Colloquium: Femtosecond optical frequency combs

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(Published 10 March 2003)

Recently there has been a remarkable synergy between the technologies of precision laser stabilization and mode-locked ultrafast lasers. This has resulted in control of the frequency spectrum produced by mode-locked lasers, which consists of a regular comb of sharp lines. Thus such a controlled mode-locked laser is a "femtosecond optical frequency comb generator." For a sufficiently broad comb, it is possible to determine the absolute frequencies of all of the comb lines. This ability has revolutionized optical frequency metrology and synthesis. It has also served as the basis for the recent demonstrations of atomic clocks that utilize an optical frequency transition. In addition, it is having an impact on time-domain applications, including synthesis of a single pulse from two independent lasers. In this Colloquium, we first review the frequency-domain description of a mode-locked laser and the connection between the pulse phase and the frequency spectrum in order 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 a brief overview of how the comb technology is affecting and will affect time-domain experiments.

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I. INTRODUCTION

Mode-locked lasers generate ultrashort optical pulses by establishing a fixed phase relationship across a broad spectrum of frequencies. Progress in the technology of mode-locked lasers has resulted in the generation of optical pulses that are only 5 fs in duration (Morgner *et al.*, 1999; Sutter *et al.*, 1999), which corresponds to less than two cycles of the laser light. Although mode locking is a frequency-domain concept, mode-locked lasers and their applications are typically discussed in the time domain. Recently, a paradigm shift in the field of ultrafast optics has been brought about by switching to a frequency-domain treatment of the lasers and the pulse trains that they generate. Understanding mode-locked lasers in the frequency domain has allowed the extensive tools of frequency-domain laser stabilization to be employed, with dramatic results.

The central concept to these advances is that the pulse train generated by a mode-locked laser has a frequency spectrum that consists of a discrete, regularly spaced series of sharp lines, known as a frequency comb. As described below, if the comb spectrum is sufficiently broad, it is possible to directly measure the two radio frequencies (rf) that describe the comb. This fact has had immediate impact in the field of optical frequency metrology/synthesis[1] and has been enabling for the recent demonstration of optical atomic clocks (Diddams *et al.*, 2001; Ye *et al.*, 2001). Because the comb spectrum can be related to phase evolution in the pulse train (Apolonski *et al.*, 2000; Jones *et al.*, 2000), these results also promise important advances in ultrafast science, specifically extreme nonlinear optics (Brabec and Krausz, 2000) and coherent control. In addition, the union of time and frequency-domain techniques has yielded remarkable results in pulse synthesis (Shelton *et al.*, 2001).

The idea that a regularly spaced train of pulses corresponds to a comb in the frequency domain and can

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[1]There have been a large number of results in the last two years. A few of the more notable ones are reported by Diddams, Jones, Ye, *et al.* (2000), Holzwarth *et al.* (2000), Jones *et al.* (2000), Stenger, Binnewies, *et al.* (2001), and Udem *et al.* (2001). For a more extensive compilation, see Cundiff *et al.* (2001).

thereby excite narrow resonances was realized more than 20 years ago by Hänsch (1976) and Baklanov and Chebotayev (1977). Teets *et al.* (1977) used a train of pulses generated externally to the laser; however, it was quickly realized that mode-locked lasers were superior, as demonstrated in a measurement of the sodium hyperfine splitting using a picosecond laser by Eckstein *et al.* (1978). Some of the concepts being developed today were described in these early papers, although the technology was insufficient to demonstrate them at the time. Advances in mode-locked laser technology (specifically the Kerr-lens mode-locked Ti:sapphire laser) renewed interest in this area (Udem *et al.*, 1999a, 1999b; Diddams, Jones, Ye, *et al.*, 2000). The observation of supercontinuum generation from nanojoule pulses in microstructured fiber by Ranka *et al.* (2000) led to the recent, sudden explosion in activity.

In the following, we first discuss the time- and frequency-domain pictures of mode-locked lasers and specifically how phase evolution of the pulses is manifest as a frequency shift. This provides the background for understanding how the absolute frequencies of the comb lines are determined, which in turn is the basis for performing absolute optical frequency metrology using mode-locked lasers. Absolute optical frequency metrology requires that optical frequencies be generated based on the microwave hyperfine transition of cesium that is used to define the second. Optical clocks are in some sense the inverse problem, using an optical frequency quantum transition as the clock oscillator and deriving microwave outputs. Returning to the time domain, we then discuss how frequency combs can be used to synthesize ultrashort pulses from two independent lasers. This is followed by a discussion of how phase stabilization is expected to impact ultrafast science, specifically extreme nonlinear optics and coherent control.

II. TIME- AND FREQUENCY-DOMAIN PICTURES OF A MODE-LOCKED LASER

Understanding the connection between the time-domain and frequency-domain descriptions of a mode-locked laser and the pulse train that it emits is crucial. In this section, we first briefly introduce mode-locked lasers to provide the necessary background. This is followed by a discussion of how, given a spectrum that spans an octave, to determine the frequency spectrum of the pulse train emitted by a mode-locked laser and how the absolute frequencies of the comb spectrum can be determined. Based on this, a prototype femtosecond comb generator is presented along with time domain cross-correlation measurements.

A key concept in this discussion will be the carrier-envelope phase. This is based on the decomposition of the pulses into an envelope function $E(t)$ that is superimposed on a continuous carrier wave with frequency ω_c, so that the electric field of the pulse is written $E(t)=\hat{E}(t)e^{i\omega_c t}$. The carrier-envelope phase, ϕ_{ce}, is the phase shift between the peak of the envelope and the closest peak of the carrier wave, as illustrated in Fig. 1.

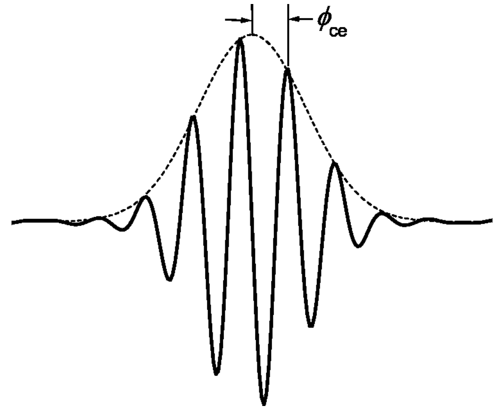

FIG. 1. An ultrashort optical pulse showing the carrier and envelope.

In any dispersive material, the difference between group and phase velocities will cause ϕ_{ce} to evolve as the pulse propagates.

A. Introduction to mode-locked lasers

Mode-locked lasers generate short optical pulses by establishing a fixed phase relationship between all of the lasing longitudinal modes (see Fig. 2) [for a textbook level discussion, see Diels and Rudolph (1996)]. Mode locking requires a mechanism that results in higher net gain for short pulses compared to continuous-wave (cw) operation. This can be done either by means of an active element or passively by means of a saturable absorption (real or effective). Passive mode locking yields the shortest pulses because, up to a limit, the self-adjusting mechanism becomes more effective as the pulse shortens (Ippen, 1994). Real saturable absorption occurs in a material with a finite number of absorbers, for example, a dye or semiconductor. The shortness of the pulses is limited by the finite lifetime of the excited state. Effective saturable absorption typically utilizes the nonlinear index of refraction of some material together with spatial effects or interference to produce higher net gain for shorter pulses. The ultimate limit on minimum pulse duration in such a mode-locked laser is due to interplay

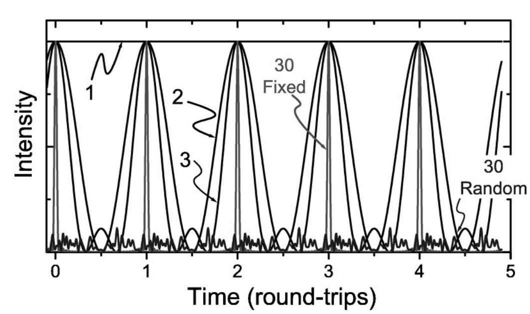

FIG. 2. Schematic of the pulse train generated by locking the phase of simultaneously oscillating modes. The output intensity for 1, 2, 3, and 30 modes is shown. The result for 30 unlocked modes is also shown.

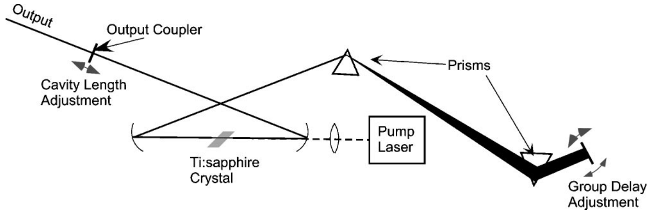

FIG. 3. Schematic of a typical Kerr-lens mode-locked Ti:sapphire laser.

between the mode-locking mechanism, group velocity dispersion (GVD), and net gain bandwidth (Ippen, 1994).

