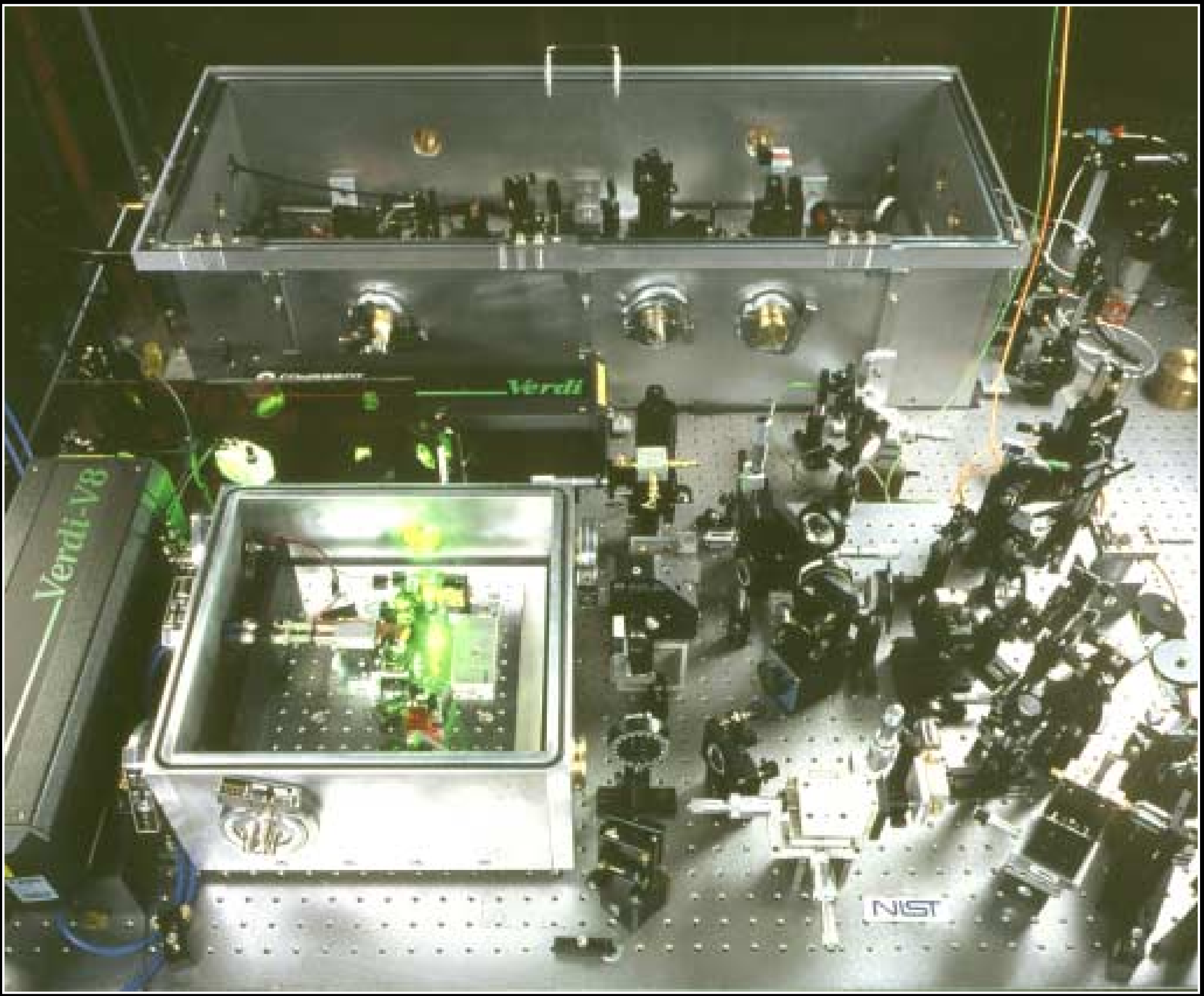
Merging the Ultrasensitive, the Ultrastable, and the Ultrafast



A New Era of Frequency Standards and Optical Frequency Measurement

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Optical frequency metrology, used today in numerous scientific and technological arenas, entails the controlled, stable generation and measurement of coherent optical waves to achieve high-precision, high-resolution measurements and process control. The use of optical frequencies in measurement science has been evolving since the earliest days of lasers. Analysis of a laser frequency at some 5 x 10^{14} Hz and a linewidth of potentially a few-milliHertz, as limited by the fundamental phase diffusion of spontaneous emission, reveals a potential dynamic range of 10^{17} resolution. This makes optical metrology one of the most potent tools yet to emerge for discovering new physics in "just the next decimal." New enabling technologies, as well as exciting developments in fundamental science, have emerged today on the heels of nearly forty years of intense research worldwide into the many diverse aspects of optical metrology. A number of ambitious, long-terms goals have recently been achieved, including:

