Optical Frequency Standards and Measurement

John L. Hall and Jun Ye

***Abstract*—This paper celebrates the progress in optical frequency standards and measurement, won by the 40 years of dedicated work of world-wide teams working in frequency standards and frequency measurement. Amazingly, after this time interval, the field is now simply exploding with new measurements and major advances of convenience and precision, with the best fractional frequency stability and potential frequency accuracy now being offered by optical systems. The new "magic" technology underlying the rf/optical connection is the capability of using femtosecond (fs) laser pulses to produce optical pulses so short their Fourier spectrum covers an octave bandwidth in the visible. These "white light" pulses are repeated at stable rates (∼100 MHz to 1 GHz, set by design), leading to an optical "comb" of frequencies with excellent phase coherence and stability and containing some millions of stable coherent optical frequencies. Optical-heterodyned differences between comb lines provides a frequency-related rf or microwave output with remarkably low added phase noise, such that in an optically-based atomic clock, the phase noise of the standards-grade microwave frequency reference dominates over that of optical reference and the fs "gear-box."**

***Index Terms*—Femtosecond (fs) lasers, frequency synthesizers, laser frequency control, optical frequency comb, optical frequency measurement, stabilized lasers.**

I. INTRODUCTION AND OVERVIEW

THE first lasers were HeNe and had narrow linewidths. However, the few millihertz linewidths predicted by the Schawlow-Townes calculation were not achieved (usually have never been achieved) because of technical limitations, mainly vibrations of the laser structure. Considering that the cavity Q greatly exceeds that of the Doppler atomic line, frequency stability was provided almost solely by the physical length reference. Locking to the Doppler line provided a useful improvement, but was soon eclipsed by locking to the narrow spectral feature at line center that results from extra saturation when both running waves interact with the same atoms—the so-called "Lamb dip." Locking to this saturated absorption feature provided several decades of stability improvement according to the narrower linewidth. The separation of gain and reference function by Lee and Skolnick allowed optimizing each function by choice of pressure. The versatility was extended to using separate absorption and gain atoms, but the required matching of emission and absorption spectra was essentially not possible before molecular absorbers were introduced [1]. Molecules have dense spectra and the likelihood of spectral overlap is greatly enhanced. The first and still one of the better systems used a Methane absorption cell with the HeNe laser gain at 3.39 μm. The first report showed relative frequency stability of 1×10^{-13} at 300 s and frequency reproducibility of 1×10^{-11}, the latter a factor 400 better than the length standard of that day, the 605.7-nm spectral line emitted by a Krypton discharge held at the triple point of nitrogen ($\sim$63 K). Modern HeNe/CH_4 and CO_2/OsO_4 systems [2] show reproducibilities near 1×10^{-13} and stabilities better than 1×10^{-14}. Similar systems using the HeNe 633-nm red line and intra-cavity absorption by molecular Iodine were soon described. Over time, many laboratories worked with the HeNe $^{127}I_2$ system and the performance improved to the level that the Comité International des Poids et Mesures was able to provide recommended operating conditions assuring frequency conformity within 12 kHz 2.5×10^{-11}. An absolute measurement of its frequency allowed redefinition of the International Meter in 1983 in terms of an adopted value for the speed of light, thus reducing by one the number of independent, base physical standards.

Before the year 2000, knowledge of the absolute frequency of such lasers was hard to obtain, mainly because their frequencies were some 50 000-fold higher than that of the Cs reference at 9.2 GHz, so no "one-step"multiplier was available. Thus, a "chain" to coherently phase-relate these two kinds of stable oscillators would have to contain many links: typically, a dozen stages of phase-locked intermediate optical oscillators would be used, along with a similar number of rf phase-locked oscillators to make up the small differences. Harmonic chains were set up by national standards laboratories like NIST, NPL, PTB, LPTF, ILP, VNIIFTRI, NRLM, and probably others. A useful simplification based on difference-frequency synthesis was pioneered by NRC using CO_2 lasers [3]. Later Telle, Meschede, and Hänsch [4] developed a related scheme of "frequency interval bisection." A recent paper discusses this epoch [5].

