Control of coherent light and its broad applications

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A remarkable synergy has been formed between precision optical frequency metrology and ultrafast laser science. This has resulted in control of the frequency spectrum produced by mode-locked lasers, which consists of a regular "comb" of sharp lines. Such a controlled mode-locked laser is a "femtosecond optical frequency comb generator." For a sufficiently broad comb, it is straightforward to determine the absolute frequencies of all of the comb lines. This ability has revolutionized optical frequency metrology and synthesis, and it has also led to recent demonstrations of atomic clocks based on optical frequency transitions. In addition, the comb technology is having a strong impact on time-domain applications, including control of the carrier-envelope phase, precision timing synchronization, and synthesis of a single pulse from independent lasers.

1 Introduction and overview

Precision phase control of femtosecond laser-based optical frequency combs has produced remarkable and unexpected progress in the areas of precision spectroscopy, optical frequency metrology, and ultrafast optics [1-9]. A phase stabilized frequency comb spanning an entire optical octave (> 300 THz) has been established, leading to a single step, phase coherent connection between the optical and radio-frequency spectral domains. Optical frequencies can thus be measured in a straightforward manner by referencing to millions of marks on a frequency "ruler" that are stable at the Hz level while covering most part of the visible spectrum. The precision comb can also serve as an accurate gearbox to transfer an optical oscillation signal down to the microwave/rf domain, thereby establishing an optical atomic clock [8,9]. We will present one of the systems based on an optical transition of iodine molecules, providing an rf clock signal with a frequency stability that is comparable to that of the optical standard, and superior to almost all conventional rf sources. To realize a high-power CW optical frequency synthesizer, a separate widely tunable single-frequency cw laser has been employed to randomly access the stabilized optical comb and lock to any desired comb component [10]. Carrier-envelope phase stabilization of few cycle optical pulses has recently been realized [3,4]. We can now achieve stabilization of the pulse-to-pulse and the "absolute" carrier-envelope phase to a level of milli-radians and tens of milli-radians, respectively [11]. This level of phase coherence is maintained over several minutes, paving the groundwork for synthesizing electric fields with known amplitude and phase at optical frequencies. Working with two independent femtosecond (fs) lasers operating at different wavelength regions, we synchronize the relative timing between the two pulse trains at the fs level [12], and also phase lock the two carrier frequencies, thus establishing phase coherence between the two lasers [13]. By coherently stitching the optical bandwidth together, a "synthesized" pulse has been generated. The simultaneous control of timing jitter (repetition rate) and carrier-envelope phase can also be used to phase coherently superpose a collection of successive pulses from a mode-locked laser. For example, by stabilizing the two degrees of freedom of a pulse train to an optical cavity acting as a coherent delay, constructive interference of sequential pulses will be built up until a cavity dump is enabled to switch out the "amplified" pulse [14]. Furthermore, the synchronization techniques we developed for pulse synthesis have also made a strong impact in the field of nonlinear-optics based spectroscopy and imaging, showing significant improvements in experimental sensitivity and spatial resolutions [15]. In short, we now appear to have all the experimental tools required for *complete* control over coherent light, including the ability to generate pulses with arbitrary shape and precisely controlled frequency and phase, and to synthesize coherent light from multiple sources.

2 Time domain consequence of frequency domain control

It is useful to first explore the connections between the disciplines of the ultra-stable and the ultra-fast. The ultra-stable field is typified by high-resolution spectroscopy and high-precision measurements carried out by continuous wave (CW) lasers that can be best described by their near delta-function frequency spectra. In contrast, the field of ultra-fast phenomena encompasses the study of sub-ps events utilizing laser pulses that approach the limit of time domain delta-functions. In fact, state-of-the-art laser sources from these two fields now share nearly the same delta-function "figure of merit" with frequency and temporal widths on the order of a few parts in 10^{15} Hz and seconds, respectively. The connection between the ultra-stable and the ultra-fast 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. Although "mode-locking" is a frequency domain concept, mode-locked lasers and their applications are typically discussed in the time domain. Recent development of frequency domain treatment of mode-locked lasers has allowed the extensive tools of frequency domain laser stabilization to be employed for ultrafast optics, with dramatic results. A key concept in this discussion is the carrier-envelope phase, ϕ_0, which is the phase shift between the peak of the pulse envelope and the closest peak of the underlying carrier wave, as illustrated in Fig. 1(a). In any dispersive material, the difference between group and phase velocities will cause ϕ_0 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) $f_{rep}= 1/\tau_{r.t.} = v_g/l_c$, 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 $\Delta\phi$) between the "carrier" phase and the envelope peak for each of the successive pulses emitted by the laser, with $\Delta\phi = \left(1/v_g - 1/v_p\right) l_c \omega_c \bmod (2\pi)$ as shown in Fig. 1(a). Here ω_c is the carrier frequency. In the frequency domain, $\Delta\phi$ yields an offset of the comb from the exact harmonics of f_{rep} by the amount $f_{ceo} = \Delta\phi\, f_{rep} /2\pi$. Hence each optical comb frequency is effectively given by $f_n = nf_{rep} + f_{ceo}$, where n is the integer (~ 1 million) harmonic number of a particular given optical comb line.