- laser-frequency stabilization to 1 Hz and below[1];
- observation of optical transitions at a few Hz linewidth (corresponding to a quality factor Q of 1.5 x 10^{14})[2];
- realization of extremely accurate optical standards (with a target of 10^{-18} based on cold atom/ion systems);
- establishment of a direct phase-coherent link between microwave and any portion of the optical spectrum.[3, 4]

Given such developments, it seems that a reasonably robust optically based atomic clock and optical frequency synthesizer will soon be within our reach. Since frequency-based metrology is used in most modern measurement applications, in parallel we can also expect tremendous growth in related research fields.

Following a general discussion of the field of optical frequency metrology, we list some of the contributions made by the National Institute of Standards and Technology (NIST) to the exciting recent developments. This work is carried out in part at JILA, a joint institute between NIST and the University of Colorado.

Issues of optical frequency standards

A few years after the laser's invention, the international frequency standard was established based on the F = 4, $m_F = 0 \leftrightarrow F = 3$, $m_F = 0$ transition in the hyperfine structure of the ground state of the cesium 133 atom.[5] The transition frequency was defined as 9,192,631,770 Hz. The resonance Q of ~10^8 was set by the limited coherent interaction time between matter and field. Since then, a tremendous amount of effort has been invested in the extension of the coherent interaction time between matter and field and in the reduction of first- and second-order Doppler shifts. In particular, recent advances in laser cooling and trapping technology have led to a hundredfold enhancement in resolution using the atomic fountain technique. With reduced velocities, the first- and second-order Doppler effects have also been eliminated or greatly reduced. Cesium (Cs) clocks based on atomic fountains have been built by several different research groups and are now operating with reported accuracy of 3 x 10^{-15} and short-term stability of 1 x 10^{-13} at 1 second, limited by the frequency noise of the local rf crystal oscillator.[6]

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Since the invention of the laser, numerous researchers have sought to establish frequency standards in the optical domain. Their work has been fueled by the fact that the superior quality of suitable optical clock transitions can translate into enhanced accuracy, reproducibility, and stability—the three key aspects of any frequency standard. Accuracy refers to a standard's intrinsic capacity to track the frequency value of a natural transition, i.e., the value associated with an unperturbed atom or molecule at rest. Reproducibility is a measure of whether a standard can be replicated in different settings under different conditions. Stability indicates the degree to which the frequency remains constant throughout the course of the operation. A stabilized laser can achieve a fractional frequency stability

$$\frac{\delta\nu}{\nu} = \frac{1}{Q}\frac{1}{S/N}\frac{1}{\sqrt{\tau}} \quad ,$$

where *S*/*N* is the recovered signal-to-noise ratio of the resonance information, and τ is the averaging time. Clearly the limits of both resolution and sensitivity of the detected resonance need to be explored, since these quantities control the timescale within which a given level of precision in measurement can be obtained. The reward is enormous: enhancement of Q (or *S*/*N*) by a factor of ten leads to a reduction of averaging time by a factor of 100.

The drive to achieve such results led to the rapid expansion of the field of nonlinear laser spectroscopy in the 1970s and 1980s. The nonlinear nature of the quantum absorption process, while limiting the attainable *S*/*N*, also permits sub-Doppler resolutions. Specific optical techniques developed for sub-Doppler resolution include saturated absorption spectroscopy, two-photon spectroscopy, optical Ramsey fringes, optical double resonance, and quantum beats. Cold samples offer an opportunity to observe the atom's natural frequency within its rest frame. Sensitive detection techniques developed during the same period, including polarization spectroscopy, electron-shelving–quantum jump, and frequency modulation optical heterodyne spectroscopy,* have led to an absorption sensitivity of 1 part in 10^{-8} and the ability to split a MHz scale linewidth by a factor of 10^4–10^5, at an averaging time of 1 s or so. All of these advances served to lay the foundation for the sub-Hertz stabilization work upon which super-coherent optical local oscillators are based.

Active frequency-control is needed to ensure the stability and accuracy of optical local oscillators because of the strong link between laser frequency and laser parameters. In a gas laser, the gain bandwidth for stimulated emission ordinarily extends over the full Doppler width of the atomic or molecular transition line. (An exception is the case in which the gain is provided by optically pumped, velocity-selected atoms in an upper state.) Solid-state laser hosts can provide a gain bandwidth of several tens of nanometers. The optical cavity, because of its much narrower resonance width, determines the lasing frequency. This scenario is fundamentally different from the case of microwave maser standards; there, the atomic line Q prevails over the cavity Q and consequently determines the output frequency. The resonant frequencies of a laser cavity—which is fundamentally a mechanical structure—are easily perturbed by environmental

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*This work was carried out by researchers at JILA and IBM.

