} = \sqrt{\int_{f_{obs}}^{f_r/2} (1/f^2)S_\nu^{f_0}(f)df}$$

where the upper bound for the integration is half the pulse repetition rate (Nyquist frequency) and the lower bound corresponds to an observation time $\tau = 1/(2\pi f_{obs})$ [113]. Experimentally, we can now control the carrier phase evolution of a pulse train extended over several minutes, inferred from the fact that the measured linewidth of f_0 is limited by the same measurement time window [114]. Furthermore, the absolute phase of each pulse is also influenced by the propagation media outside of the laser cavity, but the fluctuations in this absolute pulse phase can also be controlled to within 100 mRad over minutes. The frequency-domain-based laser control techniques thus have a profound impact to the time-domain applications, especially those where a direct knowledge or control of the carrier-envelope phase helps to understand or enhance physical effects to be measured. Normally, the absolute phase of an optical wave's electric field is not relevant, in that any shift
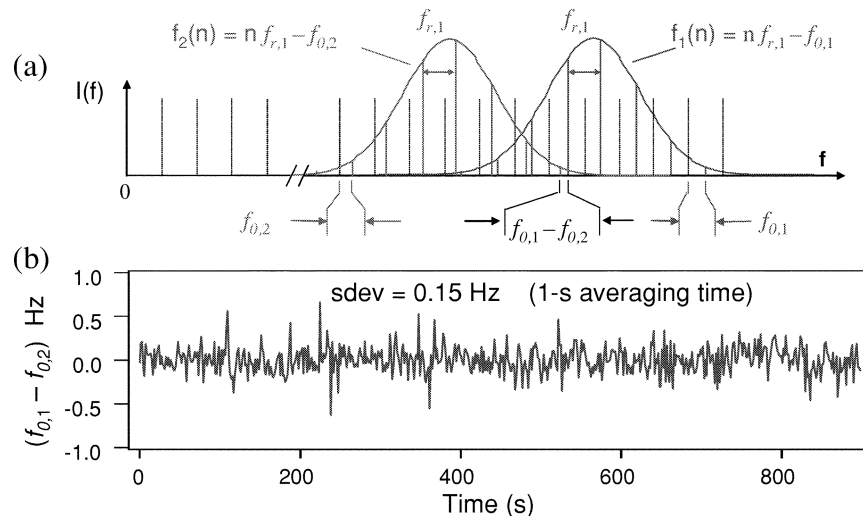
Fig. 9.   (a) Schematic and (b) result of phase locking of two carrier frequencies.

in this phase has no measurable effect. However, within the context of few-cycle pulses, the electric field does not have this invariance. This condition is due to the reference provided by the few-cycle width of the pulse envelope and the value of the electric field's phase (relative to the envelope) drastically alters the optical character of a few-cycle pulse. Examples of extreme nonlinear optics or strong field processes where the absolute phase is critical include coherent x-ray and/or attosecond pulse generations and strong-field ionization.

B. Timing Synchronization and Phase Lock of Mode-Locked Lasers

Our motivation for working with separate ultrafast lasers stems from the desire to create an arbitrary light wave-form generator, with the capability of synchronizing and phase-locking arbitrary separate mode-locked lasers of distinct optical properties in distinct regions of the spectrum with potentially high powers. The ability to generate coherent light across ultrabroad bandwidths is essential for many applications in ultrafast science and technology.

To establish phase coherence between two separate ultrafast lasers, it is necessary to first achieve a level of synchronization between the two lasers such that the remaining timing jitter is less than the oscillation period of the optical carrier wave, namely, 2.7 fs for Ti:S lasers centered around 800 nm. While other techniques are available for synchronization, such as using cross-phase modulation to passively synchronize two mode-locked lasers that share the same intracavity gain medium [115], [116], we have adopted a flexible all-electronic approach [117] for active stabilization of repetition rates to achieve synchronization with sub-fs timing jitter at a 160-Hz bandwidth [118] over an observation period of several seconds. The timing jitter rises to 1.75 fs if the integration bandwidth is increased to 2 MHz, beyond which and up to the Nyquist frequency of 50 MHz (for a 100-MHz repetition rate laser) there is negligible contribution to the timing jitter. Using a configuration of two phase locked loops working at different timing resolutions has allowed ease of operation, enhanced jitter control, rapid

switch of time delay, and working with flexible repetition rates. One limitation to the present performance is due to the intrinsic noise of the microwave mixer used for detection of high order harmonics of f_r. A microwave interferometer for carrier suppression is being constructed to overcome this limit. Reduced timing noise should be achievable by using either a single highly stable cw laser [104] or a stable optical resonator as a reference, extending the harmonic order of (f_r) well into and beyond the terahertz frequency range.

