# Towards the ultimate control of LIGHT

## Optical frequency metrology and the phase control of femtosecond pulses




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In just the past year there has arisen a remarkable convergence of two seemingly disparate subfields of optical physics: precision optical frequency metrology and ultrafast phenomena. The former field is typified by the variety of spectroscopic and fundamental physical measurements that can be made using a continuous wave (CW) laser best described by its near delta-function frequency spectrum. In sharp contrast, the field of ultrafast phenomena encompasses the study of sub-picosecond events using laser pulses that approach the limit of time domain delta-functions. In fact, at this point in time state-of-the-art laser sources from these two fields share nearly the same delta-function "figure of merit" with frequency and temporal widths on the order of a few parts in $10^{15}$ hertz and seconds, respectively In this article, we will present relevant details of these two subfields, then explore their connection, and finally highlight the most recent advances made possible by this surprising union.

## The ultrastable, the ultrafast and their connection

Over the past century, spectroscopic investigation has provided much information about the structure of the atoms and molecules that make up our universe. And during this time it has always been important to have tools that permit greater measurement resolution. As resolution increased, scientists were able to measure and understand the fine and hyperfine structures, relativistic effects, the broadening due to thermal motion, the Lamb shift, and even the small spectral shifts associated with the recoil that results from an emitted photon.[1,2] Of course, the introduction of frequency-stabilized lasers has had a dramatic impact on the field of spectroscopy, and it is now possible to investigate optical transitions at the Hz level (i.e. resolution approaching 1 part in $10^{15}$).[3] For tests of fundamental physics, such precision is not excessive—especially if one is searching for variations of fundamental constants, which **has (Author query: has or have?)** been predicted to occur at or below this same level.[4] When such stable laser sources are referenced to very narrow optical transitions they also serve as excellent oscillators for what is anticipated to be the next generation of all-optical atomic clocks. With the significant increase in the Q **ratio** of carrier frequency to linewidth **(Author query: ratio inside or outside parentheses?)** and the improved control and evaluation of systematic shifts, the inaccuracy of future clocks based on optical transitions is predicted to approach 1 part in $10^{18}$.[3] This is one thousand times better than the current microwave atomic clocks based on the 9 GHz cesium hyperfine transition.

Although, as just discussed, many physical systems exhibit and benefit from great stability, other physical systems change extremely rapidly—on the time scale of femtoseconds. This, for example, is the characteristic time scale for making and breaking bonds in many chemical systems, as well as the vibrational periods of solid-state lattices. By providing photons in very short packets, lasers that produce femtosecond pulses also have the distinct advantage of peak powers in the range of kilowatts to terawatts (when amplified).[5] For applications in nonlinear optics, this peak power can be used for the generation of dense plasmas and x-ray radiation. So it perhaps comes as a surprise that lasers with such apparent nonlinear ferocity might usefully combine paths with their extremely quiet and carefully controlled cousins from the realm of precision spectroscopy and metrology. In the regime when pulses consist of just a few optical cycles, we come to a situation in which for the first time the phase of the carrier field actually begins to be of importance. It is here that the powerful techniques of frequency-domain laser stabilization can be applied to a *femtosecond* laser to control the optical carrier with respect to the envelope on even a *sub-femtosecond* time scale. Such precise control of just a few oscillations of a high intensity laser field should dramatically impact the field of extreme nonlinear optics, including above-threshold ionization and high harmonic and x-ray generation with such pulses. In addition, once the femtosecond laser is stabilized, its broad spectral coverage provides an excellent tool for precision metrology and spectroscopy across intervals of many hundreds of terahertz.

The connection between the ultrafast and the ultrastable arises from the fact that femtosecond laser oscillators produce pulses in a periodic train via mode-locking. If one simply assumes that the femtosecond laser emits a pulse once per cavity round trip $\tau_{LL}$, then it is straightforward to show that the frequency domain

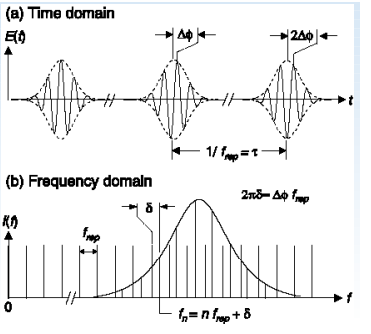



Figure 1: Time-Frequency correspondence, and the relationship between offset frequency δ and the pulse-to-pulse carrier envelope phase Δφ. (a) In the time domain, the relative phase between the carrier and the envelope of the pulse train continually increments for each successive pulse by the amount Δφ. (b) In the frequency domain, the elements of the frequency comb of a mode-locked pulse train are spaced by $f_{rep}$, and the entire comb is offset from integer multiples of $f_{rep}$ by an offset frequency $\delta=\Delta\phi\ f_{rep}/2\pi$.