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conditions such as vibration, strains, pressure and temperature. For most types of lasers, frequency noise of technical origin dominates fundamental quantum fluctuations. Solitary semi-conductor laser diodes are an exception, having typical free-running linewidths on the order of tens of MHz, (limited by the fundamental phase diffusion), and so need additional linewidth-narrowing techniques.

When they are used simultaneously, quantum absorbers and an optical cavity offer an attractive laser stabilization system. A passive reference cavity brings the benefit of a linear response and a high S/N. A laser prestabilized by a cavity offers several benefits. First, a long phase coherence time reduces the need for frequent interrogations of the quantum absorber. In other words, over a short timescale the laser linewidth is narrower than the chosen atomic transition width and thus the information about the natural resonance can be recovered with an enhanced S/N while the long averaging time translates into a finer examination of the true line center. Second, the quantum absorber's resonance basically eliminates inevitable drifts associated with material standards. Frequency stability at the 10^{-16} level has been measured with a cavity-stabilized laser.[7] The use of frequency modulation for cavity/laser lock, the so-called Pound-Drever-Hall technique developed at JILA, has become standard laboratory practice.[8] The frequency-offset optical phase-locked-loop (PLL) and similar techniques can be used to assure the tenability of a cavity/laser system.

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Work in the area of stabilization at JILA and NIST has involved a broad spectrum of lasers; from early experiments with gas lasers (He-Ne, CO_2, Ar^+) to more recent work with tunable dye lasers, optically pumped solid-state lasers (Ti:Sapphire, YAG) and diode lasers. Usually one or more atomic or molecular transitions are located within the tuning range of the laser to be stabilized. Often an absorber line can be used because of its accidental overlap with a laser transition. Sometimes the same molecule (e.g., CO_2) can serve for both the lasing action and absorption. It is increasingly common to see tunable lasers stabilized on a particular frequency obtain access to another (nearby) spectrum by means of a frequency/phase offset lock technique.

Molecular transitions of rotational and/or vibrational origin have been used for laser stabilization very successfully in the infrared with molecules such as CH_4, CO_2 and OsO_4.[9] Their natural linewidths are in the range of a kilohertz or less, limited by molecular fluorescent decay at the vibration frequency. The harmonics of these fundamental transitions, termed overtone bands, extend these ro-vibrational spectra well into the visible with similar ~ kHz linewidths. Until recently the rich spectra of the molecular overtone bands were not adopted as frequency references in the visible because of their rather limited transition strengths.[10] However, excellent S/N for these weak but narrow overtone lines has recently been achieved at JILA using a sensitive absorption technique which combines frequency modulation with cavity enhancement;[11] this opens the door for realization of high quality frequency reference grids in the visible and near infrared.[12]

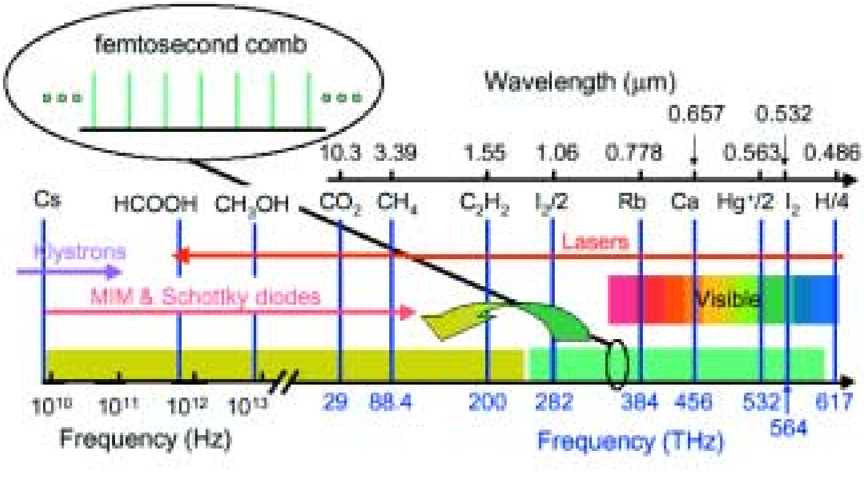

Figure 1. Frequency map of optical standards and the ultrawide bandwidth femtosecond laser comb. Shown in contrast is the electromagnetic spectrum spanning the microwave to the visible region.

