Optical Frequency Standards And Their Measurement

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Abstract. The last four years have seen a revolution in the art and practice of optical frequency measurement. Indeed, the “gearbox” represented by the spectrally-broadened fs laser’s comb has sufficiently low noise that we in the optics community have abruptly found ourselves in the tough real world well known to our rf standards colleagues who perpetually work toward more coherent interrogation oscillators. As suggested earlier by Dehmelt, a trapped, laser-cooled single-ion can form an excellent basis for a fundamental new frequency standard system. Still, laser-cooled cold gas ensembles typically offer higher S/N and so better short-term stability, but at some cost in possible shifts of the resonance spectrum. In the effort to obtain excellent stability and reproducibility from simpler (gas cell) systems, emphasis is on getting smaller shifts in the process of observing the atomic resonance. Time-domain synchronization, achieved by frequency-domain methods, brings a whole new order of experimental precision.

INTRODUCTION AND OVERVIEW

The laser frequency standards community has been busy for ∼30 years, introducing more lasers stabilized to atomic and molecular transitions, and improving their performance. By ∼1995 the situation was as indicated in Fig. 1 (top) when a number of attractive systems were under vigorous development. While they were all shown to be rather stable, as for the absolute frequencies, only a few were known because of the elaborate frequency-multiplication systems then required for the measurement. One measurement approach of the day utilized accidental near overlaps generated by wave-mixing in a nonlinear crystal. Usually a residual beat frequency in the THz range resulted, which led to the development of optical comb generators based on iterative microwave modulation within a cavity, spectral broadening in fiber, and optical frequency translator devices. The situation changed forever in 1999 when a femtosecond laser based optical frequency comb was demonstrated to be an accurate ruler for frequency measurement, expanded by the demonstration of the extreme nonlinearity of microstructure fiber when subjected to pulses from a rather modest fs laser: basically “white” light is generated. See Fig. 2. However, as indicated in the lower panel of Fig. 1, there is something really special about this “white” light: interference between the various pulses leads to an optical “comb” spectral structure whose comb-line separation is rigorously equal to the repetition rate of the pulse train. Experiments reveal no metrologically-damaging phase or frequency shifts, thus setting the stage for an explosion of accurate absolute frequency measurements. This
**FIGURE 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. Residual (~THz) frequency differences are measured via passive comb generators using intracavity microwave phase modulation. The superior new technology (lower panel) employs a regular train of short optical pulses to produce a corresponding "comb" of regularly-spaced optical frequencies.

brief review provides an overview of these results, along with some discussion of the present limitations of the measurement methods and of the sources themselves. Efforts to capture much of the good results with less complex systems are currently attractive.

**FEMTOSECOND LASER, MICROSTRUCTURE FIBER, AND THE OPTICAL COMB**

Our usual daily experience, intensified by our traditional training, presents the world as a basically linear place. Indeed, mathematical transfer functions and linear feedback concepts are powerful tools for understanding the observable world. However, there IS another domain, made accessible by ultra-short pulse lasers. For example a typical modern Ti:Sapphire laser may offer an average power of 500 mW, but this is the result of ~100 million pulses per second, each of perhaps 15 fs duration and carrying about 5 nJ. While this energy per pulse may seem miniscule, it involves only a 15 fs pulse duration and hence represents a peak power of about 1/3 MW, not quite a weak pulse power. The pulse contains only about 6 optical cycles. Important for its nonlinear applications, the beam is essentially a single TEM mode spatially and can be focused to a diffraction limited waist, such as the ~2 μm diameter of a Microstructure Fiber, and thus represents about $10^{17}$ W/m² incident (and captured) flux. The stage is set for serious nonlinearity!
Making White Light with a Microstructure Fiber

The geometry of one of the Lucent-Bell Labs fibers\(^8\) is shown in Fig. 2. The central small rod of low-loss fiber glass is seen to be supported by thin glass webs, leading to an effective "cladding" index of basically unity, as it is mostly just air. This high index of refraction contrast makes the incidence angle for total internal reflection much larger than for normal glass-clad fibers and so low-loss single-mode transmission is available for 800 nm light even with a core diameter as small as 1.7 \(\mu\)m. The dispersion is designed to be zero near 800 nm so the incident Ti:Sapphire pulses are not immediately stretched in time, maintaining their high peak power for some few cm's propagation. Of course the extreme laser intensity leads to nonlinear effects, the strongest of which arise from the intensity-dependent index of refraction. In recent JILA work\(^9\), Fortier \textit{et al.} found that the resulting phase shift is \(~3800\) rads. per \(\mu\)J of transmitted pulse energy. The rms intensity noise of the laser, integrated from 0.03 Hz- 55 kHz, leads to \(-0.5\) radian phase excursions between fundamental and second harmonic fiber signals. Recalling that the pulse's full width in the fiber is \(<50\) fs, the envelope will contain Fourier components well beyond 10 THz. Analytically, in Phase Modulation theory -- and consistent with observations in more calm electronic circuits -- it is known that the modulation bandwidth is \(~\)modulation rate \(\times\) modulation index (\(\approx\) peak phase deviation). So we see that with the UltraFast lasers we basically have a very high-order phase-modulation process, with bandwidth limitation set by uv fiber

![Microstructured fiber](image)

- dispersion zero at \(~800\) nm
- pulses do not spread
- continuum generation via self-phase modulation