Currently, the generation of ultrashort optical pulses is dominated by the Kerr-lens mode-locked Ti:sapphire (KLM Ti:sapphire) laser because of its excellent performance and relative simplicity. Figure 3 depicts a typical KLM Ti:sapphire laser. The Ti:sapphire crystal is pumped by green light from either an Ar^+-ion laser or a diode-pumped solid-state (DPSS) laser, which provides far superior performance. The Ti:sapphire crystal provides gain and serves as the nonlinear material for mode locking. Prisms compensate the GVD in the gain crystal (Fork *et al.*, 1984). Alternatively, dispersion compensating mirrors, known as chirped mirrors, can be used (Szipocs *et al.*, 1994; Xu, Spielmann, Krausz, *et al.*, 1996). Since the discovery of KLM (Negus *et al.*, 1991; Spence *et al.*, 1991), the pulse width obtained directly from the mode-locked laser has been shortened by approximately an order of magnitude by first optimizing the intracavity dispersion (Asaki *et al.*, 1993) and then using double chirped mirrors (Morgner *et al.*, 1999; Sutter *et al.*, 1999), yielding pulses that are less than 6 fs in duration, i.e., less than two optical cycles. Recently, output spectra that span an octave (factor of 2 in frequency) have been obtained directly from a mode-locked laser (Ell *et al.*, 2001), which is an important accomplishment for phase stabilization. Kerr-lens mode locking is based on a combination of self-focusing in the Ti:sapphire crystal and an aperture that selects the spatial mode corresponding to the presence of self-focusing. A textbook level discussion can be found in Silfvast (1996), while Magni *et al.* (1995) have provided a detailed analysis.

B. Frequency spectrum of a mode-locked laser

To understand how frequency-domain techniques can be used to control mode-locked lasers, we must first connect the time- and frequency-domain descriptions. Here we give an intuitive derivation of the spectrum; see Cundiff (2002) for a rigorous derivation.

To start, we ignore the carrier-envelope phase and assume identical pulses. If we just consider a single pulse, it will have a spectrum that is the Fourier transform of its envelope function and is centered at the optical frequency of its carrier. Generally, for any pulse shape, the frequency width of the spectrum will be inversely proportional to the temporal width of the envelope. For a train of identical pulses, separated by a fixed interval, the spectrum can easily be obtained by a Fourier series expansion, yielding a comb of regularly spaced frequencies, where the comb spacing is inversely proportional to time between pulses, i.e., it is the repetition rate of the laser that is producing the pulses.

To include the effect of the carrier-envelope phase, including the possibility that it evolves from pulse to pulse, it is useful to first understand how the comb spectrum arises. The Fourier relationship between time and frequency resolution guarantees that any spectrometer with sufficient spectral resolution to distinguish the individual comb line cannot have enough temporal resolution to separate successive pulses. Furthermore, this means that the successive pulses interfere with each other inside the spectrometer. The comb spectrum occurs because there are certain discrete frequencies at which the interference is constructive. Using the result from Fourier analysis that a shift in time corresponds to a linear phase change with frequency, we can readily see how the interference occurs. In Fig. 4(a), the spectral phase is plotted (modulo 2π) for pulses arriving at $t=0$, τ, 2τ, and 4τ. The $t=0$ pulse has an overall phase of $\pi/4$, this is arbitrary and chosen just for graphical convenience. It is clear that all four pulses have the same spectral phase at frequencies that are integer multiples of the repetition rate $f_{\mathrm{rep}}=1/\tau$. Thus they coherently add in the spectrometer, whereas at other frequencies they will not. Now we can include the carrier-envelope phase; we first show in Fig. 4(b) that shifting the phase of all the pulses does not change the comb frequencies, i.e., the frequency at which they all have the same phase is unchanged. In Fig. 4(c), we include an evolving carrier-envelope phase; the phase of each pulse increments by an amount $\Delta\phi_{\mathrm{ce}}$, which is $\pi/4$ in the figure. This shows the central result—evolution of the carrier-envelope phase results in a rigid shift of the frequencies at which the pulses add constructively and thus a shift of the frequency comb. Thus the optical frequencies ν_n of the comb lines can be written as

$$\nu_n = n f_{\mathrm{rep}} + f_0, \tag{1}$$

where n is a large integer of order 10^6 that indexes the comb line, and f_0 is the comb offset due to pulse-to-pulse phase shift. The comb offset is connected to the pulse-to-pulse phase shift by

$$f_0 = \frac{1}{2\pi} f_{\mathrm{rep}} \Delta\phi_{\mathrm{ce}}. \tag{2}$$

[Note that because of sign conventions, the pulse-to-pulse phase change used to construct Fig. 4(c) is actually negative, yielding a value of $f_0 = -f_{\mathrm{rep}}/8$, which is equivalent to the value of $f_0 = \frac{7}{8} f_{\mathrm{rep}}$ obtained in the figure.] The relationship between time- and frequency-domain pictures is summarized in Fig. 5.

The pulse-to-pulse change in the phase for the train of pulses emitted by a mode-locked laser occurs because the phase and group velocities inside the cavity are different. The difference in the velocities in turn arises because of dispersion in the optical elements. Since the

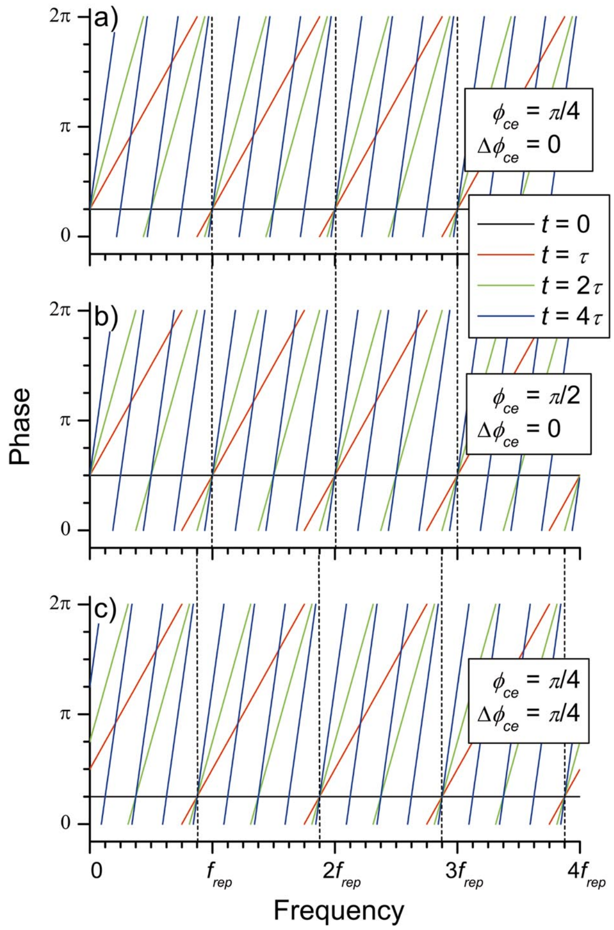

FIG. 4. Schematic showing the how pulse phase is manifest in the frequency spectrum. (a) Spectral phases of pulses arriving at $t=0$, τ, 2τ, and 4τ. These pulses will constructively interfere in a spectrometer at frequencies for which they all have the same phase (dashed vertical lines). (b) Adding a constant phase to all of the pulses does not change the constructive interference frequencies. (c) If the phase changes incrementally from pulse to pulse, the constructive interference frequencies shift (Color).

pulse is sampled once per round trip when it hits the output coupler, it is only the phase change modulo 2π that matters. Specifically,

$$\Delta\phi_{ce}=\left(\frac{1}{\nu_g}-\frac{1}{\nu_p}\right)l_c\omega_c \bmod 2\pi, \tag{3}$$

where ν_g (ν_p) is the mean group (phase) velocity in the laser cavity, l_c is the length of the laser cavity, and ω_c is the carrier frequency.

C. Determining absolute optical frequencies with octave spanning spectra

Armed with our understanding of the frequency spectrum of a mode-locked laser, we can now turn to the question of measuring the absolute frequencies of comb lines. But first, what do we mean by an absolute optical frequency? For a frequency measurement to be absolute, it must be referenced to the hyperfine transition of

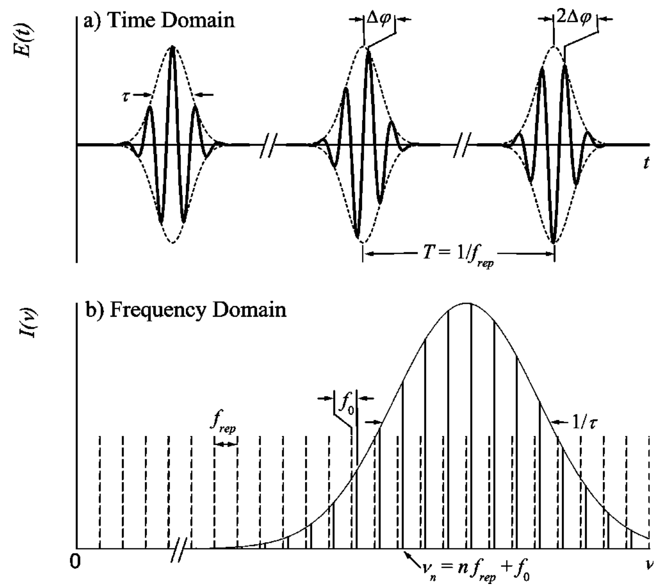

FIG. 5. Summary of the time-frequency correspondence for a pulse train with evolving phase.

^{133}Cs that defines the second. This microwave frequency is defined to be exactly 9 192 631 770 Hz, which is approximately a factor of 10^5 smaller than optical frequencies. Thus a large frequency gap must be spanned to make an absolute optical frequency measurement. Further details on absolute optical frequency metrology will be given in Sec. III. For time-domain experiments, determining the ratio f_0/f_{rep} provides a measurement of the evolution of ϕ_{ce}, which has interesting ramifications. Note that interferometers, such as a grating spectrometer or Fabry-Perot cavity, do not provide absolute frequency measurements because the length must be known absolutely, and the speed of light, which includes time, is used for the definition of length. This leads to a circular definition.

From Eq. (1) we see that determining the absolute optical frequencies of the femtosecond comb requires two rf measurements, that of f_{rep} and f_0. Measurement of f_{rep} is straightforward; we simply detect the pulse train with a fast photodiode. This is typically around 100 MHz; however, it can range from tens of MHz to several GHz. However, measurement of f_0 is not so simple; the phase does not come into the measurement of intensity, and, as we saw above, f_0 is connected to $\Delta\phi_{ce}$.