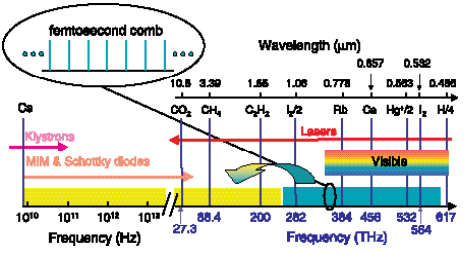



**Figure 2:** Map of the electromagnetic spectrum from the ~9.2 GHz cesium standard (Cs) to the optical. The approximate frequencies (and wavelengths) of several optical standards are identified. A significant challenge to optical frequency metrology is establishing a phase coherent connection across the 4-order of magnitude gap between Cs and the optical domain. With the new femtosecond comb technology, this problem is enormously simplified and a grid of cesium-referenced frequencies now exists across the entire visible and near infrared (green shading). **Difference frequency generation can transfer this comb to lower frequencies. (Author query: is the preceding sentence correct?)**

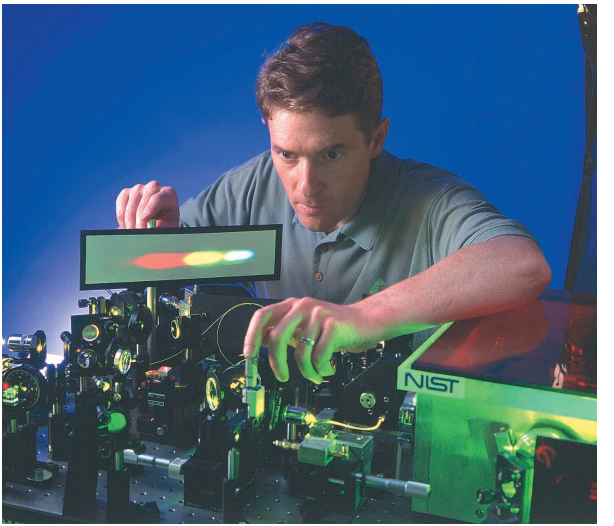
spectrum consists of a comb of discrete modes separated by the repetition frequency $f_{rep}=1/\tau_{r.t.}$. The extent of the time domain pulse train provides the frequency resolution of individual comb components, while the total extent of the frequency domain mode comb is approximately limited to the inverse of the pulse duration. However, what may not be obvious is the consequence of the carrier having a round trip cavity delay that differs from that of the envelope. This is the result of dispersion in the laser cavity, and in the time domain it is manifested as a pulse-to-pulse phase slip $\Delta\phi$ between the carrier and the envelope [see Figure 1(a)].[6,7,8] In the frequency domain, $\Delta\phi$ yields an offset of the mode comb from exact harmonics of the $f_{rep}$ by the amount $\delta=\Delta\phi\ f_{rep}/2\pi$ [see Figure 1(b)].[6,8] Based on this simple picture one can now see how the control of both $f_{rep}$ and $\delta$ makes it possible to subsequently control the pulse-to-pulse phase slip $\Delta\phi$ between the carrier and the envelope. At the same time, when both $f_{rep}$ and $\delta$ are fixed, one arrives at the unique situation of having an entire array of optical frequencies with precisely known frequencies $f_n = n\ f_{rep} + \delta$, where n is an integer. As will be described in further detail below, we are now at the point where one can produce such broad combs of oscillators across many hundreds of terahertz of bandwidth, with each comb element actively stabilized at a level approaching 1 Hz.

## Optical frequency metrology with femtosecond lasers

Any statement about the absolute frequency of a laser stabilized to an optical transition must follow from a comparison with the internationally agreed upon frequency definition based on the time unit (the second) being exactly 9,192,631,770 oscillations of the microwave field connecting cesium ground state hyperfine levels. However, a very troublesome detail in this endeavor has been **the "clockwork" that would permit the phase coherent conversion of the extremely high optical frequency (~500 THz) to a countable signal in the microwave domain [see Figure 2] (Author: is there a word missing in the preceding sentence?).** One approach has been to build frequency chains that create successive harmonics of the cesium standard, until one arrives in the optical domain.[9] This approach to optical frequency metrology is very complicated and labor intensive, and requires the scale of investment that is generally found only at national research facilities. An alternative approach essentially involves the subdivision of the octave between an optical frequency f and its second harmonic 2f. Indeed if one can directly measure the interval between f and 2f, one has accomplished the measurement of f itself. Successive bisections of the octave (yielding the frequencies 3/2f, then 5/4f, then 9/8f, and so on) with lasers and nonlinear optical elements could eventually lead to a frequency interval small enough to be directly compared to cesium.[10] Clearly, any device that would permit the measurement of the largest possible frequency gap (ideally the octave itself) would decrease the number of required bisections and simplify this scheme.