Systems based on cold absorber samples potentially offer the highest quality optical frequency sources, mainly because of drastic reductions of both linewidth and systematic errors. For example, researchers at NIST recently observed a linewidth of only a few Hz on the optical transition of Hg^+.[2] Other currently active single ion systems include Sr^+,[13] Yb^+,[14] and In^+.[15] One of the early JILA concepts[16] for use of atomic fountains for optical frequency standards resulted in studies using the neutral atoms Mg, Ca, Sr, Bi, and Ag. Such systems could offer frequency standards free from virtually all of the conventional shifts and broadenings, to the level of one part in 10^{16} - 10^{18}. Yet practical considerations regarding system realization must always include cost, size and degree of complexity. Compact, low-cost systems can be competitive even though their performance may be inferior to the ideal system by a factor of ten. One such system is the Nd:YAG laser stabilized on HCCD at 1064 nm or on I_2 (after frequency doubling) at 532 nm, with a demonstrated stability level of 4 x 10^{-15} at 300 s averaging time.[17] Figure 1 lists a number of the optical frequency standards that are either established or under development.

A pressing question

One question now being asked regards the accuracy of our knowledge of the center of resonance. Collisions, electromagnetic fringe fields, residual Doppler effects, probe-field wave-front curvature, and probe power can all generate undesired linewidth broadening and center shifts. Distortion in the modulation process and other physical interactions can produce asymmetry in the line shape of the recovered signal. In frequency modulation spectroscopy, for example, residual amplitude modulation introduces unwanted frequency shifts and instability and therefore needs to be controlled.[18] Such issues will need to be addressed before researchers can be completely comfortable about accuracy issues. A more fundamental problem related to time dilation of the reference system (second-order Doppler effect) can be resolved through accurate knowledge of the sample's velocity (for example, using a velocity selective Raman process), or by reducing the velocity to a negligible level using cooling and trapping techniques.

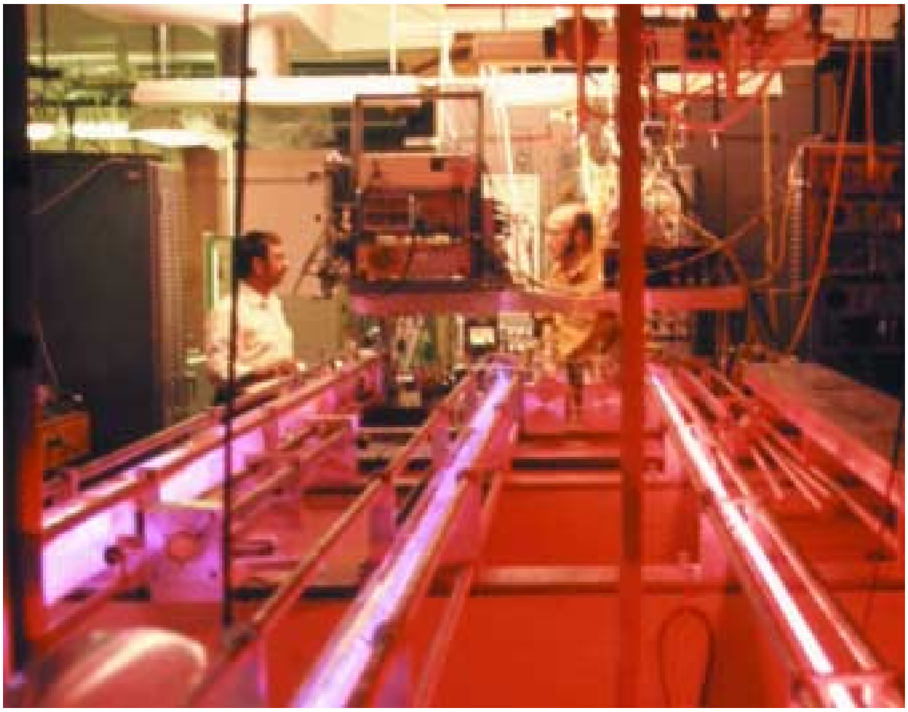

Figure 2. The first optical frequency synthesis chain at NBS (now NIST). It was used by K. M. Evenson (r) and J. S. Wells (l) and their colleagues for the measurement of the absolute frequency of the 3.39 μm He–Ne laser stabilized to a methane transition. The measurement led to accurate knowledge of the speed of light. The first absolute optical frequency measurement (iodine-stabilized He–Ne laser at 633 nm) was built on this chain.