The connection between f_0 and $\Delta\phi_{ce}$ suggests an interferometric measurement. Consider the case where the optical spectrum spans an octave in frequency, i.e., the highest frequencies are a factor of 2 larger than the lowest frequencies. If we now use a second harmonic crystal to frequency double a comb line, with index n, from the low-frequency portion of the spectrum, it will have approximately the same frequency as the comb line on the high-frequency side of the spectrum with index $2n$. Measuring the heterodyne beat between these yields a difference frequency

$$2f_n-f_{2n}=2(nf_{rep}+f_0)-(2nf_{rep}+f_0)=f_0, \tag{4}$$

which is just the offset frequency. Thus an octave spanning spectrum enables simple measurement of f_0. Note that an octave spanning spectrum is not required, it is just simplest (a number of variations are discussed by Telle *et al.*, 1999). We designate these schemes as self-referencing as they only use the output of the mode-locked laser.

Self-referencing is not the only means of determining absolute optical frequencies given an octave spanning spectrum. For example, the absolute optical frequency of a cw laser can be determined if its frequency lies close to comb line n in the low-frequency portion of the femtosecond comb spectrum. Then the second harmonic of the cw laser will lie close to the comb line $2n$. Measurement of the heterodyne beat between the cw laser frequency f_l and the frequency of comb line n gives $f_{1b} = f_l - (nf_{\mathrm{rep}} + f_0)$ and between the second harmonic of the cw laser and comb line $2n$ gives $f_{2b} = 2f_l - (2nf_{\mathrm{rep}} + f_0)$. Taking the difference,

$$f_{2b} - f_{1b} = 2f_l - (2nf_{\mathrm{rep}} + f_0) - [f_l - (nf_{\mathrm{rep}} + f_0)] = f_l - nf_{\mathrm{rep}}, \quad (5)$$

yields f_l in terms of the heterodyne beat frequencies, the repetition rate and a large integer n. Given *a priori* knowledge of f_l to within $f_{\mathrm{rep}}/4$, n can easily be determined, and thus f_l to much larger accuracy. Without additional information, n can be determined by systematically varying f_{rep}, although a more complex algorithm is required.

D. Femtosecond optical frequency comb generator

A frequency comb generator produces a spectrum that consists of a series of equally spaced sharp lines with known frequencies. Microwave comb generators are commercially available. Optical frequency comb generators have been constructed by injecting a single-frequency cw laser into a high-quality optical cavity that contains an electro-optic modulator (EOM) (see, for example, Kourogi *et al.*, 1993). Typically comb bandwidths of a few THz have been achieved using this method (Ye *et al.*, 1997). If the absolute optical frequency of the cw laser is known, then the resulting comb can be used to directly measure nearby frequencies (Nakagawa *et al.*, 1996). Alternatively, without needing to know the absolute frequency of the cw laser, a comb generator can be used to span a frequency gap in a more complex chain (Hall *et al.*, 1999; Udem *et al.*, 2000).

Based on our discussion above, it is clear that a mode-locked laser also generates an optical comb. However, there is no equivalent to the cw laser used in the EOM scheme, which can provide *a priori* knowledge about the absolute frequencies of the comb. Thus the first applications of mode-locked lasers were the second application of spanning intervals in more complex chains (Udem *et al.*, 1999a, 1999b) or between a known and unknown frequency (Diddams, Jones, Ma, *et al.*, 2000).

The discussion in Sec. II.C shows that knowledge of the absolute frequencies of the comb generated by a mode-locked laser can be easily obtained if it generates a spectrum that spans an octave. A Fourier-transform limited pulse with a full width at half maximum (FWHM) bandwidth of an octave centered at 800 nm would only be a single optical cycle in duration. Such short pulses have not been achieved; the shortest pulses generated by a mode-locked oscillator (i.e., not including external amplification and broadening) are just under two cycles in duration (Morgner *et al.*, 1999; Sutter *et al.*, 1999; Ell *et al.*, 2001). Fortunately, neither a transform limited pulse nor a FWHM of an octave is needed to implement the methods for obtaining the absolute frequencies. The pulse width is unimportant as the methods are purely frequency-domain techniques. Experimentally, it has been found that even if the power at the octave spanning points is 40 dB below the peak, it is still possible to observe strong f-to-$2f$ heterodyne beats.

Since Ti:sapphire, which has the broadest gain bandwidth of all known laser media, does not support an octave spanning spectrum, additional spectrum must be generated. This is accomplished by using self-phase modulation, which is based on a temporal variation in the index of refraction due to a combination of a short optical pulse and an intensity-dependent index of refraction (Agrawal, 1995). This can be external to the laser cavity by using an optical fiber, or internally by creating secondary coincident time and space foci (Ell *et al.*, 2001). The latter technique requires carefully designed mirrors (Kärtner *et al.*, 2000) and is challenging to implement. Therefore the former is significantly more common. Recent results have shown that additional spectral bandwidth can be obtained by minor changes in the cavity configuration of a high repetition rate laser, although it has not yet yielded sufficient intensity at the octave points for observation of f-to-$2f$ beats (Bartels and Kurz, 2002).

The amount of spectral broadening that can be obtained in ordinary optical fiber is limited, primarily because temporal spreading of the pulse, due to group velocity dispersion in the fiber, reduces the peak intensity. Using a low repetition rate laser, to raise the pulse energy, an octave-spanning spectrum has been obtained with ordinary fiber (Apolonski *et al.*, 2000). The discovery by Ranka *et al.* (2000) that microstructure fiber can have zero group velocity dispersion within the emission spectrum of a Ti:sapphire laser eliminated this difficulty and led to rapid progress in the field of femtosecond optical frequency combs by allowing broadband continuum generation with only nanojoule pulse energies. Typical results are shown in Fig. 6.

Microstructure fiber utilizes air holes surrounding a fused silica core to obtain the index of refraction contrast needed for waveguiding. This results in a much larger index contrast than can be obtained using doping. The large index contrast has two consequences, first the ability to generate a zero in the GVD at visible or near-infrared wavelengths and, second, the possibility of using a much smaller core size. The first means that the pulse does not spread temporally, and hence maintains its high peak power. In addition, it results in phase

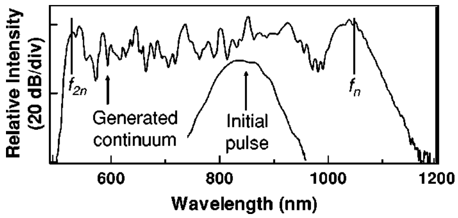

FIG. 6. Input and output spectra of a microstructured fiber (reproduced from Jones *et al.*, 2000).

matching between the generated spectral components. The second also greatly increases the light intensity in the core, thereby enhancing nonlinear effects.

We have now introduced all of the concepts and components needed to construct a femtosecond optical frequency comb generator that produces known absolute frequencies. There are several possible implementations; we will discuss the particular implementation in Fig. 7.

The heart of the comb generator is a KLM Ti:sapphire laser. A small portion of the output is detected using a high-speed photodiode to measure the repetition rate. Greater precision is obtained by measuring a large harmonic of the repetition rate, not the fundamental. A servo loop controls the repetition rate of the laser by comparing this signal to a microwave clock.

The output of the KLM Ti:sapphire laser is launched into a length of microstructure fiber. It has been found that using a minimum possible amount of spectral broadening in the microstructure fiber is best, and thus all metrology experiments start with KLM Ti:sapphire lasers that produce pulse widths of 30 fs or less. This generally results in an octave spanning spectrum for modest pulse energies and short lengths of microstructure fiber. The output of the microstructure fiber is split into two parts. One part serves as the useful output of the comb generator, while the other part is used in a f-to-$2f$ interferometer to measure f_0.

The input to the f-to-$2f$ interferometer is divided into long- and short-wavelength portions by a dichroic beam splitter. The long-wavelength portion is frequency doubled by a second harmonic crystal. The light in the two arms of the interferometer, which now have the same frequency, is recombined and detected with a photodiode. Note that the lengths of the two arms must be matched so that there is temporal overlap, including GVD in the microstructure fiber.

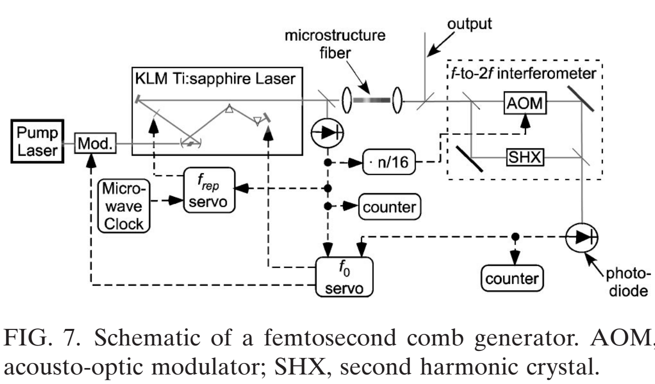

FIG. 7. Schematic of a femtosecond comb generator. AOM, acousto-optic modulator; SHX, second harmonic crystal.