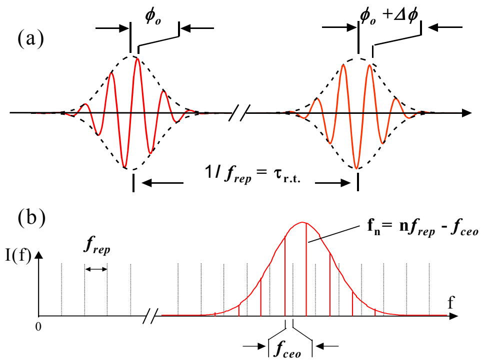

Fig. 1 Time and frequency domain representations of the comb.

Frequency-domain control of both f_{rep} and f_{ceo} makes it possible to establish a high precision fs-laser based optical comb. The bandwidth of a Ti:S based fs-laser comb already covers more than 100 nm, and it can be further extended to cover an entire optical octave using nonlinear optical effects such as that in microstructure fibers. With an octave-spanning spectrum it becomes possible to measure optical frequencies in a single step with a direct reference to the present realization of the SI unit of time, namely the Cs standard. This process can be implemented in two equivalent ways. The first approach is to use the comb to measure the frequency gap between a CW laser (f_{cw}) and its second harmonic ($2\,f_{cw}$). In other words, the octave interval $2\,f_{cw}$ - $f_{cw} = f_{cw}$, is directly expressed in quantities of f_{rep} and f_{ceo} [2,5]. The second approach, as shown in Fig. 2, is to frequency double the infrared portion of the comb spectrum and to heterodyne it with the existing visible portion of the spectrum [3]. The resulting beat frequency is the comb offset f_{ceo} . When the laser is controlled in such a fashion that both f_{rep} and f_{ceo} are linked to the Cs standard, one produces an entire array of absolute, precisely known optical frequencies.

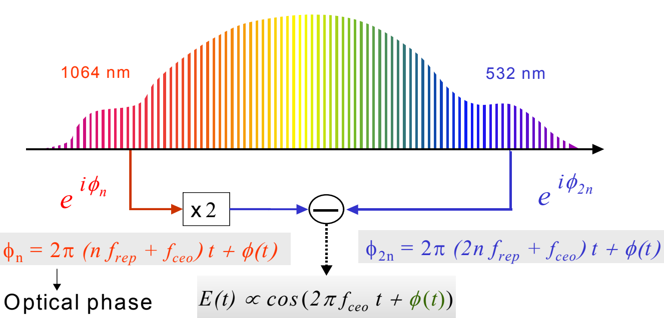

Fig. 2 Self-referenced phase sensitive heterodyne detection of $|2f_n - f_{2n}| = f_{ceo}$.

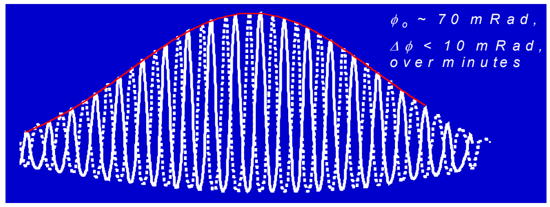

Fig. 3 Coherence of the carrier-envelope phase, as shown in cross-correlation.

For time-domain experiments, control of the frequency ratio f_{ceo}/f_{rep} establishes the evolution of $\Delta\phi$. However, establishing a long-term coherence of the carrier-envelope phase requires precise phase control of f_{ceo} (See Fig. 2). 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_{ceo} is limited by the same measurement time window [11]. Furthermore, the absolute phase of each pulse, which is also influenced by the propagation media outside of the laser cavity, can also be controlled to within 100 mRad over minutes. These results are summarized in Fig. 3. The frequency-domain-based laser control techniques 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 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.