The technology of laser frequency stabilization, refined and simplified over the years, has become an indispensable research tool in every modern laboratory where optics is used. Research on laser stabilization has been pushing the limits of measurement science. Indeed, a number of active research projects involving fundamental physical principles reap substantial benefit from the availability of stable optical sources: fields such as special relativity,[19] gravitational wave detection,[20] quantum dynamics,[21] atomic and molecular structure will need continued progress on the laser stabilization front. Recent experiments with hydrogen atoms have led to the best-reported value for the Rydberg constant and 1S-Lamb shift.[22, 23] Fundamental physical constants such as the hyperfine constant, ratio of Planck's constant to electron mass, and the electron-to-proton mass ratio are also being determined with increasing precision.[24] Thanks to the development of extremely stable phase coherent optical sources, we are on the verge of an exciting era in which picometer distance resolution can be achieved over a million kilometers in space, and a few Hertz linewidth of an ultraviolet resonance can be probed with a high *S/N*. In the time-keeping business, an optical frequency clock is expected eventually to replace the current microwave atomic clocks. In length metrology, the basic unit, the Metre, relies on stable optical frequencies. The stabilization results achieved using many different kinds of lasers invite optimism. To list just a few examples involving cw tunable lasers: milliHertz linewidth stabilization (relative to a cavity) for diode-pumped solid state lasers; dozen milliHertz linewidth for Ti:Sapphire lasers; and sub-Hertz linewidths for diode and dye lasers. Tight phase locking between different laser systems can be achieved,[25] even for diode lasers that have fast frequency noise.

Advances in optical frequency standards have resulted in the parallel development of absolute and precise frequency measurement in the visible and near-infrared spectral regions. However, until recently the measurement of optical frequencies (hundreds of terahertz) has been more primitive. This is because only a limited set of "known" frequencies has been available (see Fig. 1) and it has been hard to bridge the gap between the known frequencies and an arbitrary unknown frequency in the case in which the gap exceeds tens of GHz (or 0.01% of the optical frequency). Furthermore, even establishing a known optical frequency was difficult since the definition of the second is based on a microwave transition frequency in the cesium atom, which is ~ 50,000-fold lower than the optical frequencies of current interest (see Fig. 1).

Traditional frequency measurement takes a synthesis-by-harmonic approach. Such a synthesis chain is a complex system, involving several stages of stabilized transfer lasers, high-accuracy frequency references (in both optical and rf ranges), and nonlinear mixing elements. Integer orders of harmonics of a standard frequency are generated with the help of a nonlinear element and the output signal of a higher-frequency oscillator is phase coherently linked to one of the harmonics. Tracking and counting of the beat note or the use of a phase-locked loop (PLL) helps to preserve the phase coherence during the link. In the frequency region of microwave to mid-infrared, a harmonic mixer can perform frequency multiplication and frequency mixing/phase comparison all by itself. Cat's whisker W-Si point contact microwave diodes, metal-insulator-metal (MIM) diodes and Schottky diodes have been used extensively for this purpose. In the near-infrared to the visible (<1.5 μm), efficiencies of MIM and Schottky diodes decrease rapidly and harmonic generation is better furnished by optical nonlinear crystals. Frequency mixing (non-harmonic, up to a few tens of GHz) and phase comparison can now be directly accomplished by fast photodetectors.

Some of the phase-coherent optical frequency synthesis chains linked to the cesium primary standard are: Cs - HeNe/I_2 (633 nm);[26, 27] Cs - HeNe/CH_4 (3.39 μm);[28] Cs - HeNe/I_2 (576 nm);[29] Cs – CO_2/OsO_4 (10 μm).[30] Among these, the NIST Cs - HeNe/CH_4 was the first Cs-based frequency chain to reach the CH_4 transition at 88 THz. The scale of this endeavor is reflected in the giant lasers (HCN, CO_2,

CH_4, etc.) shown in Fig. 2. Another key NIST program involved the interferometric determination of the associated CH_4 wavelength[31] in relation to the existing wavelength standard based on krypton discharge: this work led to a definitive value for the speed of light, soon confirmed by other laboratories. Redefinition of the unit of length with adoption of c= 299,792,458 m/s became possible with the extension of the direct frequency measurements to 473 THz (HeNe/I_2 633 nm system); this was made possible 10 years later by the work of a 10-person NIST team which created a direct connection between the time and length units. More recently, thanks to improved optical frequency standards based on cold atoms (Ca)[32] and single trapped ions (Sr^+),[13] these traditional frequency chains have been able to demonstrate measurement uncertainties at the 100 Hz level.

Researchers are well aware that these frequency chains can only cover some discrete frequency marks in the optical spectrum: differences of many THz could still remain between a targeted frequency and a known reference. Over the years, a number of approaches have been tried in the search for simpler and more reliable solutions to bridge large optical frequency gaps. Some recent popular schemes include: frequency-interval bisections,[33] optical-parametric oscillation (OPO),[34] optical comb generation,[35, 36] sum-and-difference generation in the near infrared,[37] frequency division by three,[38, 39] and four-wave mixing in laser diodes.[40] In contrast to the frequency harmonic generation method described above, all of these techniques rely on the principle of difference-frequency synthesis.

