

**FROM STABLE LASERS TO OPTICAL-FREQUENCY  
CLOCKS**  
**Merging the UltraFast and the UltraStable, for a New Epoch of  
Optical Frequency Measurements, Standards, & Applications**

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This is a report on behalf of the World Team of Stable Laser and Optical Frequency Measurement Enthusiasts, even if most detailed illustrations draw mainly from our work at JILA. Specifically we trace some of the key ideas that have led from the first stabilized lasers, to frequency measurement up to 88 THz using frequency chains, revision of the Definition of the Metre, extension of coherent frequency chain technology into the visible, development of a vast array of stabilized lasers, and finally the recent explosive growth of direct frequency measurement capability in the visible using fs comb techniques. We present our recent work showing a Molecular Iodine-based Optical Clock which delivers, over a range of time scales, rf output at a stability level basically equivalent to the RF stability prototype, the Hydrogen Maser. We note the bifurcation between single-ion-based clocks – likely to be the stability/reproducibility ultimate winners in the next generation – and simpler systems based on gas cells, which can have impressive stabilities but may suffer from a variety of reproducibility-limiting processes. Active Phase-Lock synchronization of independent fs lasers allows sub-fs timing control. Copies of related works in our labs may be found/obtained at our website <http://jilawww.colorado.edu/yehall1labs>

**1. Introduction: The Laser Metrology Epoch started with a Revolution**

The stable laser epoch began with the introduction of the HeNe cw laser and exploration of its coherence properties by Javan in the early 1960's. New far-ir laser systems were found and their frequencies measured<sup>1</sup>. With the shorter wavelengths, it soon appeared possible to perform improved measurements of the speed of light. However, the traditional microwave approach, ie. measuring  $\lambda$  and  $\nu$  to calculate  $c = \lambda * \nu$ , then seemed ill-suited since no known methods could deal with and measure such high frequencies. An alternative, to determine  $c$  from measured *differences* in frequencies and wavelengths of nearby laser lines, led to development of dual wavelength lasers in Novosibirsk and design of experiments in JILA where a 30 m evacuated interferometer was established in an unused gold mine in the mountains

west of Boulder. Stability of the fringes obtained using a Lamb-dip-stabilized HeNe 633 nm laser there (and in the laboratory comparison with the incoherent 605.7 nm radiation defining the Krypton-based Metre) rapidly led to the realization that a high performance optical frequency standard was a practical necessity for success in the speed of light measurement. An important paper by Lee and Skolnick<sup>2</sup> showed that separate absorber and gain cells could be used to obtain “inverted Lamb dips” and soon led to a nice system using low pressure CH<sub>4</sub> as the absorber and HeNe as the gain medium for the 3392 nm transition. Barger & Hall’s system<sup>3</sup> worked astonishingly well, producing reproducibility in the range  $\sim 10^{-11}$  (vs.  $4 \times 10^{-9}$  for Krypton) and medium-term stability a hundred-fold better. Indeed, even now, this laser is one of the finalists for a good practical working standard for frequency, with reproducibility in the low  $10^{-13}$  domain based on telescopic resonator designs by two Russian groups, led by S. Bagayev and by M. Gubin.

To convey the sense that this represented a veritable “phase transition” in the art of defining optical frequency, consider Fig. 1 which shows interferometric fringes of the new contender, the HeNe/CH<sub>4</sub> laser system, and those of the internationally-accepted krypton standard discharge lamp, operated under the