Phase locking of separate fs lasers requires a step beyond tight synchronization of the two pulse trains. One would need effective detection and stabilization of the phase difference between the two optical carrier waves underlying the pulse envelopes [119]. As shown in Fig. 9(a), after synchronization matches the repetition rates $(f_{r1} = f_{r2})$, phase locking requires that the spectral combs of the individual lasers be maintained exactly coincident in the region of spectral overlap so that the two sets of optical frequency combs form a continuous and phase coherent entity. We detect a coherent heterodyne beat signal between the corresponding comb components of the two lasers. When synchronized, the heterodyne beat between the two combs can be recovered with an SNR of 60 dB in a 100-kHz bandwidth. Hundreds of comb pairs contribute to the heterodyne beat signal, and its amplitude is coherently enhanced when synchronized. Such heterodyne detection yields information related to the difference in the offset frequencies of the two lasers, $\delta f_0 = f_{01} - f_{02}$, which can then be controlled. By phase locking δf_0 to a frequency of a mean zero value, we effectively demand that $(\Delta\phi_{01} - \Delta\phi_{02}) = 0$, leading to two pulses trains that have nearly identical phase evolution. Fig. 9(b) shows the recorded δf_0 under phase-locked condition, with a standard deviation of 0.15 Hz at an averaging time of 1 s. This is to be compared against the unlocked case, where the standard deviation of δf_0 is a few megahertz at 1-s averaging.

The established phase coherence between the two fs lasers is also revealed via a direct time-domain analysis. A second-order auto-correlation measurement of the combined pulse clearly demonstrates the aspect of coherent synthesis [120]. For this measurement, the two pulse trains were maximally overlapped
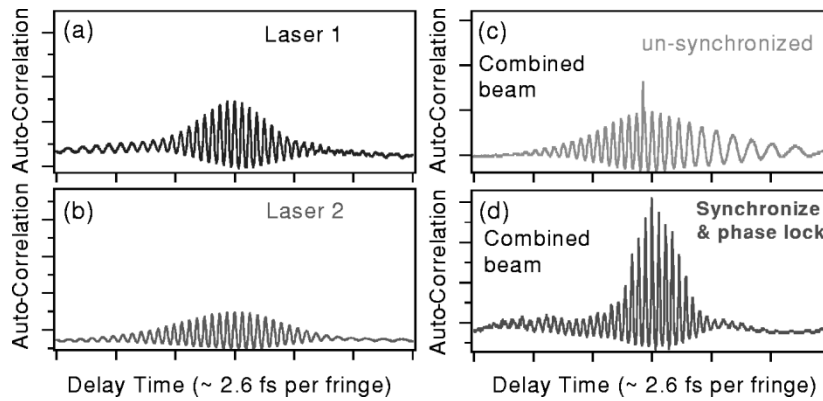
Fig. 10.   Autocorrelation measurement of a coherently synthesized pulse.

in the time domain before the auto-correlator. The auto-correlation curves of each individual laser are shown in Fig. 10(a) and (b), respectively. Fig. 10(c) shows the auto-correlation measurement when the two lasers are not synchronized. Basically, we obtain an auto-correlation of a single pulse train, with a sharp spike appearing in the data at a random position. The spike appears because, at that particular instant, the two pulses overlapped in time and the two electric fields came into phase and coherently added together. The time scale of this random interference is related to the offset frequency between the two repetition rates and is usually less than a few nanoseconds. When the two femtosecond lasers are phase locked, the auto-correlation reveals a clean pulse that is shorter in apparent duration and larger in amplitude [Fig. 10(d)]. This represents a successful implementation of coherent light synthesis, the coherent combination of output from more than one laser in such a way that the combined output can be viewed as a coherent femtosecond pulse being emitted from a single source.