Coherent bisection of optical frequency generates the arithmetic average of two laser frequencies f_1 and f_2 by phase locking the second harmonic of a third laser at frequency f_3 to the sum frequency of f_1 and f_2. These frequency-interval bisection stages can be cascaded to provide difference-frequency division by 2^n. The OPO scheme converts an input optical signal into two coherent subharmonic outputs, the signal and the idler. They are both tunable and their linewidths are replicas of the input pump except for the added quantum noise during the down conversion process. By phase locking the difference frequency between the signal and the idler to a known frequency source located in the microwave or infrared, the output frequencies can then be precisely determined. The same principle, i.e., phase-locking between the difference frequency while holding constant the sum frequency, leads to frequency measurement in the near infrared with the sum-and-difference frequency generation in optical nonlinear crystals. The sum of two frequencies in the near infrared can be matched to a visible frequency standard while the difference matches to a stable reference in the far infrared. Another important technique is optical frequency division by 3. This larger frequency ratio could simplify optical frequency chains while providing a convenient connection between visible lasers and infrared standards. An additional stage of mixing is needed to ensure the precise division ratio. Another interesting idea is to use a laser diode as both a light source and an efficient nonlinear receiver to allow a four-wave mixing process to generate phase-coherent bisection of a frequency interval of a few THz.

One of the most promising techniques in difference frequency synthesis is the generation of multi-THz optical combs by placing an rf electro-optic modulator

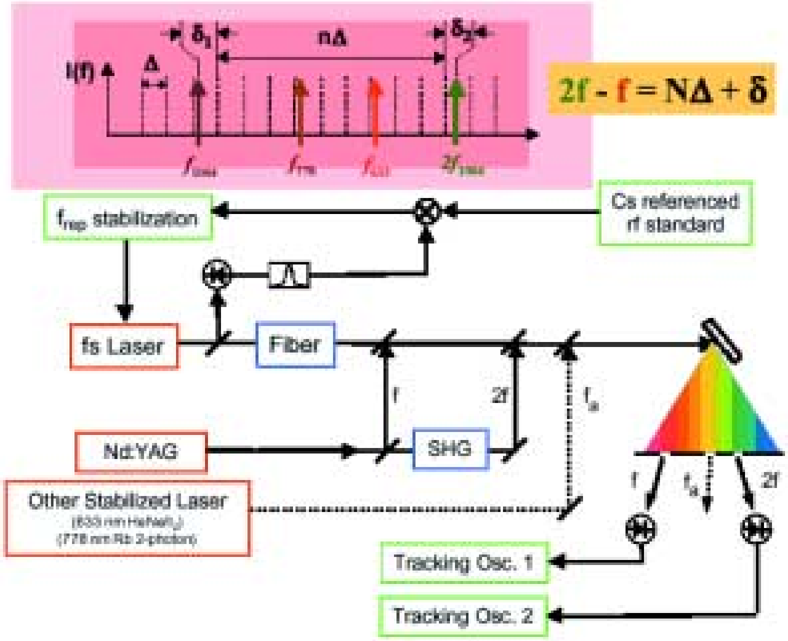

Figure 3. Femtosecond laser stabilization. Heterodyne beats between cw lasers and the corresponding comb components can be used either to count the optical frequency or stabilize the comb spectrum.

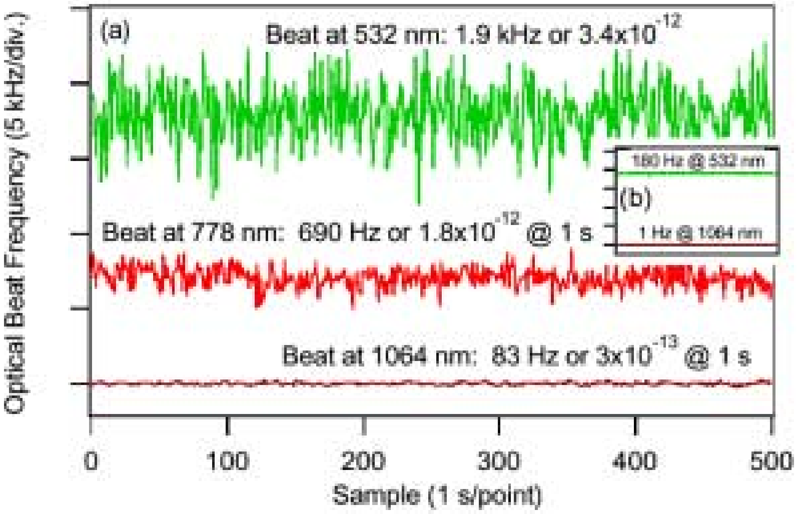

Figure 4. (a) Time record of the direct frequency measurement of cw laser frequencies with f_{rep} stabilized with respect to a microwave standard. (b) Precision phase lock between a cw laser reference and the femtosecond laser comb provides a much more stable optical comb.