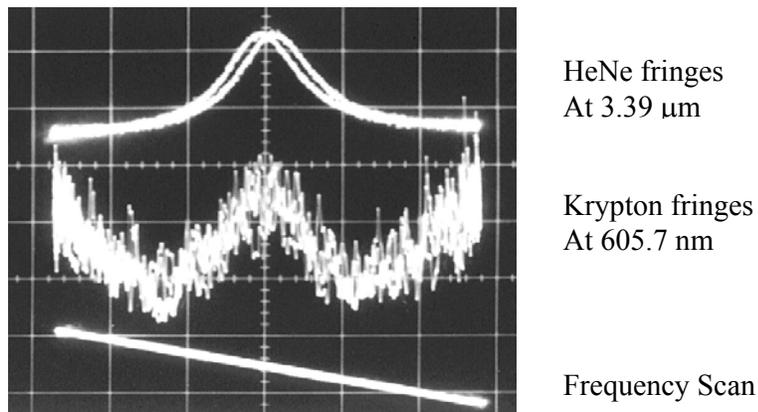


Fig. 1. A New Wavelength Standard? Interferometer fringes of Kr wavelength standard, with discharge lamp operated at the recommended current and temperature and under the metrological conditions optimised for its accurate measurement (middle trace). Top trace shows HeNe laser fringes with mirror finesse chosen to produce a similar width. Bottom trace shows accurately-linear frequency-based scan of interferometer length.

conditions recommended by the BIPM/CIPM for the attainment of the maximal wavelength reproducibility. (It was called a “wavelength” standard in those days, although surely it was actually an incoherent standard of optical frequency.) From this scope display one sees that a precision matching the Krypton’s nominal accuracy is obtained in a single sweep for the laser and only after about 30 minutes with the discharge lamp. One can reasonably discuss this 1971 data as a “revolution in optical wavelength/frequency metrology.”

The almost unbelievable miracle is that now, just 30 years later, we can simply measure such laser frequency standards directly against the standard of frequency, the 9.2 GHz hfs transition in Cs. Indeed, progress in the optical domain has been so powerful that one now has a choice of optical systems which outperform this venerable and well-researched rf standard defining the SI unit of time. After a flurry of activity in the last two years, this second measurement revolution is nearly completed! Before we discuss this new measurement paradigm, and consider where it is leading us, a useful next task of this paper is to rapidly spin across these three decades of advances.

## 2. Measuring the Stable Laser’s Frequency

As noted above, in the 1970’s a major industry soon developed with standards labs setting up chains of lasers of successively higher frequencies, each locked to some harmonic of a lower frequency one. Typically the first step at NBS used harmonics from a phase-locked klystron at 74 GHz to generate a sub-mm-wave output, to lock the frequency of the HCN laser at 890 GHz. This laser’s frequency was multiplied by 12 in another point-contact diode to reach the H<sub>2</sub>O laser at 10.7 THz. In turn this laser was multiplied another 3-fold to reach a CO<sub>2</sub> laser line at 32.1 THz. Three harmonics of the 890 GHz HCN laser were mixed with this CO<sub>2</sub> laser in a Metal-Insulator-Metal diode to lock another CO<sub>2</sub> at 29.4 THz. Finally, Evenson et al. mixed the third harmonic of this laser with a HeNe power laser at 88 THz (3.39 μm) to measure its frequency and so that of the CH<sub>4</sub>-stabilized laser<sup>4</sup>. Using the interferometer system described above, Barger and Hall measured<sup>5</sup> the wavelength of this laser against the krypton standard and so the NIST team could give a dramatically-improved value for the Speed of Light<sup>6</sup>. Other laboratories, notably NPL<sup>7</sup> in the UK and NRC<sup>8</sup> in Canada built equivalent frequency measuring chains and the measured value of *c* was basically confirmed. Many measured frequencies are tabulated<sup>9,10</sup>. Impressively, the new direct frequency measurements so far always confirm the ~10<sup>-10</sup> accuracy of the NPL frequency-controlled interferometer results!