This report gives a broad-brush view of the current situation in which we enjoy an incredible octave-spanning bandwidth produced with femtosecond (fs) laser techniques. The mutual phase-coherence within this forest of narrow "optical comb" lines makes it possible to relate an rf or microwave frequency to an optical one in a single direct step [6]. Rather than requiring years of planning and work by a team of specialists, it is now possible to set up and measure some stable laser (say a transportable transfer laser frequency reference at 532 nm) within a few hours. Any other visible color could also be measured to 13 or 14 digits in the same time—in brief, we are enjoying a broadband technology that spans from, say, 400 to 1200 nm and which has a precision and accuracy that is mainly limited by the existing rf sources and standards. Additionally, a variety of tunable laser systems are being used to investigate the spectroscopic

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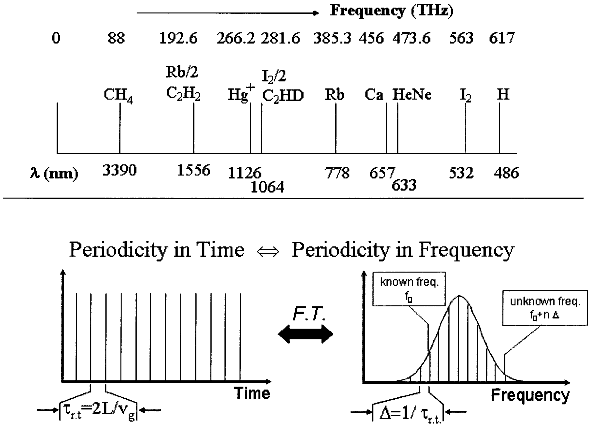

Fig. 1. Two approaches to optical frequency measurement. Top panel shows several well-studied optical frequency references on a linear frequency axis. A common separation, ~88 THz, suggests a measurement strategy based on cw lasers, with sum and difference frequency mixing. Small ($\sim$ THz) frequency residuals are measured via passive comb generators using intracavity microwave phase modulation. The new technology (lower panel) employs a regular train of short optical pulses to produce a corresponding "comb" of regularly-spaced optical frequencies.

and other properties of many new potential quantum standards, from single trapped ions to laser-cooled atomic clouds, optically-trapped atom clouds, and, of course, traditional gas cell systems, sometimes using an intra-cavity location. A number of frequency measurements have already been reported, including Yb^+ [7], Sr^+ [8], In^+ [9], Hg^+ [10], H [11], Ca [10], [12], and I_2 [13]. The performance of several of these new systems [10] already is competitive with the best rf standards based on atomic fountain realizations of the Cs hyperfine transition. We can confidently expect an exciting future as these various systems are explored and some of the interesting atomic clock applications realized.

II. BASIS FOR A NEW FREQUENCY METROLOGY

By the mid 1990s, a number of laser stabilization and spectroscopic results were known, including saturated absorption in gas cells, gas samples cooled in magnetooptic traps, and single trapped ions. Fig. 1 (top panel) shows a few selected systems, displayed on a linear frequency axis.