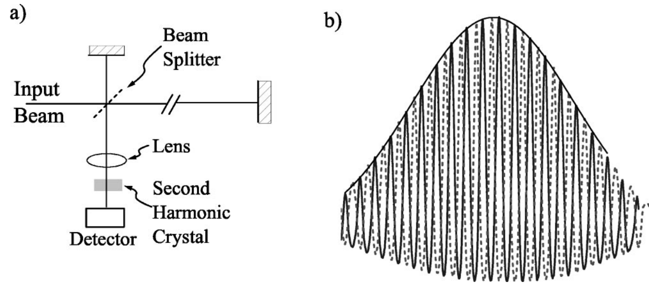

FIG. 8. Time-domain demonstration of phase stabilization. (a) Schematic of a cross correlator. (b) Two typical cross correlations showing a π phase change in $\Delta\phi_{ce}$.

The detected signal from the f-to-$2f$ interferometer contains a forest of signals including multiples of f_{rep} and f-to-$2f$ beat-note signals spaced above and below each repetition rate signal by f_0. One of the beat-notes must be chosen and isolated for counting and stabilizing the laser. If the signal-to-noise ratio is sufficiently large, an appropriate rf bandpass filter is sufficient; otherwise, regeneration with a tracking oscillator is usually employed.

The final step is to close the loop to stabilize f_0. This requires a "knob" on the laser that can be used to adjust f_0, which is determined by the difference between the intracavity group and phase velocities. One common method for adjusting f_0 is to swivel the end mirror in the arm of the laser cavity that contains the prism sequence (Reichert *et al.*, 1999), as shown in Fig. 3. Since the spectrum is spatially dispersed on this mirror, a small swivel produces a linear phase delay with frequency, which is equivalent to a group delay. An alternative method of controlling f_0 is via modulation of the pump power (Apolonski *et al.*, 2000; Poppe *et al.*, 2001). Empirically, this clearly causes a change in f_0 (Xu, Spielmann, Poppe, *et al.*, 1996). However, the details are somewhat unclear, with likely contributions from the nonlinear phase, spectral shifts, and the intensity dependence in the group velocity (Haus and Ippen, 2001). Each method has advantages and disadvantages with respect to servo speed and impact on amplitude noise.

E. Cross correlation: Time-domain measurement of f_0

The connection between time and frequency can be readily illustrated by measuring the cross correlation between successive pulses emitted by the comb generator. A cross correlator is based on the interferometric autocorrelator commonly used to measure ultrashort pulses (Diels and Rudolph, 1996), with the modification that one arm of the interferometer is longer than the other by a multiple of the cavity round-trip time (Xu, Spielmann, Poppe, *et al.*, 1996). A schematic of a cross correlator is shown in Fig. 8(a). An autocorrelation is always symmetric. However, in a cross correlator only the envelope is symmetric, and the shift of the fringes from the peak of envelope is due to the pulse-to-pulse phase shift, $\Delta\phi_{ce}$. By locking f_0 at different points, differing values of $\Delta\phi_{ce}$ are obtained, as per Eq. (2). This is shown in Fig. 8(b) where two cross correlations are shown for f_0 differing by $f_{\rm rep}/4$, yielding a shift of π in $\Delta\phi_{\rm ce}$. (Note that the correlator used to take these data has one arm that is two cavity lengths longer than the other, leading to a doubling of the measured phase shift.) Systematic variation of f_0 yields exactly the expected variation in $\Delta\phi_{\rm ce}$ (Jones *et al.*, 2000).

III. METROLOGY AND OPTICAL CLOCKS USING MODE-LOCKED LASERS

It is useful to quickly review the historical development in optical frequency metrology and its perspectives in precision spectroscopy, clock signal generation, and frequency synthesis. The outstanding spectral properties of optical frequency standards offer unprecedented resolution and precision and potentially the highest accuracy for physical measurements (Bergquist, 1996). Even at the cost of extraordinary complexity and remarkable resources, researchers have explored and constructed optical frequency synthesis chains that span the vast frequency gap between the optical and microwave spectral regions.[2] Previous effort has concentrated on measurement of a few discrete optical lines that are chosen to be optical frequency standards. Even limited frequency measurement capabilities have brought a number of significant advances in fundamental physics including determination of the speed of light (Barger and Hall, 1973; Evenson *et al.*, 1973; Jennings *et al.*, 1986) and linkage between the fundamental physical units of length and time; refinement of the Rydberg constant and the Lamb shift (Udem *et al.*, 1997; Schwob *et al.*, 1999) predicted by quantum electrodynamics; competitive measurement of various fundamental constants such as the fine structure constant (Peters *et al.*, 1997; Udem *et al.*, 1999a, 1999b) and the ratio of proton to electron mass (de Beauvoir *et al.*, 2000); and test of the relativity theory (Hils and Hall, 1990). However, until recently it was deemed an overwhelming challenge to synthesize arbitrary absolute optical frequencies. The significance of the wide bandwidth optical comb lies in the fact that it has substantially reduced this challenge.

A. Measurement of absolute optical frequency

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 lack of systematic errors. For example, recent tests have shown that the repetition rate of a mode-locked laser equals the mode spacing to within the measurement uncertainty of 10^{-16} (Udem *et al.*, 1999a, 1999b). The uniformity of the comb mode spacing has also been verified to a level below 10^{-17} (Udem *et al.*, 1999a, 1999b), even after spectral broadening in fiber. Comparison between two separate fs comb systems, both linked to a common reference source (microwave or optical), allows one to examine the intrinsic accuracy of a fs comb-based frequency measurement system, currently at a level of a few parts in 10^{16} with no measurable systematic effects (Diddams *et al.*, 2002).

Another confirmation of fs comb accuracy was provided by comparison of the measured frequency of a He-Ne laser frequency stabilized on a molecular iodine transition. Using an elaborate scheme of transfer standards and cross checks, we were able to reliably compare the absolute frequency of the laser measured with a fs comb in JILA and that measured by a traditional harmonic optical frequency synthesis chain located at NRC, Canada (Ye, Yoon, *et al.*, 2000). The difference between the two measurements is below 1.6×10^{-12}. While the accuracy of this test has fewer digits, it is comforting to find such an agreement in a direct comparison of the two synthesis methods and the two national labs. With a broad reference line and rather large shifts with operating parameters, the He-Ne/I_2 system is not suitable to provide an accuracy check below the level of 10^{-12}.

As mentioned in Sec. II, one simple use of a fs comb is to span a frequency difference between two transitions while only stabilizing $f_{\rm rep}$. In practice such a basic arrangement places a heavy burden on the electronic measurement system and limits the attainable measurement precision and accuracy. A typical approach is to stabilize the fs comb directly to a known microwave standard or an optical frequency standard (see Sec. III.B). Experimental observation has clearly confirmed that the actual limitation in precision with fs comb-based measurements is the quality of the radio frequency reference sources (Ye, Hall, and Diddams, 2000). For example, commercial Cs clocks have a stability $\sim5\times10^{-12}/\tau^{1/2}$ and can be calibrated to an accuracy $\sim1\times10^{-14}$.

There has been an explosion of absolute frequency measurements using fs comb methods in the last two years. Not surprisingly, the most accurate results come from optical standards that are based on transitions with extraordinary quality factors. One of the most precise measurements is the determination of the Hydrogen 1S-2S transition frequency at the Max-Planck Institute for Quantum Optics (MPQ) in Garching (Niering *et al.*, 2000). The MPQ team took advantage of the high stability of a transportable Cs fountain clock as the reference for their fs comb. The result was quoted at the 1.8×10^{-14} accuracy level. Some national labs have both trapped ion teams and primary frequency standard teams that form a powerful collaboration. After investing many years of effort on the Hg^+ trap (Young *et al.*, 1999), a team at the National Institute of Standards and Technology (NIST) in Boulder can now determine the frequency of an electric quadrupole transition of Hg^+ ion to $<1\times10^{-14}$ (Udem *et al.*, 2001). Yb^+ was measured with similar accuracy at the Physikalisch-

[2]See Evenson *et al.* (1973); Pollock *et al.* (1983), Clairon *et al.* (1985), Weiss *et al.* (1988), Acef *et al.* (1993); Schnatz *et al.* (1996), Bernard *et al.* (1999) for examples of results using synthesis chains.

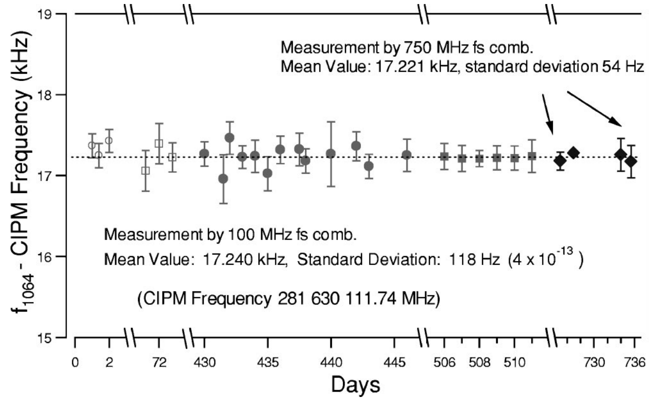

FIG. 9. Long-term frequency measurement record of the frequency doubled Nd:YAG laser stabilized on the a_{10} hyperfine component of the $R(56)$ 32-0 transition of $^{127}I_2$ at 532 nm. The agreement between the two optical comb systems is excellent. (Reproduced from Jones *et al.*, 2002.)

Technische Bundesanstalt (PTB) in Braunschweig (Stenger, Tamm, *et al.*, 2001). Excellent results are also obtained for frequency determination of a spin-forbidden intercombination line in cold Ca atoms at both NIST and PTB (Stenger, Binnewies, *et al.*, 2001; Udem *et al.*, 2001). 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 (Karshenboim, 2001).