### 2.1. *Extending the Chains into the Visible – re-definition of the Metre*

The several national metrology labs saw opportunity in the many possible stabilized lasers, each adopting their favourite system for further study. At NBS a system was set up by Jennings et al, based on the measured CH<sub>4</sub> frequency, to extend the measurement range to the visible HeNe Iodine-stabilized laser at 633 nm<sup>11</sup>. With this frequency value, confirmed by other national laboratories, it became feasible and attractive to eliminate the Kr wavelength standard by adopting a conventional value for the speed of light. Thus in 1983 the CIPM announced adoption of the value  $c = 299,792,458$  m/s, exactly. Some reflections on this epoch appeared recently<sup>12</sup>, but the main message is that these multistage chains are really hard work! One typically can obtain a frequency multiplication factor of only two in the near visible because we must work with frequency-doubling and mixing in crystals: the stronger nonlinearity of the MIM diodes is no longer effective at these high frequencies. The first coherent and most impressive rf-visible chain of this traditional type was built by Schnatz et al. at PTB to measure<sup>13</sup> their Ca optical frequency standard: this employed 11 lasers, 7 special microwave sources and their associated phase locking circuits, with 12 laser frequency-mixing stages leading to 7 laser phase-locks and at least 2 counted beat frequencies. A frequency uncertainty of only 430 Hz ( $<10^{-12}$ ) was obtained for the Ca 456 THz (657 nm) transition. A measurement<sup>14</sup> of the Sr<sup>+</sup> transition at 445 THz was made by Bernard et al. at the NRC in Canada soon after, using a folded version of the chain concept, whereby harmonics of *difference* frequencies are successively multiplied to eventually become equal to one of the optical frequencies (~30 THz with CO<sub>2</sub> lasers). The measurement uncertainty was only 200 Hz. Using the Optical Frequency Interval Divider concept<sup>15</sup>, Udem et al. in 1997 used the Garching chain to provide a high precision measurement of H 1S-2S frequency<sup>16</sup> and in 1999 von Zanthier et al. measured the In<sup>+</sup> clock transition<sup>17</sup>. These were fundamental achievements, fully successful from a scientific perspective. But a big liability of the method is its lack of versatility: each particular new frequency to be measured will need its own setup.

### 2.2. *Many Optical Frequency Standards – the “standard” interval*

By 1995, a number of frequency standards were developed and their performance characterised. Fig 2 depicts this situation. While the laser systems are selected at “random,” inspection of Fig 2 shows that there tends to be a relatively standard frequency interval of 90 THz between them. What variations exist could be taken up if we just had a source capable of spanning intervals  $<20$  THz. In fact such a

versatile tool had been developed by Kourogi et al, in the form of an optical frequency comb generator<sup>18</sup>, based on strong microwave excitation of a low loss optical phase modulator contained within a cavity of moderate finesse. This work directly demonstrated sidebands covering ~4 THz. If broader bandwidth were desired, it could be obtained by amplifying the original modulator’s optical output in a fiber amplifier to the ~1 W level, and then sending this output into a low-dispersion fiber. The concentrated field distribution and extended length for the nonlinear interaction provided output bandwidths up to 30 THz<sup>19</sup>. This tool allowed connection between known and “unknown” frequency sources and at 1.5 μm was applied by Nakagawa<sup>20</sup> to make a C<sub>2</sub>H<sub>2</sub> atlas. At JILA, one such measurement<sup>21</sup> was made of the Nd:YAG laser stabilized to Iodine at 532 nm, based on the known reference HeNe line at 633 nm and the Rb two-photon reference at 778 nm. This I<sub>2</sub>-based system is of particular interest for its remarkable stability as we shall see.

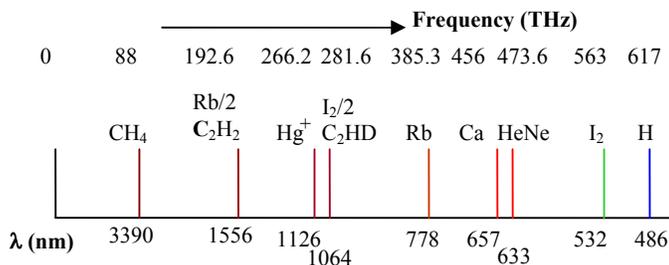


Fig. 2. A representative collection of well-studied optical frequency references. The amazing thing to note is the near-regularity of the optical frequencies! With the 88 THz HeNe laser and Periodically-Poled LiNbO<sub>3</sub>, one can get interestingly close to spanning these intervals. Just add Kourogi’s <30 THz comb and the frequency synthesis job is done! A 1995 Dream.