The community has come into this new epoch in two steps. Fig. 1 (top panel) shows the situation in 1995 when it had been noticed that differences between the frequencies of various stabilized lasers tended to be a constant value ~88 THz, conveniently the frequency of the CH_4-stabilized HeNe at 3.39 μm. Usually, one had a THz or two leftover, which could be effectively measured using the modulation-produced optical comb generators pioneered by Kuorogi [14]. Such a measurement of the 532-nm I_2-stabilized Nd:YAG laser frequency was reported as late as 1998 [15]. By 1999, the main ideas were in place for the modern epoch. Chebotayev recognized that using a repetitive pulse train would define a spectral comb [16] already in 1977. Several additional issues were considered by Hänsch, including his 1978 paper [17] which showed a clear frequency measurement between two spectral lines in terms of comb frequency intervals (Fig. 1, lower panel). The width was increased to 20 THz with the advent of short-pulse fs lasers [18]. A major breakthrough of 1999 was the Lucent Bell Laboratories report [19] of spectral broadening from $\sim$10% to a bandwidth exceeding an optical octave, which was made possible using an optical fiber of special design, the so-called photonic crystal fiber. This fiber design consists of a central core, surrounded by fine hollow tubes that provide airspaces just outside the core. By design, such a fiber can exhibit single-mode propagation over a wavelength range greater than 2:1, with a wide wavelength interval over which the pulse group-velocity dispersion is nearly zero. The large index of refraction drop from glass to the near unit value in the mostly-air region increases the limiting angle of total internal reflection, leading to a much smaller core diameter than customary single-mode fiber. This 20-fold area reduction is coupled with a long distance ($\sim$10 cm) over which the input pulse bandwidth will have the same group velocity. The modern fs laser may have an inter-pulse interval of about 2 ns, while the pulse durations may be 15 fs. The result is that we are feeding $>$10 kW pulses into a fused-silica rod of a few square microns area. This power density of about 1/2 TW/cm^2 sets the stage for serious optical nonlinearity; by deep phase modulation, thanks to the intensity-dependent refractive index, basically white light is generated by a single excitation pulse. The fiber is not melted or damaged, however, and is ready to receive the next of the few 100 million pulses we feed it each second. What is the spectrum of an "infinite" train of white light pulses? It is clear to expect the periodicity in the time domain will manifest itself as a periodicity in the frequency domain. However, in general, the comb of frequencies—when extended to near zero frequency—will not match up with the repetition rate and the frequency zero. This arises in the fs laser because the cavity modes are defined in terms of the phase velocity, while the envelope repetition rate is fixed by the group velocity. As these velocities generally differ, the details of the optical oscillations under each successive fs pulse differ: Basically, a phase slip between successive output pulses appears as an extra optical phase. So, the general formula of a comb-line frequency is

$$f_n = f_{\text{ceo}} + n f_{\text{rep}}. \tag{1}$$

Here, f_n is the optical frequency of comb-line n, with $n \sim 5 \times 10^6$, for a pulse repetition rate of 100 MHz. The carrier-envelope offset frequency is f_{ceo} and the phase slip per pulse is δ, with

$$\delta = 2\pi \left(\frac{f_{\text{ceo}}}{f_{\text{rep}}} \right). \tag{2}$$

We turn now to the frequency stabilization of this comb of optical frequencies. We have just seen that the comb has two degrees of freedom that will need controlling. Early experiments utilized almost any handy rf clock to stabilize the pulse repetition rate. Of course this works, but few rf sources are clean enough that a sharp line is still present after phase multiplication by a factor of 5×10^6. A compromise is to stabilize the fs laser only for slow drifts, but this is only partly satisfactory due to vibrations that affect the fs laser and modulate all the comb frequencies. By far, the best solution is to begin with a highly stable optical source, such as the I_2/Nd:YAG system at 532 nm. Fig. 2 shows the setup where two frequencies, f and $2f$ from the reference laser, are compared with their corresponding

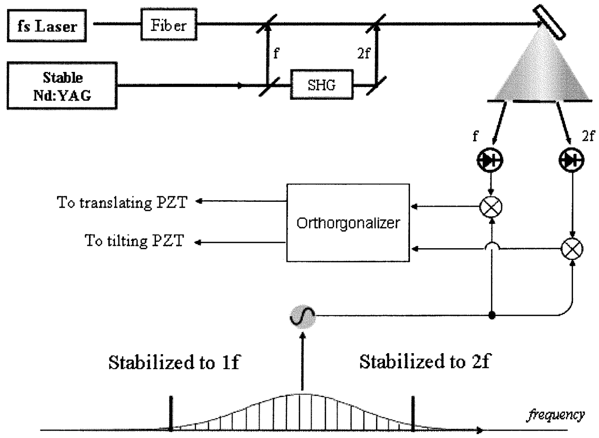

Fig. 2. Prototype of an optically-based clock. The fs laser has two frequencies needing stabilization. Because these appear differently in the two beat frequencies, full orthogonalization is possible (see text).