This leads to the idea of using a broadband frequency comb to measure intervals between optical frequencies. Such an experiment was first carried out in the late seventies when the frequency domain mode comb from a picosecond laser was used to demonstrate the basic potential of mode-locked lasers as an "optical ruler" in the measurement of frequency gaps in sodium.[11] This route was pursued further in the seventies and eighties,[12,13] but the attainable bandwidths were never sufficiently large to make it a widespread technique for optical frequency metrology. Although broadband femtosecond Ti:sapphire lasers have existed since the beginning of the 1990s, their use in optical frequency metrology remained un-tapped until researchers at the Max-Planck-Institut für Quantenoptik (MPQ) conclusively showed that such lasers could play a crucial role in this field. In the very first demonstration, a 20 THz wide frequency comb from a commercial femtosecond laser was used to measure the absolute frequency of the cesium $D_1$ line and thereby provide a new determination of the fine structure constant $\alpha$.[14] Furthermore, the uniformity and accuracy of the femtosecond laser comb were verified to the level of $3x10^{-17}$ and $6x10^{-16}$, respectively.[15]

As discussed above, large comb bandwidths are desirable for measuring optical frequencies. For this rea-




son, one would seek to use a femtosecond laser that produces pulses of the shortest duration. To date, the very shortest optical pulses generated directly from an oscillator consist of approximately two optical cycles and have 3dB bandwidth greater than one-half an octave (centered near 800 nm).[16] The well-known process of self-phase modulation in silica optical fiber can also act to broaden the spectrum of a femtosecond pulse. In experiments at JILA in Boulder, Colorado, such broadening permitted the comparison of two optical standards separated by 104 THz,[17] a frequency interval that would have been viewed as insurmountable even one year before. At the MPQ, similar broadening of a femtosecond frequency comb to 44 THz was used in a frequency chain that made the most precise measurement to date of any optical frequency—the 121 nm 1S-2S transition in atomic hydrogen[1]— with an uncertainty of only 46 Hz ($1.8 \times 10^{-14}$).[18]

Although such large attainable bandwidths already present a great simplification in the measurement of optical frequencies, the introduction of novel microstructure silica fibers (also called photonic crystal fibers) has provided the ultimate simplification through the generation of a complete octave of bandwidth in the visible and near infrared [see Figure 3].[19] The microstructure fiber consists of a very small (~1.5 micron diameter) silica core surrounded by a ring of air-filled holes. The unique dispersion properties of the fiber shifts the zero of the group velocity dispersion to wavelengths in the range of the Ti:sapphire laser (~800 nm). What is quite surprising about the microstructure fiber is that in spite of the high peak powers and complicated nonlinearities involved, the very stable comb structure of the femtosecond laser is transferred to the newly generated comb components via four-wave mixing.[20] The result is a vast array of discrete, evenly spaced modes that fill the entire spectrum of Figure 3.

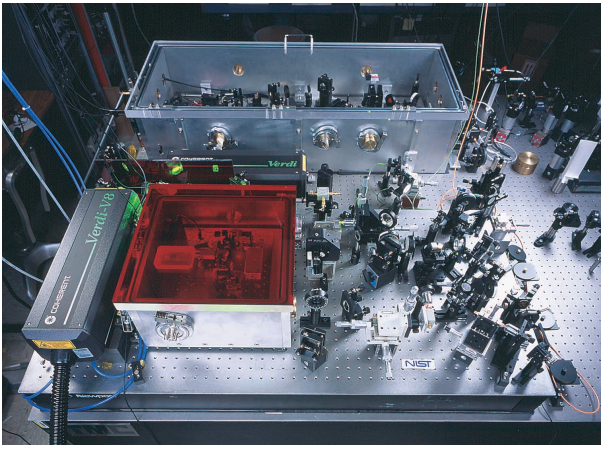
With this octave-spanning spectrum, it is now possible to measure optical frequencies in a single step with a direct reference to the cesium microwave standard. This is accomplished when the mode spacing ($f_{rep}$) of the femtosecond comb is locked with feedback control to an rf synthesizer (that itself is referenced to cesium). The octave interval between the frequency f and its second harmonic 2f can be simply expressed as $2f\text{-}f = f = n\, f_{rep}$. The first implementation of this idea was carried out with the 282 THz fundamental frequency of an iodine-