### 3. The Femtosecond Comb and Nonlinear Fiber arrive

Already in the mid to late 1970’s, Ted Hänsch<sup>22</sup> and Venia Chebotayev<sup>23</sup> were thinking about the Fourier spectral picture for the short ps pulse lasers which were beginning to be available. A single pulse with a short duration would have a broad, unstructured continuum as its spectrum. If we had a regularly occurring train of pulses, an instrument of reasonable spectral resolution would combine a number of these pulses coherently together due to its intrinsically low temporal resolution. Inter-pulse interferences would then lead one to expect a “comb” spectrum, composed of many spectral lines with a common frequency separation equal to the

repetition rate of the input train. These would be spectrally-sharp if the input laser had a stable repetition frequency. Indeed, in 1978, Eckstein, Ferguson & Hänsch measured<sup>24</sup> some hfs intervals using the pulse repetition rate as the effective frequency “ruler.”

In 1993 the remarkable Ti:Sapphire laser was shown by Asaki et al. to be capable of generating pulses<sup>25</sup> as short as 10 fs. Very short indeed, but even for these pulses the spectral width was “only” about 150 nm, centered at ~800 nm. The final magic component was the introduction at the 1999 CLEO of Photonic Crystal Fiber, PCF, by Ranka, Windeler, and Stenz<sup>26</sup>. They showed that a modest commercially-available fs laser would generate a full rainbow of colours as its light passes through even a rather short length of this fiber. Of course, the fiber *is* intrinsically nonlinear (but at a very low level) due to the Kerr effect, basically an intensity-dependent increase of the index of refraction. Because the fiber is fabricated as a rod surrounded by air-filled voids, there is a large discontinuity in the index of refraction, which translates into a relatively large critical angle for total internal reflection. In diffraction-controlled optics, such a large angular aperture could be coupled with a small spatial aperture to achieve low-loss single-mode propagation. Thus for a single mode “holey” fiber, the effective core diameter may be something like 2 micrometers, leading to an effective area of around  $A=3 \times 10^{-12}$  m<sup>2</sup>. Really Small! The fs laser, on the other hand, produces perhaps 300 mW of time average output, but the pulse duty factor is only about  $10^{-6}$  (10 fs wide pulses every 10 ns). Not all the power will enter the fiber, so we can suppose an input peak power of 100 kW entering the aperture A, which corresponds to an internal intensity of  $3 \times 10^{16}$  W/m<sup>2</sup>. This is a seriously high power density even for fused silica, one of our least nonlinear materials. The result is the index is strongly driven ( $\Delta\Phi \sim 30$  rads) and so an iterative and massive self-phase-modulation occurs. The “modulation frequency” for this process is related to the fs pulse envelope components (50-100 THz) so a huge spectral broadening can result in just a few steps. Another property of the multi-structured fiber is that it can be designed to have basically zero group-velocity-dispersion at the chosen wavelength, so the high power impulse and all the Bessel modulation sidebands propagate together coherently to continue the spectral conversion from “broad line” of the short impulse to a pure continuum output. Of course the basic repetition rate spectral periodicity is maintained by the interpulse interference in low bandwidth detection/analysis. It seems completely natural to use the name “fs laser comb” to describe such an articulated spectrum of perhaps 1 million equally-spaced spectral needles.