3 Coherent pulse synthesis and applications

Our motivation of working with separate ultrafast lasers stems from the desire of creating 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 ultra-broad 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 (see Fig. 4). A flexible all-electronic approach [12] for active stabilization of repetition rates to achieve synchronization has recently enabled us to achieve sub-fs timing jitter at a 160 Hz bandwidth over an observation period of several seconds. This synchronization lock can be maintained for several hours. A major limitation to the present performance is due to the intrinsic noise of the microwave mixer used for detection of high order harmonics of f_{rep} . Timing noise below 0.1 fs should be achievable if one uses either a single highly stable cw laser or a stable optical resonator as a reference, extending the harmonic order of (f_{rep}) well into and beyond the THz frequency range.

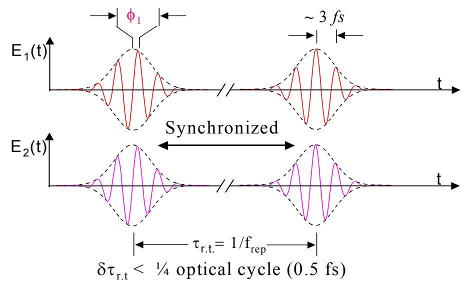

Fig. 4 Synchronization between two independent mode-locked lasers.

Phase locking of separate femtosecond (fs) lasers requires a step beyond tight synchronization. One would need effective detection and stabilization of the phase difference between the two optical carrier waves underlying the pulse envelopes. As shown in Fig. 5(a), after synchronization matches the repetition rates ($f_{rep\ 1} = f_{rep\ 2}$), 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 spectrally overlapping comb components of the two lasers. Such heterodyne detection yields information related to the difference in the offset frequencies of the two lasers, $\delta f_{\mathrm{ceo}} = f_{ceo1} - f_{ceo2}$, which can then be controlled. By phase locking δf_{ceo} to a mean zero value, we effectively demand that $(\Delta\phi_1 - \Delta\phi_2) = 0$, leading to two pulse trains that have nearly identical phase evolution. Fig. 5(b) shows the recorded δf_{ceo} 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_{ceo} is a few MHz at 1 s averaging.

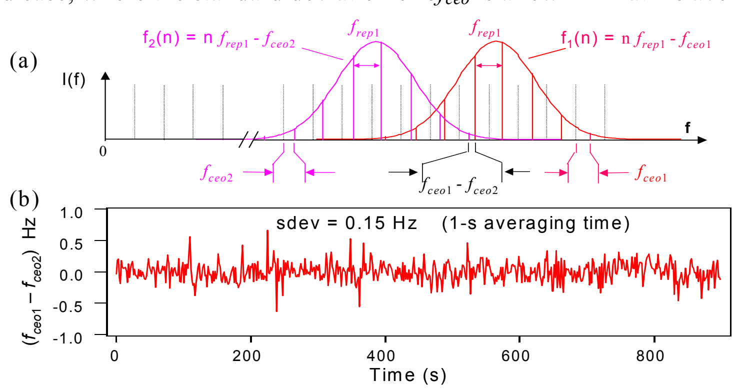

Fig. 5 (a) Schematic and (b) result of phase locking of two carrier frequencies.

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 [13]. For this measurement, the two pulse trains were maximally overlapped in the time domain before the auto-correlator. The auto-correlation curves of each individual laser are shown in Fig. 6(a) and 6(b), respectively. Fig. 6(c) shows the auto-correlation measurement when the two lasers are not synchronized. Basically we obtain an auto-correlation 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 femtosecond lasers are phase locked, the auto-correlation reveals a clean pulse that is shorter in apparent duration and larger in amplitude (Fig. 6(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.

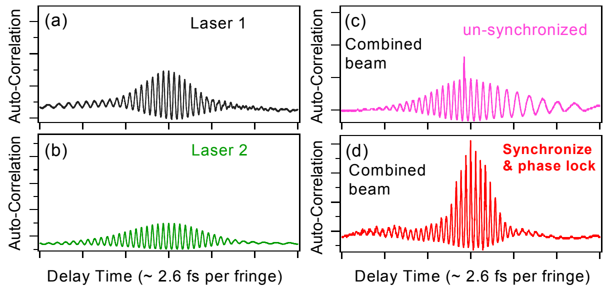

Fig. 6 Autocorrelation measurement of a coherently synthesized pulse.