stabilized CW-YAG laser and its second harmonic.[21] A more elegant approach, which eliminates the need for any CW lasers, is to frequency double the infrared portion of the spectrum of Figure 3 and heterodyne it with the existing visible portion of the spectrum.[22,23] The resulting beat frequency is the offset δ shown in Figure 1. As already noted, when the laser is controlled (with piezo-electric transducers, for example) in such a fashion that both the radio frequencies $f_{rep}$ and δ are locked to the cesium standard, one then has an entire array of optical frequencies with precisely known frequencies $f_n = n f_{rep} + \delta$. In effect, such a femtosecond laser then operates as an optical frequency synthesizer. Very recently, researchers at the University of Karlsruhe have produced an octave-spanning frequency comb directly from a mode-locked Ti:sapphire laser (without any spectral broadening in a nonlinear fiber) and locked the offset δ using the scheme just described.[24]

It seems appropriate at this point for one to ask about the accuracy and precision of measurements made with such broadband combs. Two very recent experiments have addressed the accuracy issue. In one case, two femtosecond-based frequency chains were compared side-by-side in a measurement of the same frequency and found to agree with an uncertainty of $5x10^{-16}$.[23] In the other experiment, a femtosecond frequency chain was compared to a traditional harmonic frequency chain operating at the NRC in Ottawa, Canada. In this case, the longstanding 633 nm He-Ne optical standard was measured by both systems with an uncertainty of 220 Hz, limited by the He-Ne laser itself. [25] In every experiment done to date the limiting element in terms of the precision has been the stability of the radio frequency or microwave oscillator that controls the mode spacing $f_{rep}$. In fact, measurements indicate that once the low frequency thermal and acoustic perturbations have been removed, the stability of the repetition rate of the femtosecond laser rivals (and exceeds) that of the best crystal oscillators available. Even phase locking one of the modes of the femtosecond laser to an ultrastable CW laser does not drastically improve the stability elsewhere in the spectrum.[26] The best way to improve this situation is to remove the external microwave source completely and control both $f_{rep}$ and δ via all-optical means. This has recently been accomplished such that the stability of the iodine-stabilized CW laser is transferred to every element of the octave-spanning comb at the level of $3x10^{-13}$.[26] In this case, the system can operate as an all-optical clock that provides a stable 100 MHz radio frequency in the form of $f_{rep}$.

## Time-domain consequences of frequency-domain control

At the most basic level, the output of a mode-locked laser is an amplitude-modulated, carrier wave, shown schematically in Figure 1(a). In the case of sub-10 fs pulses, now commonly generated by mode-locked Ti:Sapphire lasers, the pulse width is only a few cycles in duration. Normally, in a femtosecond laser cavity, the carrier wave and the pulse envelope travel at different velocities, causing the relative phase between the carrier



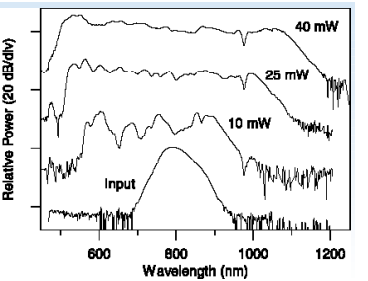



**Figure 3:** Output spectrum from the silica microstructure fiber for different amounts of coupled power. The input spectrum from the laser is also shown for reference. Beneath the envelope lies an evenly spaced comb of frequency modes.

and the envelope of the pulse train to continually increment for each successive pulse emitted by the mode-locked laser. Another problem is that fluctuations in the laser's operating parameters work to randomize the carrier-envelope phase from pulse to pulse.

Historically, the absolute phase of an optical wave's electric field has not been relevant, in that any shift in this phase had no measurable effect. However, within the context of few-cycle optical 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. Shown schematically in Figure 1(a), the value of the electric field's phase (relative to the envelope) drastically alters the optical character of a few-cycle pulse. One clear example of this condition is coherent X-ray (or high harmonic) generation. In addition to X-ray generation, other examples of extreme nonlinear optics or strong field processes where the absolute phase is critical include attosecond pulse generation and strong-field ionization.[27]

In general, the absolute carrier-envelope phase, $\phi$ of the $m^{th}$ pulse is given by $\phi_m = \phi_0 + m\,\Delta\phi$ where $\phi_0$ is unknown constant phase offset and m is an integer labeling individual pulses. As already described, the quantity $\Delta\phi$ is the pulse-to-pulse phase slip, which is related to the offset frequency and pulse repetition rate via $2\pi\delta = \Delta\phi\, f_{rep}$. By directly referencing the carrier-envelope phase evolution of the output pulse train to the pulse repetition rate itself,[22] (thus establishing a definite integral fraction of $\delta/f_{rep}$) a pulse train with constant carrier-envelope phase can be easily obtained with a pulse picker driven at the appropriate sub-harmonic of $f_{rep}$. As a result, one can now synthesize waveforms consisting of just a few optical cycles.