C. New Light Sources and Nonlinear Spectroscopy

The capability of synchronization of a passively mode-locked laser to an external reference, or to a second laser, has many applications. The present level of synchronization would make it possible to take full advantage of the femtosecond time resolution in applications such as high power sum- and difference-frequency mixing [121], novel pulse generation and shaping [122], new generations of laser/accelerator based light sources, or experiments requiring synchronized laser light and x-rays or electron beams from synchrotrons [123]. Indeed, accurate timing of high intensity fields is essential for several important schemes in quantum coherent control and extreme nonlinear optics such as efficient X-ray generation. Two recent applications that have been developed in our laboratories include tunable, subpicosecond pulse generation in the IR [124] and coherent anti-Stokes Raman scattering (CARS) microscopy with two tightly synchronized picosecond lasers [125], [126]. The flexibility and general applicability of the two-laser-synchronization approach are clearly demonstrated in the straightforward generation of programmable light sources for these applications.

Mid-infrared ultrashort light pulses are of primary importance for the study of ultrafast dynamics in chemical reactions,

molecular vibrations, or the application of femtosecond IR spectroscopy to problems in solid-state physics. In particular, experiments in coherent control require a flexible, tunable source for MIR fs pulses. New techniques for controlled pulse shaping in the MIR [127], [128] in combination with such adaptable sources promise to enable abundant progress in the science of coherent control. Previously reported techniques for generating high-repetition-rate MIR sources use frequency mixing of pulse spectrum directly from a Ti:sapphire (Ti:s) laser [129], [130] or from an optical parametric oscillator (OPO) system [131]. Using two highly synchronized passively mode-locked Ti:s lasers allows construction of a flexible and compact source for the generation of femtosecond MIR pulses. The individual tunability of the two Ti:s lasers would, in principle, enable DFG generation spanning a spectral coverage from 3 μm down to the terahertz region. By tuning a GaSe crystal's phase-matching angle as well as the wavelength separation of the two input pulses, MIR pulses from 7.5 to 12.5 μm are generated, with the measured upper (lower) tuning range limited by the MIR monochromator (the nonlinear crystal). The MIR pulse duration is expected to be <400 fs. The average power of the pulses exceeds 15 μW, which, to our knowledge, represents the highest power in the >7.5 μm spectral range obtained from direct DFG of Ti:s pulses. More importantly, precision setting of the time delay between the two original chirped-pulse trains allows rapid switching of the MIR wavelength as well as programmable amplitude modulation. The repetition rate of the MIR source can also be set to a flexible value below that of the original Ti:s lasers by working the two lasers at different repetition rates. Fig. 11 shows the cross-correlation measurement of the two stabilized mode-locked Ti:s lasers using both sum (SFG) and difference frequency generation (DFG).