Another direction is to explore the everyman's frequency measurement system where one can consider tradeoffs such as a ~ tenfold accuracy loss for a $\sim 10^3$ scale reduction for the apparatus. Femtosecond combs based on low power mode-locked lasers (such as diode lasers) and supercontinuum generation fibers may offer portable versions of a frequency measurement device. 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 10–1000 s (Ye *et al.*, 1999). We have measured the absolute frequency of such a system over the past three years (summarized in Fig. 9) (Jones *et al.*, 2002). At present the long-term self-reproducibility is limited to about 3×10^{-13}. Better stability and reproducibility are expected from an optimized iodine spectrometer (Cheng *et al.*, 2002; Ishibashi *et al.*, 2002).

Cell-based optical standards have played an essential role in length metrology (Quinn, 1999). For different national laboratories to establish a common base for the length standard, an assortment of wavelength-reference lasers realized separately at each laboratory need to be regularly intercompared. The traditional practice has been to hold regular conventions, organized by the Bureau International Poids et Mesures (BIPM), where stabilized lasers from different national labs were gathered at the same physical location and directly compared. A more economic and precise approach to carry out this task can now be accomplished by local calibrations of the length standard lasers with GPS systems to an accuracy $\sim1\times10^{-14}$. Another rather pleasant outcome of the absolute frequency calibration process is the associated capability of unambiguous tests of the manufacturing process of reference cells and their long-term variations (Jones *et al.*, 2002).

B. Optical atomic clock

Measurement of optical frequencies in terms of radio frequency standards has become a rather straightforward task based on the development of fs combs. However, as the measurement precision is pushed to an ever-higher level, the stability limitation imposed by available radio frequency standards used for fs comb stabilization becomes an important issue. Instead of running the fs comb from microwave frequencies up to optical frequencies, it appears to be advantageous to run the comb in the other direction. In other words, the fs comb is actually stabilized by an optical frequency standard and produces stable clock signals in the radio frequency domain, leading to a so-called optical atomic clock (Diddams *et al.*, 2001; Ye *et al.*, 2001). Of course there are highly stable rf signal sources available, such as the hydrogen maser or the 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. Stepping down the stability level by a couple orders of magnitude, portable optical frequency standards offering compact, simple, and less expensive system configurations have also shown competitive performance with (in)stability near 1×10^{-14} at 1–10-s averaging time. Another strong argument favoring optical stabilization of a 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. Granted, the notorious phase noise accumulation in a traditional frequency multiplication chain does not apply to an optical comb based on a mode-locked laser since mode locking requires phase locking of all participating comb components.

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 kHz to the sub-Hz domain are available by selection of an atomic transition with a natural decay time τ_0 in the 100-s to 1-s domain. In principle, one could obtain $\sim 1/(2\tau_0)$ interactions per second with approximately only twofold 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 a 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 ~1/(SNR) in 1 s. Taking the resonance linewidth into account leads to a frequency uncertainty $\delta\nu(\text{at 1 s})\sim\Delta\nu/(\text{SNR}) = (2/N\tau_0)^{1/2}$. In case that the interrogation time, T_R, is shorter than the actual lifetime of the transition under study (we assume the Ramsey separated-field method), the fractional frequency (in)stability is then given by

$$\sigma_y(\tau) = \frac{\delta\nu}{\nu_0} = \frac{1}{\omega_0\sqrt{NT_R\tau}}. \qquad (6)$$

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. As an example, the 400-s lifetime of the Ca $^1S_0$-$^3P_1$ transition at 657 nm ideally could provide a <100-mHz laser stability at 1 s, using 10^6 atoms. Noting that the transition frequency is 456 THz, this leads to 1.6×10^{-16} projected fractional stability! Stability within a factor of 25 of this ideal value has recently been reported (Oates *et al.*, 2000). Of course, so many atoms in a small volume may bring some problems, such as collisional shifts, etc. Another approach is to use just a single ion, such as Hg^+ with $\tau_0 \sim 90$ ms, which leads to about 1.0-Hz possible stability at 1 s. The transition frequency in this case is 1064 THz ($\lambda \sim 282$ nm), so one can expect $\sim 1\times10^{-15}$ stability at 1 s (Rafac *et al.*, 2000).

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 straightforward 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.

To realize an optical atomic clock, an optical comb needs to be stabilized to a pre-selected optical frequency source at a precision level that exceeds the optical standard itself. As discussed in Sec. II, a comb system has two degrees of freedom, $f_{\rm rep}$ and f_0, and both need to be controlled. We need to have two experimental observables to recover the information relating to $f_{\rm rep}$ and f_0. This step can be accomplished with two different but fundamentally related approaches. The first approach [scheme A, Fig. 10(a)] uses the self-referencing technique to recover f_0, which can then be stabilized with respect to either $f_{\rm rep}$ or an auxiliary stable rf source. It is worth noting that stabilization of f_0 to a few mHz is more than adequate, as it yields fractional frequency noise of $<10^{-17}$ for an optical carrier. A heterodyne beat between one of the comb components and the cw laser ($f_{\rm cw}$), which acts as the optical frequency standard, yields information about fluctuations in $f_{\rm rep}$. After appropriate processing, this error signal is used to stabilize the phase of $f_{\rm rep}$ coherently to $f_{\rm cw}$, thereby producing a clock signal output in the rf domain derived from $f_{\rm cw}$.

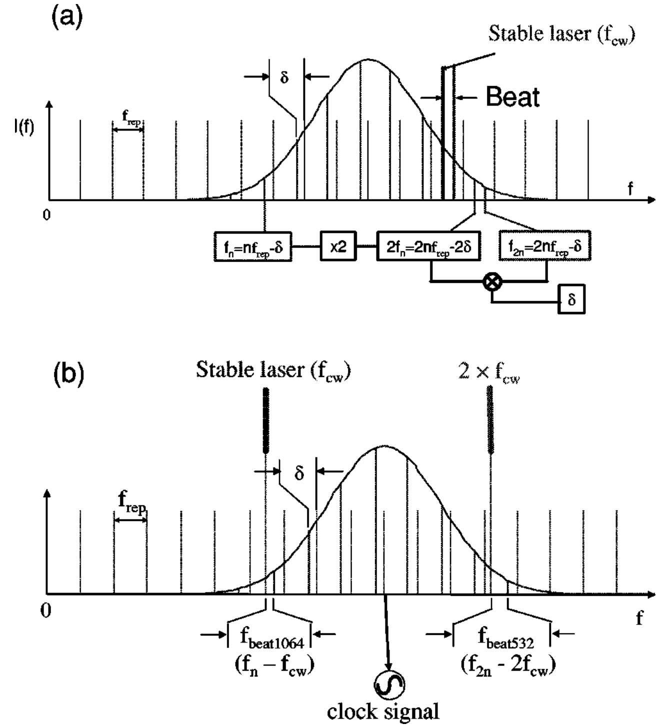

FIG. 10. Two equivalent schemes for the implementation of optical clocks using an octave-spanning optical frequency comb. In the self-referencing approach, shown in (a), frequency doubling and comparison are accomplished with the comb itself. In the second approach, shown in (b), the fundamental and the second harmonic of the cw optical frequency standard are used for error signal derivation and comb stabilization.

The second approach [scheme B, Fig. 10(b)] uses two beat signals between a cw stabilized laser ($f_{\rm cw}$) and its second harmonic ($2f_{\rm cw}$) and two respective comb components in the closest neighborhoods of these two cw frequencies. One immediately sees that we are taking the same advantage of the octave bandwidth of a fs comb, except that in the second case it is the cw laser that is frequency doubled 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 $f_{\rm rep}$, respectively.

One of the JILA optical frequency standards is a diode-pumped solid state Nd:YAG laser ($f_{\rm cw}$) with its second harmonic ($2f_{\rm cw}$) locked on a hyperfine component of an iodine transition $[R(56)32\text{-}0,a_{10}]$ near 532 nm; this system offers an (in)stability of 4×10^{-14} at 1 s (Hall *et al.*, 1999; Ye *et al.*, 1999). To implement the clockwork using scheme A, we employed an acousto-optic modulator (AOM) in the beam path of the pump laser before entering the fs laser cavity to stabilize f_0 with a bandwidth exceeding 50 kHz. Experimentally we have achieved not only a mHz stability for the value of f_0 when measured by a frequency counter, but also a mHz scale linewidth for f_0. The beat signal between a comb component near 1064 nm and the stabilized Nd:YAG laser can be recovered with a SNR of 30–40 dB at 100-kHz bandwidth. This beat signal can be phase locked to a rf signal derived either in a self-consistent manner from f_{rep}, which we want to stabilize, or from a moderately stable rf signal source. For example, an rf source at 100 MHz with a stability of 10^{-11} will add only mHz noise to the optical frequency standard, leading to a stability degradation no worse than a few parts in 10^{18}. When referenced to the optical standards, the instability of the beat signal phase lock has also been demonstrated to be $<10^{-17}$ at 1-s averaging time.

The same Nd:YAG laser has also been used to derive clock signals with scheme B. In this case, both beat signals, $f_{\mathrm{beat}\,1064}=nf_{\mathrm{rep}}+f_0-f_{\mathrm{cw}}$ and $f_{\mathrm{beat}\,532}=2nf_{\mathrm{rep}}+f_0-2f_{\mathrm{cw}}$, are regenerated electronically with rf tracking oscillators/filters, then mixed to produce control signals related to f_{rep} and f_0, namely, $s_{\mathrm{ctrl}\,1}=f_{\mathrm{beat}\,532}-f_{\mathrm{beat}\,1064}=nf_{\mathrm{rep}}-f_{\mathrm{cw}}$ and $s_{\mathrm{ctrl}\,2}=f_{\mathrm{beat}\,532}-2f_{\mathrm{beat}\,1064}=-f_0$. The frequency/phase variations arising in both f_{rep} and f_0 are therefore directly manifested in the two control variables $s_{\mathrm{ctrl}\,1}$ and $s_{\mathrm{ctrl}\,2}$ and are linked to the optical frequency standard f_{cw}. These two signals drive two servo transducers to close the feedback loops. For clock signal generation, essentially we need to use only the information of $s_{\mathrm{ctrl}\,1}$ to control l_c and thus stabilize f_{rep} with respect to f_{cw}. The variable f_0 can be actually left free running since it has been effectively taken out of the control equation. In practice, we use l_c to control the phase of $s_{\mathrm{ctrl}\,1}$ to that of another stable oscillator in the rf domain, leading to definitive phase coherence between f_{rep} and f_{cw}. For a 100-MHz fs laser with an intracavity prism pair, we have achieved a tracking (in)stability between f_{rep} and f_{cw} at a level of $<10^{-15}$.