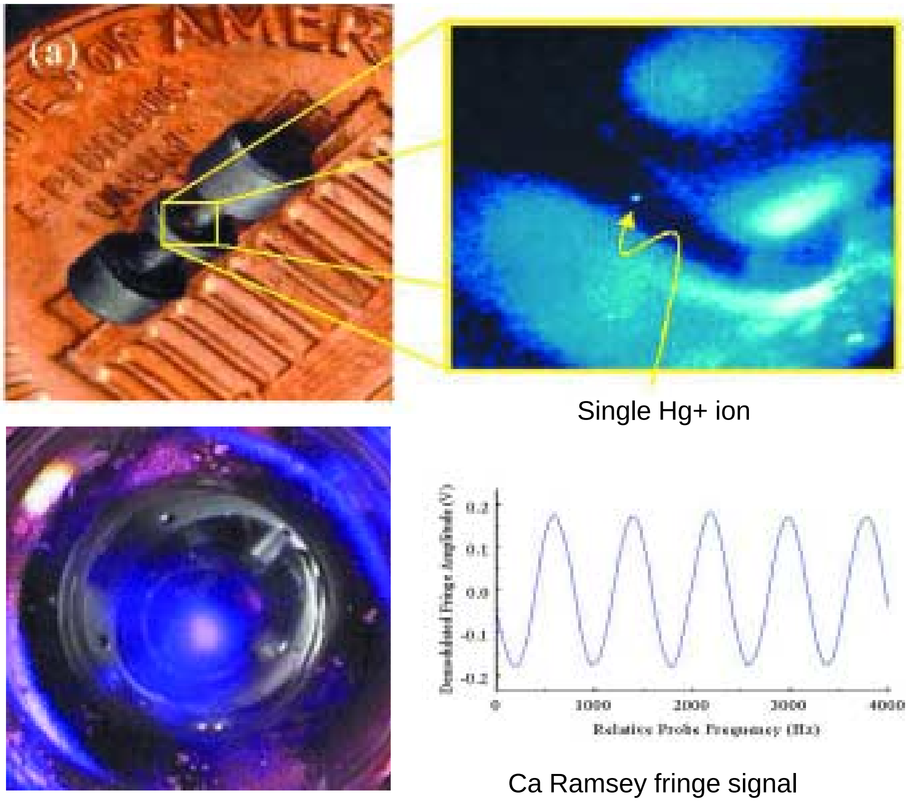

Figure 5. The NIST work on (a) Single mercury ion and (b) cold Ca atoms (with detected Ramsey fringe shown at the bottom) provides ultrastable frequency information in the optical spectrum. *(Courtesy of J. C. Bergquist (a) and L.W. Hollberg (b), NIST, Boulder)*

(EOM) in a low-loss optical cavity for enhanced modulation efficiency.[35] The optical cavity resonates with the carrier frequency and all subsequently generated sidebands, leading to a rich spectrum of frequency-calibrated lines spanning a few THz. The uniformity of the comb frequency spacing was carefully investigated.[41] By 1998, this comb generator approach had produced a spectrum extending a few tens of THz,[42] nearly 10% of the optical carrier frequency. With development of the optical frequency comb generator, a number of other interesting applications have started to emerge. At JILA/NIST, we built on the exciting development with our own unique comb generators, one with the capability of single comb line selection[43] and the other with efficiency enhancement via an integrated OPO/EOM system.[44]

But the real turning point for frequency measurement was the development of ultrawide bandwidth optical frequency combs based on Kerr-lens mode-locked femtosecond lasers. The idea of using a pulsed laser as a frequency ruler was proposed in the late 1970s by both T. W. Hänsch and V. P. Chebotayev.[45, 46] Straight-forward Fourier transformation relates a periodic pulse train of period $\tau_{r.t.}$ to a frequency domain comb spectrum 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 equal to the inverse of the pulse duration. The appearance of ultrashort pulse Ti:Sapphire lasers in the 1990s provided the bandwidth necessary to achieve rapid progress in the field: Ted Hänsch's group quickly verified the frequency uniformity of the femtosecond laser comb to a level of 3×10^{-17} and demonstrated the first optical frequency measurement using a 20 THz wide comb.[47, 48] This work marked the dawn of a new era in optical frequency metrology.