Another important application is in the field of nonlinear-optics-based spectroscopy and nanoscale imaging. For example, vibrational imaging based on coherent anti-Stokes Raman scattering (CARS) spectroscopy is a powerful method for acquisition of chemically selective maps of biological samples [132]. In CARS microscopy, pulsed pump and Stokes beams are focused tightly to a single focal spot in the sample to achieve a high spatial resolution. The third-order nonlinear interaction produces a signal photon that is blue-shifted (anti-Stokes signal) with respect to the incident beams. Strong CARS signals are obtained whenever the frequency difference between the pump
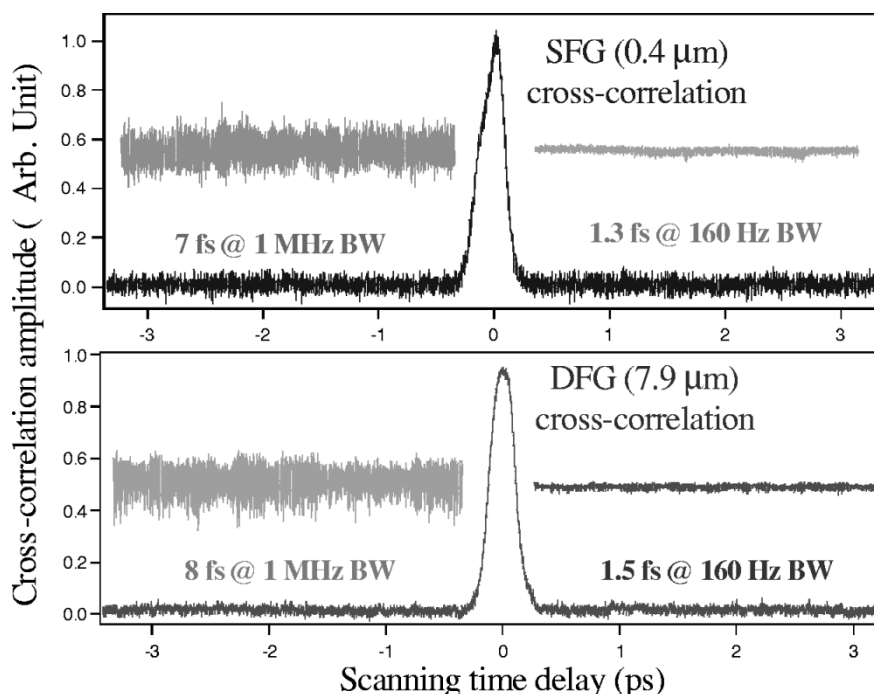
Fig. 11. Simultaneous sum and difference frequency generations from two stabilized femtosecond lasers. Noise lines in the figure represent timing jitter between the two femtosecond lasers, measured with the optical cross correlation technique.

and Stokes coincides with a Raman-active vibrational mode, which gives rise to the molecule-specific vibrational contrast in the image. Practical applications of the CARS microscopy technique require pulsed light sources; pulses with temporal widths of 1–2 ps are ideal for matching to the vibration bandwidths in order to optimize the CARS signal, with minimized nonresonant background and compromising spectral resolution [133]. Also, the laser system employed should deliver at least two different frequencies, ω_p (pump) and ω_s (Stokes), such that $\omega_p - \omega_s$ can be tuned between 500 and 3500 cm^{-1}, corresponding to the biologically interesting region of the vibrational spectrum. Using two independently optical oscillators, an important technical challenge is control of the time coincidence of the individual laser pulses. The strength of the CARS signal depends directly on the temporal overlap of the incident pulses, and any timing-jitter among the relevant pulses will lead to signal fluctuations that will, in turn, degrade the image. Tight synchronization of two independent picosecond lasers with residual timing jitter amounting to less than 1% of the pulse duration eliminates all the jitter-related noise from the CARS images and significantly enhances the image quality [126]. For example, using two tightly synchronized picosecond lasers, we are able to achieve significant improvements in experimental sensitivity and spatial resolutions for CARS microscopy [125], as evidenced in Fig. 12.

D. Extreme Nonlinear Optics and Coherent Control

The expression "extreme nonlinear optics" refers to experiments where the optical pulses are so intense (and the pulse duration so short) that the electric field of the pulse becomes a relevant physical quantity [122]. In this regime, the electric field is strong enough to distort the potential well for an electron bound to an atom to such a large extent that "above-threshold
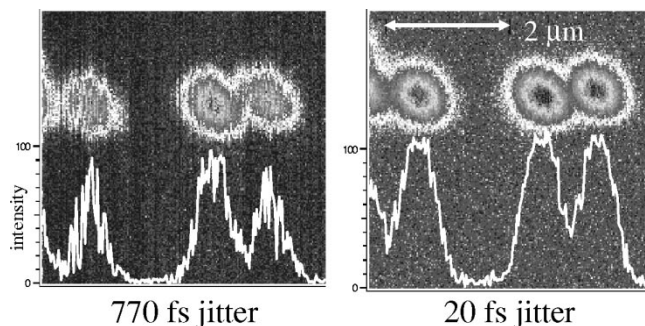

Fig. 12. CARS image of 1-μm polystyrene beads under two different synchronization conditions. Laser frequency difference matches the Raman shift of 1600 cm^{-1}.