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 optical standard. To characterize the system, the optical clock signal is compared against other well-established microwave/rf frequency standards. The international time standard, the Cs clock, should certainly be one of the references; however, the short-term stability of a commercial Cs atomic clock is only $\sim 5\times 10^{-12}$ at 1 s. For improved short-term characterization of the fs comb clock, we also use a NIST-maintained hydrogen maser signal. The comparison typically involves a heterodyne beat experiment between the two signal sources with a frequency counter recording the resultant beat frequency fluctuations over a period of time. The time record of the beat frequency can be used to determine the Allan variance (Allan, 1966) that displays the frequency noise vs its characteristic time scales. For a set of N frequency measurements f_n, each with a sampling time τ, the corresponding Allan variance is defined as

$$\sigma_y^2(\tau)=\frac{1}{2(N-1)}\sum_{n=1}^{N-1}(f_{n+1}-f_n)^2. \tag{7}$$

Allan variance analysis is a powerful technique developed to separate and isolate processes based on their time scales. It is useful to identify various time scales at which fluctuations occur. Figure 11 summarizes the comparison results of the 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. The data for the optical standard itself were obtained from heterodyne experiments between two similar laser stabilization systems, although further tests revealed stability degradation when an optical signal is transferred to the rf domain (Diddams *et al.*, 2001).

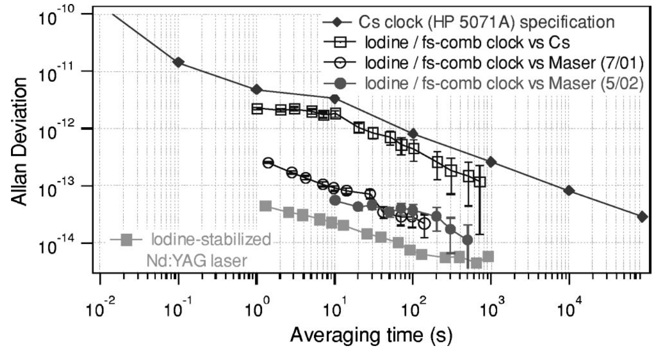

FIG. 11. Allan deviation obtained from heterodyne comparison between a hydrogen maser and an optical clock is displayed by curves with circles, with the open circles obtained from the 100-MHz comb system and the filled circles from the 750-MHz comb system. For comparison, we also display the Allan deviation associated with the commercial Cs atomic clock (worst case specification) (curves with diamonds) and the Allan deviation of the iodine stabilized laser (curves with squares). (Reproduced with modifications from Ye *et al.*, 2001.)

However, we still face some technical challenges on the road to making an optical clock a reliable scientific device. Further developments in technological areas necessary for an advanced optical atomic clock include: (i) highly accurate cold-atom optical frequency standards and portable high stability optical frequency standard, (ii) development of ultrastable optical local oscillators suitable for the most demanding spectroscopic tasks, (iii) stabilization and control of wide-bandwidth optical combs, including exploration of novel generation and detection techniques and approaches to reduce noise, (iv) reliable, stable, and compact ultrafast laser technology for practical implementation of optical clocks, (v) high stability and high accuracy rf output from an optical clock at a level that does not degrade its stability, (vi) development and utilization of femtosecond combs for intercomparison of optical frequency standards and the cesium primary standard, (vii) development of frequency and time transfer methods over extended fiber optic links that can support the next generation of atomic frequency standards.

C. Optical frequency synthesizer

As will be discussed in Sec. IV, a future goal in ultrafast technology is to demonstrate arbitrary pulse synthesis in the time domain, including the capability of phase-coherent stitching of distinct optical bandwidths. Complementary to that time-domain capability, it is desirable 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. Such a capability would allow great simplification in precision laser spectroscopy.

With the development of an optical 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 1-Hz level. 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. 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 reference to the stabilized optical comb. A self-adaptive search algorithm first tunes the LD to a specified wavelength region with the aid of a wavelength measurement device (100-MHz resolution). A heterodyne beat signal between the LD's frequency and that of a corresponding comb line is then detected and processed. For fine tuning, a rf source provides a tunable frequency offset for the optical beat. Once the LD frequency tuning exceeds one comb spacing, we reset the radio frequency offset back to the original value to start the process over again. The LD frequency can thus be tuned smoothly in an inchworm manner along the comb structure. Experimentally we verify this tuning process by using the modes of an independent optical cavity to monitor the LD frequency. When the entire optical comb is stabilized to an ultrastable optical frequency standard, such stability can be faithfully transferred to another cw laser located hundreds of THz away.

We have demonstrated two fundamental aspects of an optical frequency synthesizer; namely, continuous, precise frequency tuning and arbitrary frequency setting on demand (Jost *et al.*, 2002). Figure 12 shows the self-adaptive random search of any targeted comb position by the single frequency cw laser. Part (a) shows the LD's coarse tuning under the guidance of a wavelength meter. Once the LD is tuned to within the desired spectral range (limited by the resolution of the wavelength meter), the fs comb takes over the guiding and sets precisely the LD's frequency to any specified position according to the cavity transmission, as shown in Fig. 12(b). Clearly the same procedure can be applied when a natural atomic or molecular resonance is the target. The total search time is on the order of a minute. Finally, in Fig. 12(c) we show that controlled, rapid switching of the LD frequency can be implemented, within a reasonable frequency gap. The typical settling time is 2.5 ms, limited by a mechanical resonance in the piezotransducer used for the LD servo.

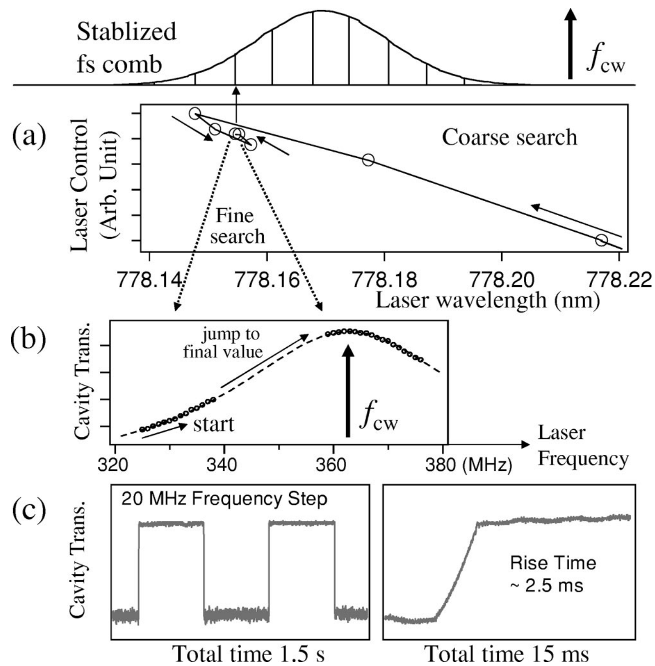

FIG. 12. Random search of and stabilization to the targeted comb position by a single frequency cw laser, with an initial coarse guiding given by (a) a wavelength meter, followed by (b) a controlled frequency seeking. The entire searching procedure within a 0.5-nm spectral region finishes on a 1-min time scale. Also shown is (c) controlled fast switching of the laser diode frequency. (Reproduced from Jost *et al.*, 2002.)

IV. OTHER APPLICATIONS OF FEMTOSECOND COMBS

Prior to the development of femtosecond comb technology, mode-locked lasers were used almost exclusively for time-domain experiments. Although the femtosecond comb technology has primarily impacted the frequency domain applications described in Sec. III, it is having an 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 (Yoon *et al.*, 2000). As discussed below, these applications put stringent requirements on the carrier-envelope phase coherence. Once long phase coherence times are 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.,

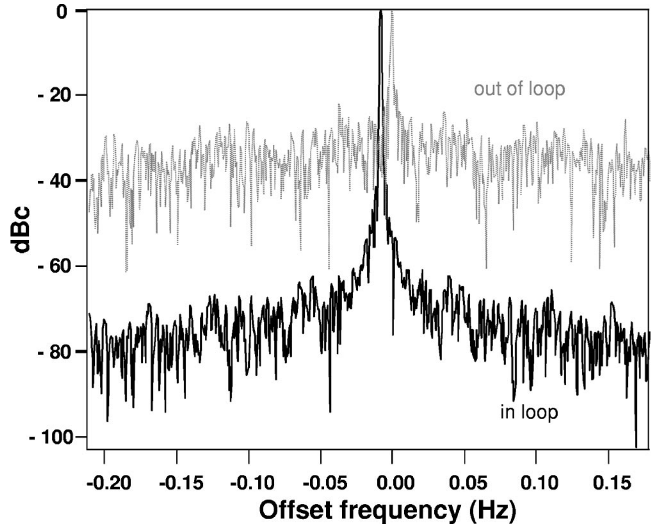

FIG. 13. Spectra of f_0 beat signal (offset for clarity) as measured with a fast Fourier transform spectrum analyzer. The in-loop measurement (solid) was taken with a resolution bandwidth of 0.976 mHz, while the out-of-loop measurement (dashed) was taken at 0.488 mHz. The linewidths are resolution limited.