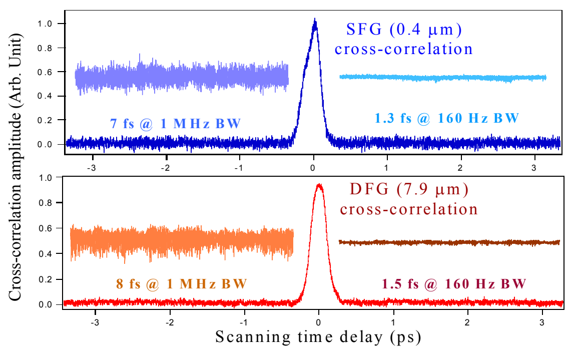

Fig. 7 Simultaneous sum and difference frequency generations from two stabilized femtosecond lasers.

The capability of stabilizing and working with two mode-locked lasers opens many applications. It may be particularly important in the generation of tunable femtosecond sources in other previously unreachable spectral regions. Fig. 7 shows the cross-correlation measurement of the two stabilized mode-locked Ti:s lasers using both sum (SFG) and difference frequency generation (DFG). The DFG signal produced by a GaSe crystal can be tuned from 6 micron and onto any longer wavelength regions with a high repetition rate (the same as the original laser's) and a reasonable average power (tens of microwatts). Arbitrary amplitude waveform generation and rapid wavelength switching in these nonlinear signals are simple to implement. Another important application is in the field of nonlinear-optics based spectroscopy and nanoscale imaging. For example, using two tightly synchronized ps lasers, we are able to achieve significant improvements in experimental sensitivity and spatial resolutions for coherent anti-Stokes Raman scattering (CARS) microscopy [15], as evidenced in Fig. 8.

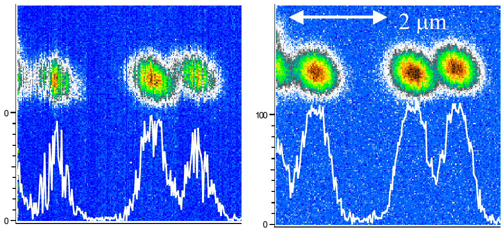

Fig. 8 CARS image of 1-μm polystyrene beads under two different synchronization conditions. The laser frequency difference matches the Raman shift of 1600 cm^{-1}.

4 Optical atomic clocks and frequency synthesizers

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. 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 [5,16,17]. 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 a ~ 10-fold accuracy loss for a ~ 10^3 scale reduction for 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 absolute frequency of such a system over the past three years (summarized in Fig. 9). At present the long-term reproducibility 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 can now be accomplished by local calibrations with GPS systems to an accuracy ~ 1×10^{-14}.

Experimental observation has clearly confirmed that the actual limitation to precision in fs comb-based measurements is the quality of the radio frequency reference sources [5,6]. For example, commercial Cs clocks have a stability ~ $5 \times 10^{-12}/ \tau^{1/2}$ and can be calibrated to an accuracy ~ 1×10^{-14}. 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 have the fs comb actually stabilized by an optical frequency standard and produces stable clock signals in the

RF domain, leading to a so-called “optical atomic clock” [8,9]. Recent experimental demonstrations support the concept that, in the future, the most stable and accurate frequency standards will be based on optical transitions.

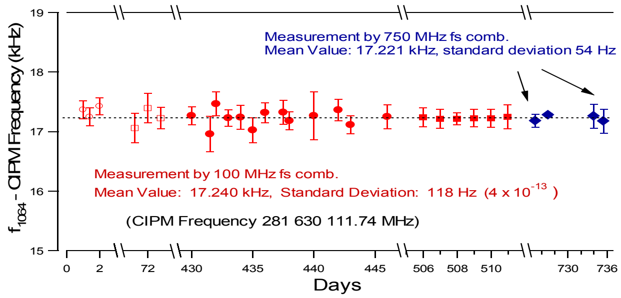

Fig. 9 Long-term measurement record of the absolute frequency of an I_2 transition.