Due to the direct connection between frequency and time domains, both can provide a diagnostic of the carrier-envelope phase lock. After locking the carrier-envelope phase, a frequency counter can measure the variation of the offset frequency $\delta$. In recent experiments, a maximum point-to-point excursion of 5.6 Hz in $\delta$ has been observed (for a counting time of 0.1 s), which is equivalent to a maximum carrier-envelope phase slippage of 0.56 cycles/sec, thereby demonstrating long-term time stability of the phase lock.[28] Time domain measurements confirming phase control require a specially designed cross-correlator in order to measure the phase shift between successive pulses.[7,22] From the cross-correlation, the relative carrier-envelope phase can be measured directly in the time domain [see Fig 4(a)]. By changing the lock point of the offset frequency and subsequently measuring the carrier-envelope phase, a comparison can be made with the theoretical expected relation $\Delta\phi = 2\pi\delta/f_{rep}$. A plot of the experimentally determined relative phases for various offset frequencies, along with the theoretically expected linear relation is shown in Figure 4(b). The excellent agreement conclusively demonstrates the first precise waveform synthesis on a femtosecond time scale of electromagnetic fields.[29]

As noted previously, the frequency domain techniques described in this article can stabilize the carrier-envelope phase to a constant $\phi_0$, but unknown value. A similar, but refined measurement or an additional technique, such as ionization of a noble gas or tunnel ionization from a metal surface, will be necessary to measure and control $\phi_0$.

## Summary and outlook

The ability to control optical fields to within a few hertz over many hundreds of terahertz of bandwidth has collectively accelerated both the frequency metrology and ultrafast communities into new frontiers. For the first time, small-scale spectroscopy labs now have tools that permit them to generate and measure optical frequencies across the entire visible and near infrared spectrum. Furthermore, as shown in Figure 2, the difference frequency between any of the components of the synthesized frequency comb provides access to mid and far-infrared frequencies—not to mention the microwave domain. The necessary clockwork is finally available such that the high stability ($10^{-15}$ range in 1 s) of superior optical oscillators, such as those based on a single mercury ion and cold calcium atoms,[3,30] may be transferred to the countable microwave domain. As the technology progresses, it is possible that compact optical frequency standards coupled with a simple femtosecond laser system will replace current microwave standards in some applications. In ultrafast technology and applications, a similar revolutionary era has also begun. Well-controlled femtosecond pulses will permit new approaches to phase-sensitive quantum coherent control of atomic and molecular wavepackets.[31] And since independent fs lasers can now be phase-locked together, two-color coherent pump-probe experiments may be performed in chemical or solid-state systems. With amplification and external-cavity pulse envelope shapers, complete control over phase and amplitude of

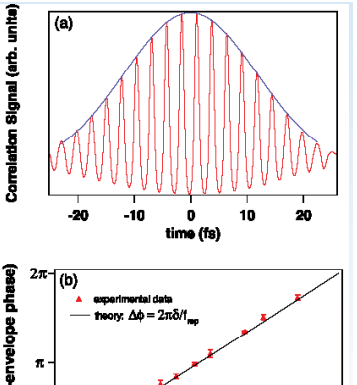



**Figure 4:** (a) Cross-correlation data between two successive pulses emitted from the phase-controlled femtosecond laser. The blue curve is a fit of the correlation envelope. For this data, the pulse-to-pulse phase shift $\Delta\phi$ is approximately $\pi$. (b) Plot of $\Delta\phi$ as a function of the normalized offset frequency $\delta$. The solid line is the expected linear relationship.

gigawatt-level optical fields on a sub-cycle (~1 fs) time scale will soon be possible. With this enabling technology, efficient extreme ultraviolet (xuv) and attosecond X-ray pulses may soon be realized, thereby opening the door for new time-resolved X-ray studies of chemical reactions and atomic structure. The preceding list is by no means complete, and the rapid realization of these new tools will most likely generate an ever-increasing range of applications in both the time and frequency domains.

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