### *3.1 Development of the fs comb techniques for frequency measurements*

The Garching group, under Ted Hänsch's leadership, were the pioneers in applying the ultrafast laser comb technique to measuring frequency intervals which arose in their measurements of frequencies in the fundamental spectrum of atomic Hydrogen. Previously the "frequency-interval divider method" had been used<sup>15</sup> with 5 stages to span the 2.1 THz interval between the 7<sup>th</sup> harmonic of the CH<sub>4</sub>-stabilized standard and 1/4 of the excitation frequency for the 1S – 2S transition. By 1999, in a beautiful paper<sup>27</sup>, Reichert et al. discussed the fiber-generated spectral broadening concept, in combination with frequency-interval division to measure a 45 THz interval which effectively was 1/2 the frequency of the CH<sub>4</sub>-stabilized laser. This interval was obtained as the sum of a large number of fs comb-line intervals and consequently could be related to the measured/stabilized pulse-repetition rate of the fs laser. At the same time, a relationship would be established with the Hydrogen transition of interest. This technique was used first by Udem et al. to measure the Cs D1 transition<sup>28</sup>, and marks the beginning of the epoch of direct optical frequency measurement whereby an optical frequency can be obtained directly in terms of the pulse repetition rate, without requiring any other known optical standards. Later that year, the Hydrogen frequency was measured by Reichert et al.<sup>29</sup> with this method, followed in 2000 by Niering et al.<sup>30</sup> with the definitive H 1S-2S measurement using the transportable Cs fountain.

Of course such dramatic progress attracted others into this field. Diddams et al.<sup>31</sup> at JILA followed Kourogi's EOM comb-generator system, adding intra-cavity OPA gain to suppress the role of losses and even offer self-sustained Optical Parametric Oscillation. The intracavity EO Phase Modulation connected mainly adjacent frequency components, but with careful tuning one could obtain a >5 THz interval filled with coherent comb components separated by the common 330 MHz FSR. Extension to broader bandwidths would need careful attention to compensation of the intracavity dispersion, arising mainly from the EOM crystal. A functionally-similar device – the fs optical impulse generator – was already known, and it became clear that this fs Ti:Sapphire technology was the system of choice. For the intended frequency metrology purpose, not only is it more robust and convenient, it has the fundamentally important advantage of spectral cross-coupling over the whole pulse spectral width. This will drastically reduce the phase noise within the comb. Basically the Ti:Sapphire laser modelocking process depends on a time-gating effect whereby the high peak intensity of a coherent superposition of modes leads to Kerr-effect lensing in the Ti:Sapphire crystal. With suitable cavity adjustment, the losses can be lower in the presence of this additional positive "Kerr-

lens” and so a self-stabilized short pulse evolves, with bandwidth limited finally by uncompensated dispersion which spectrally affects the cavity recurrence time. New JILA colleagues, Margaret Murnane and Henry Kapteyn, had earlier developed better group-velocity compensation strategies and were kind to share their technology with our cw laser group. A decisive advantage was offered by JILA colleague Steve Cundiff who earlier had been a researcher at Lucent Technologies’ Bell Labs in the group where the microstructure fiber work had been done: we were able to obtain a sample of this material in late 1999. Of course almost instantly the JILA team also had a broadband “white” output from the fiber. And of course spectral components near 1064 nm and 532 nm could be heterodyned with corresponding cw frequencies coming from our well-studied optical frequency standard (“JILA-West”) based on Iodine spectroscopy with a doubled Nd:YAG laser. So using Hänsch’s comb method, one could measure the optical frequency  $f$  as the difference between the green  $2f$  and the IR frequency  $f$  by counting the comb-lines in between. Then any visible frequency could be obtained as an interval from the now-known Iodine-stabilized fs laser comb. A multi-laboratory paper celebrated arrival of this single-step rf-visible coherent connection<sup>32</sup>.

One interesting nuance of the fs laser arises from the existence of two kinds of boundary conditions. Of course the laser oscillation is formed of normal cavity modes, satisfying phase-integral boundary conditions on the mirrors. In contrast, the repetition rate of the output pulses is connected instead to the loop circulation time and thus involves the *group* velocity, rather than the *phase* velocity. This distinction means that the optical pulses in fact are not exactly the same pulse-after-pulse, in that the envelope may be slipping relative to the underlying cavity mode frequencies. The small phase slip per pulse will, after some time, lead to a full  $2\pi$  cycle of phase difference: the inverse of this recursion time will therefore appear in the optical spectrum as a common frequency shift of the entire comb from the position of exact harmonics of the repetition rate. The language of the “carrier-envelope phase slip rate”  $f_{\text{ceo}}$  is often used to describe this frequency offset. The bottom line is that the optical comb spectrum needs to be described by two numbers: the repetition rate  $f_{\text{rep}}$  (and its harmonics) AND the carrier-envelope offset frequency,  $f_{\text{ceo}}$ . With only a single laser, how can we measure this offset?