The advantage of an optical frequency standard is apparent if we examine the frequency stability of an atomic clock. Resonance natural widths, Δν, 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 μs to 1 s domain. In principle, one could obtain ~ $1/(2\tau_0)$ 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*) ~ $\sqrt{N}$ 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 ~ 1/(*SNR*) in 1 s. Taking the resonance linewidth into account leads to a frequency uncertainty $\delta\nu$(at 1 s) ~ $\Delta\nu/(SNR)$ = $(2/N\tau_0)^{1/2}$. In case that the interrogation time, T_R, (we assume the Ramsey separated-field method) is shorter than the actual lifetime of the transition, the fractional frequency (in-)stability is given by $\sigma_y(\tau) = \delta\nu/\nu_0 = 1/\left(\omega_0\sqrt{NT_R\tau}\right)$. In this expression, ω_0 ($=2\pi\nu_0$) is the clock transition frequency, and τ ($\tau > T_R$) is the total averaging time. Clearly, higher stability is attained if we can increase ω_0, from a microwave to an optical frequency.

To realize an optical atomic clock, an optical comb ideally should be stabilized to a pre-selected optical frequency source at a precision level that exceeds the optical standard itself. A comb system has two degrees of freedom, f_{rep} and f_{ceo} . We need to have two experimental observables to recover the information relating to f_{rep} and f_{ceo} . 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_{ceo}, which can then be stabilized with respect to either f_{rep} or an auxiliary stable RF source. It is worth noting that stabilization of f_{ceo} 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_{cw}), which acts as the optical frequency standard, will then yield information about fluctuations in f_{rep}. After appropriate processing, this error signal is used to stabilize the phase of f_{rep} coherently to f_{cw}, thereby producing a clock signal output in the RF domain derived from f_{cw}. The second approach [scheme B, Fig. 10(b)] 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 we are taking the same advantage of the octave bandwidth of a fs comb, except in the second case the cw laser 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_{ceo} and f_{rep}.

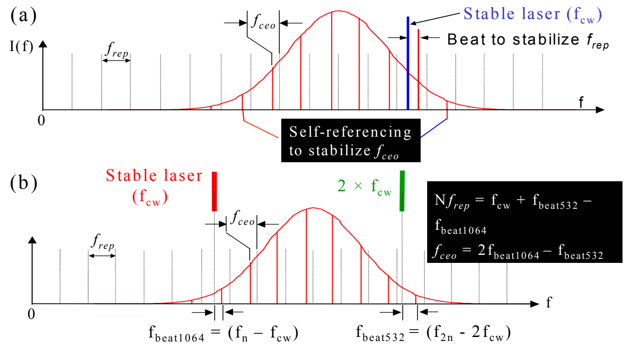

Fig. 10 Two schemes to extract clock signal from an optical source.

Our optical frequency standard is a 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 system offers an (in-)stability of 4×10^{-14} at 1 s. To implement the clockwork using scheme A, we first stabilize f_{ceo} by self-referencing. We achieve not only a mHz stability for the mean value of f_{ceo} , but also a mHz scale linewidth for f_{ceo} . The beat signal between a comb component near 1064 nm and the stabilized Nd:YAG laser is recovered with a *SNR* of ~ 40 dB at 100 kHz bandwidth. This beat signal is phase locked to an RF signal derived either in a self-consistent manner from f_{rep}, which we want to stabilize, or from an adequately stable RF signal source. Even with a stability of 10^{-11}, an RF source at 100 MHz 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. To derive clock signals with scheme B, both beat signals, $f_{beat1064} = nf_{rep} + f_{ceo} - f_{cw}$ and $f_{beat532} = 2nf_{rep} + f_{ceo} - 2f_{cw}$, are regenerated electronically with an rf tracking-oscillator/filter, then mixed to produce control signals related to f_{rep} and f_{ceo} , namely $s_{ctrl1} = f_{beat532} - f_{beat1064} = nf_{rep} - f_{cw}$ and $s_{ctrl2} = f_{beat532} - 2f_{beat1064} = -f_{ceo}$. The frequency/phase variations arising in both f_{rep} and f_{ceo} are therefore directly manifested in the two control variables s_{ctrl1} and s_{ctrl2} and are linked to the optical frequency standard f_{cw} . For clock signal generation, we need to use only s_{ctrl1} to control l_c , and thus stabilize f_{rep} with respect to f_{cw} , while leaving the variable f_{ceo} free-running. Tracking (in-)stability between f_{rep} and f_{cw} is now at a level of < 10^{-15}.