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

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. For coherent pulse synthesis, it is obviously important because we need to maintain carrier-envelope coherence between the two lasers. For experiments sensitive to ϕ_{ce}, it is difficult to determine how ϕ_{ce} affects the outcome if ϕ_{ce} is varying wildly during the measurement.

Although the cross-correlation measurements presented in Sec. II.E demonstrate some degree of phase coherence, they are actually very 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 of f_0 is a much more sensitive approach 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

$$\Delta\phi_{ce}^{RMS}|_{\tau_{obs}} = \sqrt{\int_{1/2\pi\tau_{obs}}^{\infty} \frac{1}{f^2} s_\nu^{f_0}(f)\,df}$$

for an observation time τ_{obs} (Ye *et al.*, 2002). Figure 13 shows a measurement of the spectrum of f_0 (Fortier *et al.*, 2002). Both an in-loop measurement (the measured f_0 is used in the servo loop to stabilize the laser) and an out-of-loop measurement, using a second f-to-$2f$ interferometer, are shown. The linewidth is limited by the measurement time of 2048 s. The standard definition for coherence time is the τ_{obs} at which 1 radian of phase fluctuations have accumulated. These results give a measurement limited coherence time that is greater than 163 s.

We would like to emphasize that carrier-envelope phase coherence is not the same as optical coherence. A process that shifts the position of the pulse without changing the ϕ_{ce} of the pulse destroys the optical coherence, but does not affect the carrier-envelope phase coherence.

B. Timing synchronization of mode-locked lasers

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:sapphire 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 (Leitenstorfer *et al.*, 1995; Wei *et al.*, 2001), we have chosen a flexible all-electronic approach for active stabilization of repetition rates to achieve synchronization (Ma *et al.*, 2001).

We place two Kerr-lens mode-locked Ti:sapphire lasers (Asaki *et al.*, 1993) in a mechanically and thermally stable environment for the synchronization experiment. To synchronize the two lasers, we use two phase locked loops (PLL's) working at different timing resolutions. One PLL compares and locks the fundamental repetition frequencies (100 MHz) of the lasers. An rf phase shifter between the two 100-MHz signals can be used to control the (coarse) timing offset between the two pulse trains with a full dynamic range of 10 ns. The second, high-resolution PLL compares the phase of high-order harmonics of the two repetition frequencies, for example, the 140th harmonic at 14 GHz. This second loop provides enhanced phase stability of the repetition frequency when it supplements and then replaces the first PLL. A transition of control from the first PLL to the second PLL can cause a jump in the timing offset by at most 35.7 ps ($\frac{1}{2}$ of one 14-GHz cycle), whereas the adjustable range of the 14-GHz phase shifter is 167 ps. The servo action on the slave laser is carried out by a combination of transducers, including a fast-piezoelectric-actuated small mirror, a regular mirror mounted on a slow piezo with a large dynamic range (~180 Hz–100 MHz), and an acousto-optic modulator placed in Laser II's pump beam to help with fast noise. The unity gain frequency of the servo loop is about 200 kHz and the loop employs three integrator stages in the low-frequency region (Shelton *et al.*, 2002).

To characterize the timing jitter, we focus the two pulse trains so that they cross in a thin BBO crystal cut for type-I sum frequency generation (SFG). The

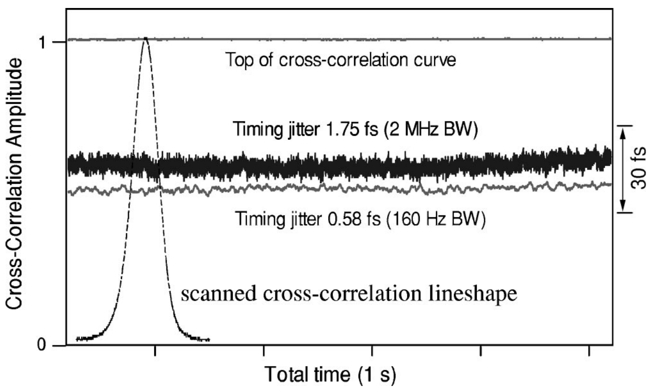

FIG. 14. Timing jitter between two synchronized fs lasers. The dotted curve is the cross-correlation signal of the two lasers when the relative pulse timing is scanned across the overlap region. Timing jitter determined from the intensity fluctuations of the SFG intensity is shown over a period of 1 s, using two different low-pass bandwidths. (Reproduced from Shelton, Ma, *et al.*, 2002.)

crossed-beam geometry produces an intensity SFG cross-correlation signal. The Gaussian cross correlation (obtained when the two lasers are free-running) is about 161 fs FWHM. (No extra-cavity dispersion compensation is used, the pulses would be 20 fs in the transform limit.) The top trace in Fig. 14 shows that the SFG signal, recorded with a 2-MHz bandwidth, has no detectable intensity fluctuations when the two laser pulses are maximally overlapped (at the top of the cross-correlation peak). The two middle traces are recorded with 2-MHz and 160-Hz bandwidths, respectively, when the timing offset between the two pulse trains is adjusted to yield the half maximum intensity level of the SFG signal. The slope of the cross-correlation signal near half maximum can be used to determine the relative timing jitter between the two lasers from the corresponding intensity fluctuations. Timing jitter is calculated from the intensity noise using the slope of the correlation peak, with the scale of the jitter indicated on the vertical axis of Fig. 14. The rms timing noise is thus determined to be 1.75 fs at a 2-MHz bandwidth and 0.58 fs at a 160-Hz bandwidth. For detection bandwidths above 2 MHz, the observed jitter does not increase. We have recorded such stable performance over several seconds. The synchronization lock can be maintained for several hours. However, the intensity stability of the SFG signal is found to strongly correlate with the temperature variations in the microwave cables. A careful study of the servo error signal inside the feedback loop reveals that a major limitation to the present performance is actually due to the intrinsic noise of the 14-GHz phase detector, a double balanced mixer. Integration of the intrinsic noise level in the mixer produces the lowest rms timing jitter limit for the synchronization loop and it determines the rms timing jitter limit for the 1–160-Hz frequency range to be $\sqrt{2.6\times10^{-3}\ \mathrm{fs}^2/\mathrm{Hz}\times160\ \mathrm{Hz}}$ ≈ 0.64 fs. To achieve even better performance, one can use either a single highly stable cw laser or a common stable optical resonator to control a high-order harmonic of the repetition frequency, well into the THz or tens and hundreds of THz frequency range. Timing noise at 0.1 fs should be achievable.

The ability to synchronize a passively mode-locked laser to an external reference, or to a second laser, has many applications. Previous work in electronic synchronization of two mode-locked Ti:sapphire lasers has demonstrated timing jitter of at best a few hundred fs. Therefore the present level of synchronization would make it possible to take full advantage of this time resolution for applications such as high power sum- and difference-frequency mixing (Kaindl *et al.*, 2000), novel pulse generation and shaping (Shelton *et al.*, 2001), new generations of laser/accelerator-based light sources, or experiments requiring synchronized laser light and x rays or electron beams from synchrotrons (Schoenlein *et al.*, 1996). 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, sub-ps pulse generation in the IR (Foreman *et al.*, 2003) and coherent anti-Stokes Raman scattering (CARS) microscopy with two tightly synchronized ps lasers (Potma *et al.*, 2002). 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.

C. Phase lock between two mode-locked lasers

Phase locking of separate femtosecond (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 (Shelton, Foreman, *et al.*, 2002). As illustrated in Fig. 15(a), after synchronization matches the repetition rates ($f_{\mathrm{rep1}}=f_{\mathrm{rep2}}$), 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. 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 $(\Delta\phi_{\mathrm{ce1}}-\Delta\phi_{\mathrm{ce2}})=0$, leading to two pulse trains that have nearly identical phase evolution.

The two independent mode-locked Ti:sapphire lasers each operate at a 100-MHz repetition rate, with one centered at 760 nm and the other at 810 nm. The bandwidth of each laser corresponds to a sub-20-fs transform limited pulse. When synchronized, the heterodyne beat between the two combs can be recovered with a 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 the lasers are syn-

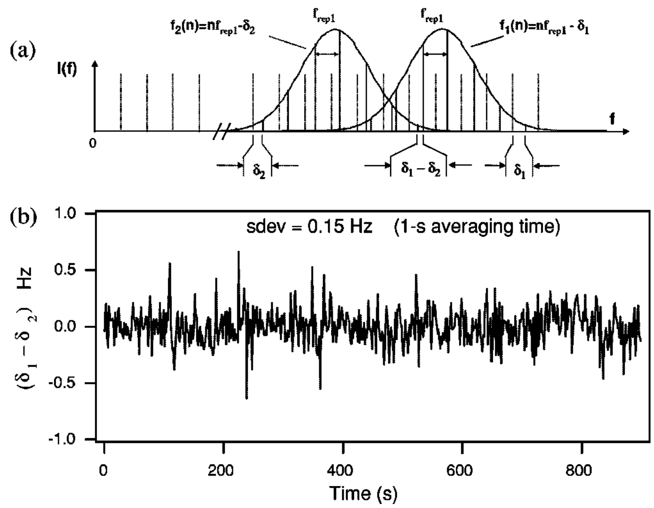

FIG. 15. Phase locking of two synchronized mode-locked lasers. (a) The principle behind the synchronization and phase locking of two independent femtosecond lasers. After tight synchronization, one needs to control $f_{01}-f_{02}=0$ in order to merge the two comb families into one. (b) Record of the beat frequency between the two laser carrier signals when they are synchronized and phase locked. The beat frequency under the locked condition shows a standard deviation of 0.15 Hz at 1-s averaging time.