ionization" occurs, which typically displays a threshold with respect to the electric field of the pulse. A dependence of this physical process on the pulse carrier-envelope phase is, therefore, expected for sufficiently short pulses and if the threshold is close to the maximum field in the pulse. Direct evidence for such phase dependence has been observed in high harmonic generation [134]. Photoelectron yields measured in opposite directions have also produced evidence for a phase dependence in above-threshold ionization [135]. The development of ultrafast lasers has also led to coherent control in molecular and reaction dynamics [136], [137]. Many of the techniques are sensitive to the phase of the applied fields. To date, only the relative phase between two laser fields, or the relative internal phase of a femtosecond pulse, has been demonstrated to have physical impacts. Some new high-order nonlinear schemes explore interference between pathways involving n photons and m photons in certain interband transitions in semiconductor material. When n and m have opposite parity, a dependence on ϕ_{CE} will occur for excitation by single ultrashort pulse. The use of a single pulse

with known ϕ_{CE} has not been demonstrated; however, the interference phenomenon has been demonstrated by using a pair of phase controlled pulses to ionize rubidium [138] and control of electrical currents in bulk semiconductors [139]. In both cases, there is a connection between spatial direction and the relative phase ϕ_{CE}. These coherent control phenomena present an interesting means of measuring ϕ_{CE} as well as a potential application of phase-controlled pulses.

With phase control of laser oscillators now implemented, the next logical step is a phase-stabilized pulse amplifier [140] to realize the full potential in extreme nonlinear optics and novel coherent processes. Ordinarily, the peak power offered by pulses emitted from a simple oscillator is not sufficient to drive the high-order nonlinear processes of interest. However, it has recently been shown by Baltuška *et al.* that conventional amplifiers can be used to amplify ultrashort optical pulses while simultaneously controlling the carrier-envelope offset frequency [134]. We have proposed and demonstrated a novel alternative approach for pulse amplification without the use of an active gain medium [141]. Our method relies on coherent superposition of successive components from a pulse train to increase the amplitude of a single pulse while reducing the repetition frequency. This requires not only a suitable delay mechanism to line up successive pulses, but also the ability to control the phase evolution of the electric field lying under the pulse. These requirements are similar to those in work already demonstrated in coherent pulse synthesis from separate femtosecond lasers [120]. An amplification scheme based on coherent addition would maintain the carrier-envelope phase coherence of the original oscillator.

A passive optical cavity is an ideal candidate to temporarily store and coherently enhance the pulsed electric field. To ensure efficient coupling into the cavity and subsequent power buildup, the repetition rate and carrier-envelope phase of the input pulses must match that of the pulse circulating inside the passive cavity. The equivalent frequency domain requirement is that all frequency components making up the pulse train are tuned into resonance with corresponding cavity modes. The cavity decay time is directly proportional to the overall cavity finesse and is predetermined to match with the desired pulse amplification factor. An important technical issue is the dispersion control of the passive buildup cavity. This scheme has now been implemented for a picosecond laser amplifier. With unoptimized cavity mirrors, our passive approach has already achieved a factor of 50 pulse amplification in the switched output [148], which is already more than a factor of five better than the output of a cavity dumped Ti:s laser. The work on a "gainless" femtosecond laser amplifier is also in progress.

V. SUMMARY

Thanks to the development of self-referenced optical frequency combs, the ability to measure and synthesize arbitrary optical frequencies with high precision is now well established. When combined with optical atomic frequency references these systems are beginning to provide optical sources with high stability, accuracy, and well-controlled phase. The ability to control of the optical frequency accurately, and to some

degree even the optical phase for extended periods of time, is a relatively new capability that is opening new opportunities in science, metrology, and other applications. We have tried to emphasize that the methods of connecting, measuring, and converting optical frequencies have broader applications than just frequency metrology, and they are beginning to have an impact on other areas, such as time-domain spectroscopy, reduced timing jitter, advanced communications, and tests of fundamental physics. Precise coherent control of optical fields obtained by referencing optical frequency combs to atomic transitions can simultaneously provide high spectral resolution, extremely broad spectral coverage, and correspondingly high temporal resolution and control. The technologies and applications are advancing very rapidly, as will be evidenced by other papers in this special issue. The amazing rate of progress is due in large part to the newly synergistic interactions between the scientific fields of precision spectroscopy, laser cooling and trapping, metrology, and ultrafast optics. The combined skills and broader perspective brings new light to optical sciences and new tools for the future.

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