Jones et al. at JILA introduced<sup>33</sup> an interesting technique to simplify the use of fs frequency comb metrology by making the spectrum “self-calibrating”. This “self-calibrating” idea turns on the fact that the fiber-broadened spectrum covers an octave frequency range. A comb component  $f$  in the near IR,  $f_n = n f_{\text{rep}} + f_{\text{ceo}}$ , can be matched with an emitted comb component in the yellow, near  $2f$ ,  $f_{2n} = 2n f_{\text{rep}} + f_{\text{ceo}}$ . Additional light can be produced at this color also by frequency-

doubling part of the IR power to obtain  $2 f_n = 2*(n f_{\text{rep}} + f_{\text{ceo}})$ . Combining the two sources of yellow light leads to a common beat frequency of  $f_{\text{ceo}}$  between each pair of components. Use of an AOM frequency shifter in one arm of this “interferometer” translates the desired information away from the spectral clutter at  $0, f_{\text{rep}}, 2 f_{\text{rep}}, \dots$ . Straightforward phase-locking techniques then let us choose from a number of lockpoints, for example 16, stabilizing  $f_{\text{ceo}} = m *(f_{\text{rep}}/16)$ . See Jones et al.<sup>33</sup> for details. For frequency synthesis purposes a smaller step may be desirable: we are using 1000 steps with our GHz-rate fs laser. More generally, it may be useful to organize the fs laser system so that exactly equivalent optical pulse shapes are produced, say after an integer number of pulses. In such case, time-domain systems with a “pulse-picker” could be assured that each next selected pulse is equivalent. (Actually, it is the *rate* which is stabilized so some slow phase drift may eventually be expected, although this is small because the servo’s effective integrator is numerical and the noise processes are observed to have zero mean.)

### 3.2 Application of the fs comb to frequency measurements

In the last two years there has been an explosion of absolute frequency measurements using fs comb methods, beginning with the Garching measurement of the Cs D1 line<sup>28</sup>, closely followed by the high precision Hydrogen measurements<sup>29,30</sup>. As now observed by all groups, the actual limitation with fs-based measurements in fact is the quality of the available rf source. For example commercial Cs clocks have a stability  $\sim 5 \times 10^{-12}/\tau^{1/2}$  and can be calibrated to an accuracy  $\sim 1 \times 10^{-14}$  via careful and extended use of common-view GPS<sup>34</sup>. The Garching solution to the rf clock problem was to collaborate with colleagues at LPTF/ENS in Paris who have developed a transportable Cs fountain clock. The resulting H 1S-2S result<sup>30</sup> was quoted at the  $1.8 \times 10^{-14}$  accuracy level and certainly set a high standard for the field. A JILA-NRC-BIPM collaboration showed equivalence of the comb and chain metrology approaches at the  $1.6 \times 10^{-12}$  level using a 633 nm transfer laser<sup>35</sup>, as well as the need for a +7 kHz correction of the adopted HeNe frequency<sup>36</sup>.

So the next progress can be expected in two ways: some national labs have both trapped ion teams and primary frequency standard teams. Surely these collaborations will be hard to beat! A NIST team gave the frequency<sup>37</sup> of  $\text{Hg}^+$  to  $<1 \times 10^{-14}$ .  $\text{Yb}^+$  was measured<sup>38</sup> with similar accuracy at PTB. Excellent results are obtained also for Ca at both NIST<sup>37</sup> and PTB<sup>39</sup>. Attention is particularly drawn to the contributions by NIST, PTB, NPL and others in this Symposium.

Another direction is to explore the “everyman’s frequency measurement system” where one can entertain tradeoffs such as a ~10-fold accuracy loss for a ~10<sup>3</sup> scale reduction for the apparatus. This next proposal may be optimistic, but 20 mW fs diode lasers already exist and super-fibers exist that give us white light with even less power. Maybe we can dream of a battery-powered fs comb system ...