comb-lines. From (1), we can see the two comb frequencies will be

$$f_n = nf_{\text{rep}} + f_{\text{ceo}} \tag{3a}$$

$$f_{2n} = 2nf_{\text{rep}} + f_{\text{ceo}} \tag{3b}$$

and so we can find f_{rep} and f_{ceo} separately

$$nf_{\text{rep}} = (f_{cw} + f_{b,1064} - f_{b,532}) \tag{4a}$$

$$f_{\text{ceo}} = f_{b,532} - 2f_{b,1064} \tag{4b}$$

where $f_{b,1064}$ and $f_{b,532}$ are the signed difference frequencies of the reference frequencies f_{cw} and $2f_{cw}$ above their respective comb-lines.

With our fs laser of 100-MHz repetition rate, these beats have respectively about 25-dB and 30-dB S/N ratios in a 100 kHz measurement bandwidth. At this level, a phase-tracking oscillator is useful to avoid dropped counts. Our 750-MHz laser gives beats about 10 dB better S/N and allows direct counting. Error information derived from these frequencies may be used in several ways to control the fs laser. These frequencies are natural for a laser physicist to consider as a good basis. However, it is also reasonable to focus on the laser transducers we may control: what are their characteristic speeds and what do they do to the laser? (see the discussion in [20]). As a simple illustration here, we note that generation of a stable rf frequency from an optical reference needs to involve only f_{rep} .

A recent paper illustrated use of an optical standard to generate a stable rf frequency [13]. Using the error signal generation ideas noted above, a 100-MHz fs laser was accurately slaved onto our stable 532-nm source. This optical reference system [21] has a stability represented by $4 \times 10^{-14}/\tau^{1/2}$, which transferred to the fs repetition rate with a precision of 2.8×10^{-15} at 1 s. This produces some 5×10^6 stable optical reference frequencies; thus the locking of the fs "gear-box" is adequate for the present optical references, although their improvement seems clearly possible [22]. Comparison of the 8-GHz component in the photodetector's spectrum was made with several traditional rf sources, with results shown in Fig. 3.

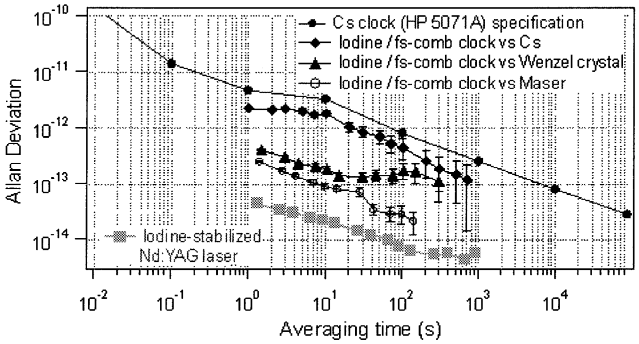

Fig. 3. Stable 8-GHz rf clock output from molecular iodine frequency standard, as compared to several high-quality rf standards. The top Allan deviation line is spec published for the Cs standard; next is our measurement versus hp Cs. The middle line results used a very quiet 5-MHz quartz oscillator, phase-locked to Cs after 300 s. The next-to-lowest trace is measured relative to a NIST hydrogen maser, relayed to us via an optical fiber. These noise levels quantitatively agree with results deduced by NIST for this source. The lowest trace shows the noise between two different I_2-stabilized Nd:YAG lasers at JILA [13], [21].

Essentially, these data suggest that the best rf stability can now be obtained by starting with a suitable optical system and gear the rate down with a locked fs laser! What the best rf systems do have is superb engineering which leads to drifts so low that integration over week-long intervals still reduces the noise. The optical systems are rather immature by comparison and usually reach some flicker floor after a few thousand seconds. However, the development of optical standards is very rapid at present so this situation can be expected to improve, leading to even larger advantages being expected for future optical sources. Fig. 4 shows the long-term frequency standard performance for our JILA gas-cell-based Iodine-stabilized Nd:YAG system. These absolute frequency data show an absolute frequency which is stable over two years, measurable to 6×10^{-14} or better in a day relative to 1-GHz timing marks based on the NIST H maser (kindly transmitted to us by NIST colleagues using optical fiber from NIST). The H-maser, the JILA laser, and the link all contribute to the noise in these early measurements.