To characterize the system, the optical clock signal is compared against other well-established microwave/RF frequency standards, including the primary time standard, the Cs clock. However, the short-term stability of a commercial Cs atomic clock is only ~ 5×10^{-12} at 1 s. For improved short-term characterization of the fs comb clock, we also use a NIST-maintained hydrogen maser signal. The time record of the heterodyne beat frequency between two signal sources is used to determine the Allan deviation that displays the frequency noise vs. its characteristic time scales. Fig. 11 summarizes the comparison results of the optical clock against the Cs and H-maser references. The Allan deviation reaches 1×10^{-14} at 500 s, slightly worse than the maser itself, most likely due to phase noise in the fiber link between NIST and JILA. The data for the optical standard itself were obtained from heterodyne experiments between two similar laser stabilization systems.

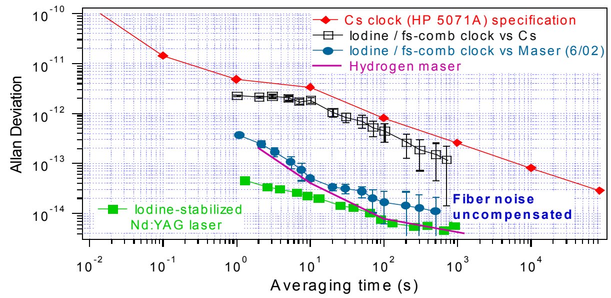

Fig. 11 Comparison of an optical atomic clock against other rf sources.

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. 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. Of course we still face some technical challenges in making an optical clock a reliable scientific device. Further necessary developments include: Highly accurate and stable optical frequency standards; Reliable, stable, and compact ultrafast laser technology with long-term control of wide-bandwidth optical combs; High stability and accuracy optical-RF connection; Development of frequency and time transfer methods that can support the next generation of atomic frequency standards.

An optical frequency grid with stable lines over a large optical bandwidth is useful for a number of applications. However, often times we desire a single-frequency optical-“delta”-function that that would allow access in the frequency domain to any optical spectral feature of interest with a well-defined optical carrier wave. Realization of such an optical frequency synthesizer (analogous to its radio-frequency counterpart) would allow great simplification in precision laser spectroscopy. One can foresee an array of diode lasers, each covering a successive tuning range of ~ 10 – 20 nanometers and emitting some reasonably useful power, that would collectively cover most part of the visible spectrum. Each diode laser frequency will be controlled by the stabilized optical comb, and therefore related to the absolute time/frequency standard, while the setting of the optical frequency will be done through computer control to any desired value. For the first step, we have implemented a computer based control system that allows a widely tunable diode laser to tune through a targeted spectral region at a desired frequency step while maintaining reference to the stabilized optical comb. Suitable software allows the laser to scan on an arbitrary but exact pattern anywhere within its tuning range.

We have demonstrated two fundamental aspects of an optical frequency synthesizer; namely continuous, precise frequency tuning and arbitrary frequency setting on demand. Figure 12 shows the self-adaptive random search of any targeted comb position by the single frequency CW laser. Part (a) shows the CW laser's coarse tuning under the guidance of a wavelength meter. Once the laser is tuned to within the desired spectral range, the fs comb takes over the guiding and precisely sets the CW laser frequency to any specified position according to the cavity transmission signal and stabilizes the diode laser there until further notice, as shown in Fig. 12 (b). The total search time is on the order of a minute [10].

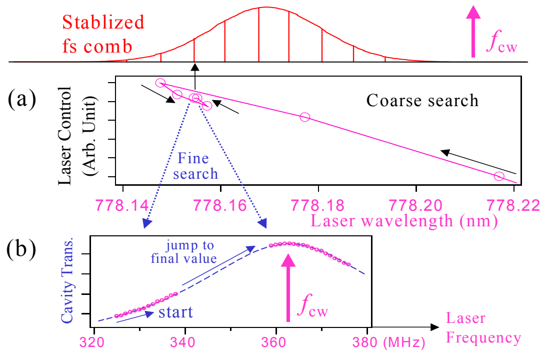

Fig. 12 Optical frequency synthesizer referenced to a stabilized fs comb.

5 Conclusions

The recent developments in femtosecond comb generators have enabled breakthroughs in optical frequency metrology, optical frequency synthesis and optical atomic clocks. 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. Thanks are due to J. Jost, R. Shelton, L.-S. Ma, and H. Kapteyn. Our research is funded by NIST, NSF, ONR, NASA, and Research Corporation.

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