Our JILA group has been exploring the issues of locking the fs laser tightly to a stable optical clock and deriving thereby an rf output with an interesting level of stability<sup>40</sup>. With outputs from the 532 nm Iodine-stabilized system, we form optical beats at 1032 nm,  $f_{\text{beat}, 1064} = f_{\text{cw}} - (n f_{\text{rep}} + f_{\text{ceo}})$  and at 532 nm,  $f_{\text{beat}, 532} = 2 f_{\text{cw}} - (2n f_{\text{rep}} + f_{\text{ceo}})$ . These ~30 dB S/N (100 kHz rbw) beats are regenerated with tracking oscillators and combined to form two orthogonal control error signals:  $f_{c1} = f_{\text{beat}, 1064} - f_{\text{beat}, 532} = n f_{\text{rep}} - f_{\text{cw}}$ ; and  $f_{c2} = f_{\text{beat}, 532} - 2 * f_{\text{beat}, 1064} = f_{\text{ceo}}$ . In the 100 MHz fs system, with dispersion-compensating prisms, for control elements we have fast

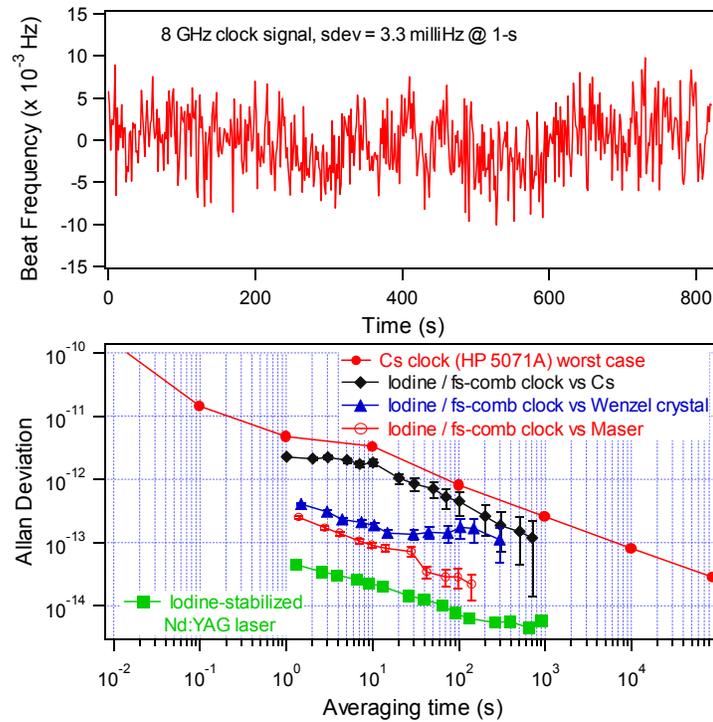


Fig. 3. Comparison of Iodine Molecular Clock rf output with other rf sources.

PZT control of the cavity length and slower angular control of the swivel mirror located in the spectrally-dispersed zone after the prisms. (This swivel mirror primarily affects the group velocity dispersion and hence the pulse repetition rate, while the PZT controls the optical frequencies directly and the repetition rate only slightly.) The  $f_{c1}$  and  $f_{c2}$  error signals are processed by independent servo loop controllers and fed to swivel and cavity length PZT's respectively. Adjustable cross-feed-forwards serve to reduce the locking noise  $\sim 2x$  due to improving the orthogonalization of control, although the high speed of the  $f_{ceo}$  /PZT loop makes this less essential. With feedforward, the present error of the control system is  $< 1$  Hz (1s) for both loops<sup>40</sup>. This  $< 3.5 \times 10^{-15}$  transfer capability is of course not fully needed for our present house reference laser, JILA-West, which exhibits a stability of "only"  $5 \times 10^{-14} / \tau^{1/2}$ . So we expect that the rf signal derived from this rather simple apparatus should have the same stability as JILA-West, namely 2 orders better than the commercial house Cs clock at JILA.