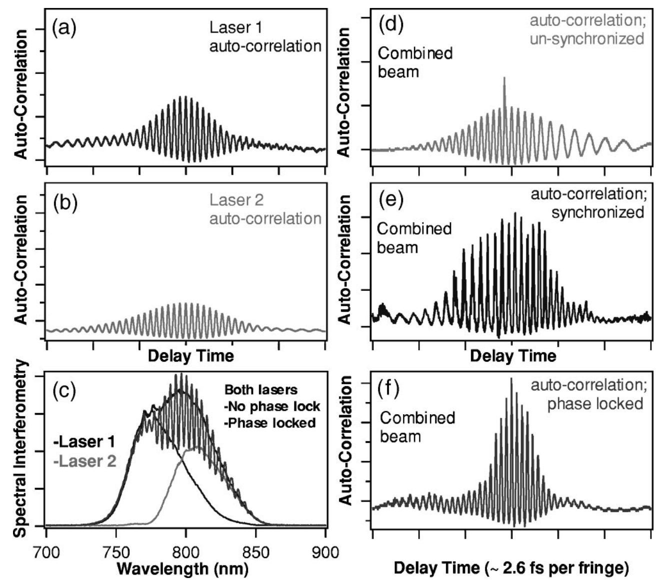

FIG. 16. Second-order autocorrelation measurement of the combined pulse. Second-order autocorrelation data of the two individual lasers are shown in (a) and (b). In (c), the spectral interferometry data indicate that the spectra of the two lasers are separated, with interference fringes clearly present in the overlap region of the two spectra. The pulse trains from the two lasers are combined and sent to a collinear autocorrelator. Data in (d) represent the case when the two lasers are not synchronized. The two fields are in phase only briefly, resulting in a random sharp spike in the autocorrelation curve. Data in (e) are obtained when the two lasers are synchronized but not phase locked. The increase of the autocorrelation amplitude is about 2.7 times over the previous unsynchronized case. When the two lasers are both synchronized and phase locked, the second-order autocorrelation data in (f) show that the pulse width is narrowed and the pulse amplitude is increased by >20%. Notice the large difference in noise between the phase locked and unlocked autocorrelation curves. (Reproduced from Shelton *et al.*, 2001.)

chronized. By stabilizing δf_0 to a mean value of 0 Hz, the carrier-envelope phase slip per pulse of one laser will accurately match the second laser. Locking of δf_0 to 0 Hz is implemented using an acousto-optic modulator (AOM). One laser beam passes through the AOM and is offset by the drive frequency of the AOM. This avoids the need to process the beat signal in the troublesome frequency range around zero frequency. The beat is then phase locked to the drive frequency of the AOM, effectively removing the AOM frequency. When unlocked, the carrier beat frequency has a standard deviation of a few MHz with 1-s averaging time. Figure 15(b) shows the recorded beat frequency signal under the phase locked condition. With an averaging time of 1 s, the standard deviation of the beat signal is 0.15 Hz.

The established phase coherence between the two femtosecond lasers is also revealed via a direct time-domain analysis. For example, we have employed spectral interferometry analysis of the joint spectra of the two pulses to produce interference fringes that correspond to phase coherence between the two pulse trains persisting over the measurement time period. A cross-correlation measurement between the two pulse trains also manifests the phase coherence in the display of persistent fringe patterns.

A more powerful and straightforward demonstration of the coherently synthesized aspect of the combined pulse is through a second order autocorrelation measurement of the combined pulse. For this measurement, the two pulse trains were maximally overlapped in the time domain before the autocorrelator. The autocorrelation curves of each individual laser are shown [Figs. 16(a) and (b), respectively]. The spectra of the lasers are centered around 760 and 810 nm. Under phase lock, the spectral interferometry measurement displays clear interference fringes within the overlapping spectra [Fig. 16(c)]. Fringe visibility is reduced when the measurement time is increased, due to the increased phase noise between the two lasers. An interesting autocorrelation measurement is obtained when the two lasers are not even synchronized [Fig. 16(d)]. Basically we obtain an autocorrelation of a single laser pulse, with a sharp spike appearing in the data at a random position. The spike appears because, at that particular instant, the 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 ns. When the two lasers are synchronized but not phase locked, the resulting autocorrelation measurement indicates increased signal amplitude compared to the unsynchronized case, typically by a factor of 2.7 [Fig. 16(e)]. However, as expected, this signal displays considerable random phase noise within the autocorrelation interference fringes. When the two femtosecond lasers are phase locked, the autocorrelation reveals a clean pulse that is often shorter in apparent duration and larger in amplitude [Fig. 16(f)]. Note that we did not attempt to recompress the light pulses outside the laser cavities to very short duration, and the pulses are dispersively broadened to 50–70 fs. The width of the central fringe pattern in an interferometric autocorrelation is more characteristic of the overall bandwidth of the pulse than of the pulse duration, and can result in a trace that appears deceptively short. However, the data clearly show the difference between a coherently phase-locked pulse train [Fig. 16(f)] and one with a complex and random frequency substructure [Fig. 16(e)]. Averaging of hundreds of the autocorrelation scans also consistently shows an amplitude enhancement by more than 20% when the phase is locked. We have therefore demonstrated 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 (Shelton *et al.*, 2001).

D. Extreme nonlinear optics

The expression extreme nonlinear optics refers to experiments where the optical pulses are so intense that the electric field of the pulse is relevant, not the intensity profile, as is the case in the perturbative regime (Brabec and Krausz, 2000). In this regime, the electric field can be so strong that it distorts the potential energy well for an electron bound to an atom to such a large extent that ionization occurs via tunneling. Such tunneling ionization (known as above threshold ionization when more than the minimum number of photons needed for ionization are absorbed) typically displays a threshold with respect to the electric field of the pulse. This causes it to depend on the phase of pulse for sufficiently short pulses if the threshold is close to the maximum field in the pulse. Recently, measurements of the photoelectron yield in opposite directions have yielded evidence for a phase dependence in above threshold ionization (Paulus *et al.*, 2001).

A pulse of extreme ultraviolet to soft x-ray light can be generated if the ionized electron slams back into the ion, which occurs for a linearly polarized incident pulse (Spielmann *et al.*, 1997; Durfee *et al.*, 1999; Brabec and Krausz, 2000). Since an x-ray pulse is emitted for each half cycle of the incident field, the spectrum of the emission consists of lines that are harmonics of the incident laser field, and thus this process is known as high harmonic generation. Since each x-ray pulse is thought to have a duration of approximately 100 as (Christov *et al.*, 1997), this has received much attention as a means of generating attosecond pulses (Drescher *et al.*, 2001). Just as for above threshold ionization, this process is thought to be extremely sensitive to the carrier-envelope phase, with fluctuations in the early experiments being attributed to the fact that the carrier-envelope phase was uncontrolled (Spielmann *et al.*, 1997; Durfee *et al.*, 1999). Controlling the carrier-envelope phase will yield advances in attosecond pulse generation, although significant work is still required to preserve the phase through an amplifier.

E. Coherent control

Control of molecular reactions is a central goal of chemistry. The development of the laser led to predictions that light fields could be used to control reaction pathways; for a recent review see, for example, Rabitz *et al.* (2000) and Shapiro and Brumer (2000). Many of the techniques are sensitive to the phase of the applied fields and thus dubbed coherent control. To date, only the relative phase between two laser fields, or the relative internal phase of a femtosecond pulse (i.e., its chirp) has been demonstrated to have physical impacts. Some new schemes explore interference between pathways involving n photons and m photons. When n and m have opposite parity a dependence on ϕ_{ce} will occur for excitation by a 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 (Yin *et al.*, 1992) and to control electrical currents in bulk semiconductors (Hache *et al.*, 1997). In both cases, there is a connection between spatial direction and the relative phase, which is ϕ_{ce}. Calculations show that a detectable signal injection current should occur in semiconductors for a transform limited pulse of 6 fs or shorter (Sipe, 2001). These coherent control phenomena present an interesting means of measuring ϕ_{ce} as well as a potential application of phase controlled pulses.

V. SUMMARY

The recent developments in femtosecond comb generators have enabled breakthroughs in optical frequency metrology, optical frequency synthesis, and optical atomic clocks. Femtosecond combs have been built or are being built for these applications around the world. Although not large on an absolute scale, the number is a large multiple of the number of frequency multiplication chains ever built. Indeed, here in Boulder, between NIST and JILA, there are more femtosecond-comb-based frequency chains than were operating in the world prior to 1999! Thus we are confident that precision absolute optical frequency metrology and synthesis is becoming a common laboratory tool.

The time-domain applications engendered by femtosecond combs are just being realized. They also promise very exciting results in the near future.

ACKNOWLEDGMENTS

The authors would like to acknowledge the many contributors to the work on femtosecond combs at JILA. Special acknowldgement goes to Jan Hall for building the foundation of precision laser stabilization at JILA over the last 40 years. Others include S. A. Diddams, S.

Foreman, T. M. Fortier, T. W. Hänsch, L. Hollberg, K. Holman, D. J. Jones, R. J. Jones, H. C. Kapteyn, L.-S. Ma, M. M. Murnane, R. Shelton, and T. Udem. Our research was funded by NIST, NSF, ONR, NASA, DARPA, and the Research Corporation. Both authors are staff members of the Quantum Physics Division of NIST, Boulder.

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