The ability to stabilize and control the wide-bandwidth frequency comb is of critical importance to its effective use. Basically there are two degrees of freedom associated with the motion of the comb. This is the result of dispersion in the laser cavity, leading to a difference in the phase and group roundtrip delays. In the frequency domain, this effect is manifested as an offset (δ) of the mode comb from exact harmonics of the f_{rep}. In the time domain, δ yields a pulse-to-pulse phase slip $\Delta\phi$ between the carrier and the envelope, given by $\Delta\phi=2\pi\ \delta/f_{rep}$. An effective stabilization approach is to lock the pulse repetition rate f_{rep} to a low-noise microwave source while at the same time locking one of the comb components to a stable cw laser. When both f_{rep} and δ are fixed, the entire comb spectrum can be represented with precisely known frequencies $f_n = n\ f_{rep} + \delta$, where n is an integer. At the same time, the frequency domain control of both f_{rep} and δ makes it possible to subsequently control the pulse-to-pulse phase slip $\Delta\phi$ between the carrier and the envelope.

The next step comes from the extension of the comb bandwidth to span an entire optical octave. This capability was achieved through the introduction of novel microstructure silica fibers (also called photonic crystal fibers). The unique dispersion properties of the fiber shift the zero of the group velocity dispersion to the Ti:Sapphire wavelength range.[49] An octave-bandwidth optical comb provides a direct connection between the microwave standard and a single laser frequency, with the interval between its frequency f and its second harmonic 2f expressed as $2f - f = f = n\ f_{rep}$, where f_{rep} is linked to a known microwave frequency. The first implementation of this idea was carried out at JILA with the 282 THz fundamental frequency of an iodine-stabilized cw Nd:YAG laser and its second harmonic,[3] leading to an improved determination of the absolute frequency of this important standard.[50] The basic scheme is shown in Fig. 3. A more elegant approach, which eliminates the need for any cw lasers, is to frequency double the infrared portion of the comb spectrum and heterodyne it with the existing visible portion of the spectrum. The resulting beat frequency is the offset δ; it can be set to any predetermined rf frequency, leading to the desired time domain control of the carrier-envelope phase $\Delta\phi$.[51]

Thus measurement of absolute optical frequencies has suddenly become a rather simple and straightforward task. Established standards can now be easily recalibrated[52] and measurement precision has reached an unprecedented level.[53] What is the next step? With the stability of the optical frequency comb now limited by the microwave reference used for phase locking f_{rep}, direct stabilization of comb components based on ultrastable optical references appears to hold great promise. Our initial demonstration of precision phase control of the comb shows that a single cw laser (along with its frequency doubled companion, as shown in Fig. 3) can stabilize all comb lines (covering one octave of the optical frequency spectrum) to a level of 1 Hz to 100 Hz at 1 s.[54] In Fig. 4 (b) we show phase lock results that are more than 10 times better than the previous frequency measurement data from a microwave-stabilized femtosecond laser comb (Fig. 4 (a)). With control orthogonalization, we expect the system will be improved further so that every comb line is phase locked to the cw reference below the 1 Hz level. Now we can generate a stable microwave frequency directly from a stable laser, essentially realizing an optical atomic clock. At the same time, an optical frequency network spanning an entire optical octave (> 300 THz) is established, with millions of frequency marks stable at the Hz level repeating every 100 MHz, forming basically an optical frequency synthesizer. The future looks very bright, considering the superior stability ($<10^{-14}$ at 1 s) offered by the optical oscillators based on a single mercury ion and cold calcium atoms developed at NIST.[55] Indeed, within the next few years it will be exciting to monitor the friendly competition between research groups working on the Cs fountain clock and those working on an optical clock based on Hg^+ or Ca, shown in Fig. 5.

A phase stable femtosecond comb represents a major step towards ultimate control of light field as a general laboratory tool. Many dramatic possibilities are ahead of us. For ultrafast science, stabilization of the relative phase between the pulse envelope and the optical carrier should lead to more precise control of pulse shaping and timing, attractive for many interesting experiments in the area of extreme nonlinear optics and quantum coherent control. For example, a phase-coherent wide-bandwidth optical comb can induce the desired multi-path quantum interference effect for a resonantly enhanced two-photon transition rate.[56] Multi-pulse interference in the time domain yields an interesting variation and generalization of the two-pulse based temporal coherent control of the excited state wave packet. The impact of the femtosecond comb approach in the optical frequency metrology community can be summarized in one sentence: Every major metrology lab in the world has started a femtosecond laser project!

Acknowledgments

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