Of course, testing the attained performance in the rf domain is challenging when one has but one high-grade source. We have improved the short-term stability of our commercial Cs system by loosely phase-locking a selected low-noise quartz oscillator so that for  $t < \sim 100$  s we have free-running quartz, and migrate back to Cs at longer times. The Allan Deviation plot, Fig. 3, shows the comparison of our optical standard with several other references, starting at the top with Cs specification, Cs measurement, then the phase-locked filtered quartz oscillator, and an H maser signal relayed to us by NIST colleagues using a buried optical fiber.

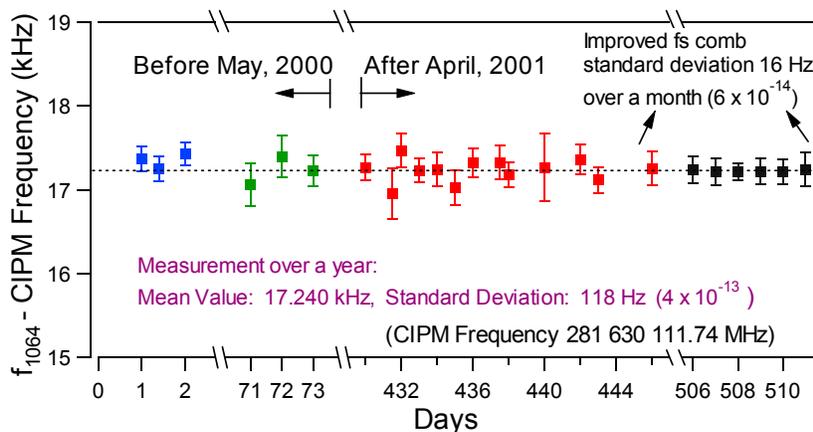


Fig. 4. Long-term stability of Molecular Iodine Clock, vs Cs (UTC-NIST)

The lowest trace shows the nominal performance of the optical reference itself, obtained from comparison of two comparable Iodine-stabilized systems. Of course this kind of research is an unending story of improvements. This makes the optically-derived rf standard rather exciting for some tasks as it already shows better performance at times below  $\sim 1$  day. With ultranarrow lines from ion-trapped species, one can expect good long-term stability and reproducibility as well.

The long-term performance of the vastly simpler Iodine gas cell approach may be summarized by Fig. 4, which shows a reproducibility of  $\pm 4 \times 10^{-13}$  over a year during the evolution of our techniques. Over the last month of data, the deviations are  $\pm 6 \times 10^{-14}$ . It is important to note that this is nearly the noise of 1 day of measuring the frequency against Cs, although we suppose there may also be some noise/drift of our optical system itself at these very long times – perhaps due to shifts of some systematic offsets in our locking system.

#### 4. Real Clocks produce Synchronized Pulses

We've shown that a frequency-stabilized laser, expressed in the rf domain with the help of a suitable fs laser “gearworks,” can form a superior rf frequency standard. The atomic clock also produces “clicks” of very high quality as well, just a few fs wide. Considering that the entire comb is strictly phase-coherent, in fact we may use tools of the frequency domain to establish useful properties in the time domain. One clear example of this idea can be expressed by the task of accurate synchronization of two pulse lasers, probably of different pulse repetition rates. We find immediate success in stabilizing two lasers of different colors such that the pulses collide with  $< 10$  fs precision on every 10 (9) pulses of the 100 MHz (90 MHz) lasers. Then using equal repetition rates, Shelton et al<sup>41</sup> used heterodyne beats between the two lasers in the region of spectral overlap to enable phase-locking together the two comb sources, so they share both the same  $f_{\text{ceo}}$  as well as repetition rates – and so are effectively just part of a broader comb. Thus one synthesises a combined pulse of greater amplitude, wider spectral bandwidth and shorter temporal duration. Improved intrinsic laser stability allows  $< 2$  fs timing jitter, along with fast ( $\sim 50$   $\mu\text{s}$ ) time-offset programming<sup>42</sup>. This synchronization capability will be of great utility in time domain experiments, for example two-color multiphoton scanning of nanoscale structures.

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