The sharing and comparison of *optical* frequencies by fiber transmission will be an interesting capability. We recently sent our stable 1064 nm signal via the Boulder Regional Area Network fiber to NIST and back to JILA, a 6.89-km round-trip distance. Even though the fiber is in buried conduit for almost all of its travel, the returned beam was spectrally broadened by about 1 kHz. The near-gaussian shape is consistent with a large number of modest, but rather slow, perturbations. Using the fiber phase-noise compensation system [23] the linewidth was reduced to 50 millihertz with >20 dB S/N in a preliminary experiment. This is an encouraging result as the 35-μs time delay already limits the control bandwidth to ~5 kHz. It will be interesting to attempt longer fiber runs to investigate the "repeater" laser requirements when we try to disseminate an optical frequency by means of a fiber network.

An important fs servo technique [24] is called "self-referencing, " wherein the offset frequency f_{ceo} is phase-stabilized to a rational fraction of f_{rep}. This has the consequence that the optical field shape under the pulse envelope repeats exactly after a defined number of pulses.

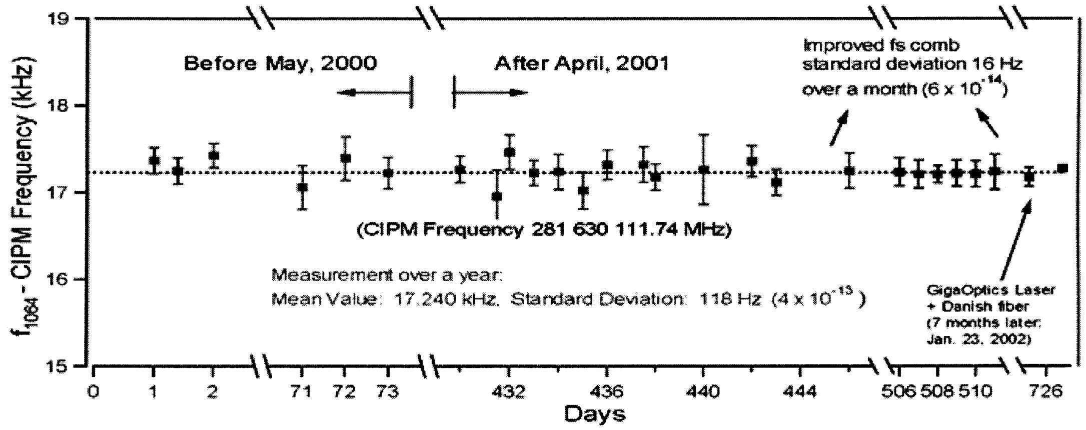

Fig. 4. Long-term frequency measurement of JILA-W iodine-stabilized laser, referenced to NIST Cs.

A related important direction is the use of frequency-domain phase-locking techniques to control time-domain relationships between laser pulses, and coherently "stitch together" fs lasers with differing output spectra. Work at JILA has shown the removal of jitter between two separate fs lasers [25], recently to the sub-fs level [26]. Controlled offsets can be provided to the repetition-rate locking loop to provide stable programmable time delays for the controlled laser. By also phase-locking together the two combs in their spectral overlap region, one can synthesize a more powerful pulse of increased bandwidth. Under proper phase control, such pulses do exhibit the expected time-domain shortening [25]. One future application area will be in chemistry and biochemistry experiments using various pump-probe nonlinear spectroscopies, all of which will be strengthened by accurate timing and removal of timing jitter [27].

This truly is the beginning of a golden age of progress in the mutually-contributing fields of ultra-stable frequency-stabilized lasers, ultrafast fs lasers, ultra-nonlinear optical photonic crystal fibers, and ultra-sensitive spectroscopies.

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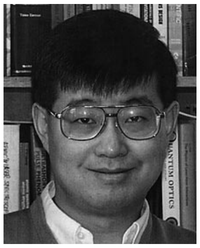

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