FIGURE 2. Serious NonLinear Optics: Octave-level Spectral Broadening in a MicroStructure Fiber. Top left shows an electron-microscope image of the fiber cross-section. The central "optical fiber" is 1.7 \(\mu\)m diameter, the surrounding holes 1.4 \(\mu\)m diameter. Many different fiber designs are now available to provide other zero dispersion wavelengths and dispersion spectral gradients. Lower left shows tight-focus objective lenses that map the fs laser into the core and recollimate the emerging "white" light. Spectra are shown at the lower right.
absorption, IR guiding losses, and by dispersion. Visually, blue light is visible within a millimeter of the fiber's input facet. With laser pulse input width of 70-100 fs, one instead can see the highly shifted blue light evolve from green, orange and red precursor lights some meter(s) upstream. One may wonder if, under these serial conversions with lower "modulation" frequencies and phase modulation depths, would there be any larger amount of phase noise developed?

**Spectral Structure of the Comb's “White Light”**

An important issue about the fiber-broadened “white” light is its spectrum. Of course the strict time periodicity of the pulses will be Fourier imaged as the spectral interval periodicity between the various optical comb lines. From the nonlinear systems point of view, one might have expected the optical comb frequencies to be harmonics of the repetition rate. This is not generally the case: we do not have the optical frequencies by a 5-millionth order nonlinearity working on the 100 MHz repetition rate. Rather the envelope's time structure of a single fs pulse sets a bandwidth or modulation rate for the phase modulation process, and the peak FM index sets the spectral extent as explained earlier. The repetition rate appears naturally as the spectral periodicity when one thinks about the summation over all the time domain pulses and asks what are the conditions for constructive interference between several pulses, each of which in isolation would indeed be a white light continuum, without detailed spectral structure.

An instructive picture envisions the fs laser as operating with a light “bullet” running around inside. The laser has laser oscillation possibilities on any of many modes, which satisfy phase-stationary boundary conditions on the cavity mirrors. We form a pulse envelope by adding together many such waves to form a packet, basically a laser carrier wave with an envelope function. But now something new happens. First of all, the ultrafast mode-locking mechanism associated with the traveling light “bullet” basically phase locks all participating laser modes within the relevant spectral bandwidth. The localized light bullet will travel with the system’s group velocity, which is not in general equal to the phase velocity. This leads to a pulse-pulse phase slippage effect. Every time we have the laser pulse impacting the mirror and launching another output pulse, it has already traveled an extra meter or two and developed a larger position/phase offset between the “carrier” wave and the envelope function’s peak. Thinking about the spectral consequences, we have to agree the sampled carrier wave’s phase has evolved more than we estimated from the cavity round-trip travel time. Effectively the output frequencies of the comb are spectrally-displaced from the harmonic positions relative to the repetition rate by this extra phase slippage. Thus we have an expression for the comb frequencies as follows\textsuperscript{10}:

\[ f_n = nf_{rep} + f_{ceo} \quad , \]

where \( f_{ceo} \) is called the carrier phase-envelope offset frequency. It is the carrier-envelope offset phase step per transit, \( \delta \), normalized as a fraction of a cycle - times the number of transits per second, \( f_{rep} \), i.e.

\[ 2\pi f_{ceo} = f_{rep} \delta \quad . \]
Making Frequency Measurements with an Octave-Spanning Comb

Using phase-chirped mirrors, it is feasible to build a Ti:Sapphire laser without dispersion-compensating prisms, leading to an ~1 GHz repetition rate\textsuperscript{11}. The octave-spanning Fiber-generated comb thus has ~0.5 million comb lines of ~50 nW each. Heterodyne with an Nd:YAG reference laser, and separately with its 532 nm second harmonic (used for the Iodine-stabilization), leads to rf beats of >40 dB observed S/N ratios (300 kHz BW). This is just sufficient to allow basically error-free direct counting, without use of a phase-tracking oscillator. As may be seen from Eq. 1, the two beat frequencies will each contain \( f_{\text{rep}} \) and \( f_{\text{eco}} \), but with 2-fold different coefficients for \( f_{\text{rep}} \). We find it helpful to form the control signals by differencing these two beats, thus relating \( f_{\text{rep}} \) to the Nd:YAG optical reference frequency. Similarly, dividing the green beat signal frequency by 2 and subtracting it from the IR beat frequency yields \( f_{\text{eco}} \), independently. The optimal use of these control signals to stabilize the Ti:Sapphire laser typically involves a PZT-based change of the laser cavity length for setting \( f_{\text{rep}} \) and rapid adjustment of the laser's pump power via an AOM for control of \( f_{\text{eco}} \). With some effort the phase noise in these locking loops is so small (\(< \leqslant 1 \) rad) that the full frequency stability of the optical reference laser system is transferred to the comb\textsuperscript{12}. Using these and related schemes, the frequencies of three well-evolved optical standards were measured, as shown in Fig. 3.

**FIGURE 3.** Early Comb-based Frequency Measurements. a) Measured optical frequency of the I\(_2\)-stabilized Nd:YAG reference laser, shown as an offset from CIPM-recommended value; b) Absolute frequency of the I\(_2\)-stabilized HeNe laser at 633 nm, before and after an international intercomparison; c) Frequency measurements of two JILA systems based on the Rb two-photon line at 778 nm, F,G,H and the J data are for the two JILA systems; d) Histogram of frequency measurements F, G, H.
Of course many groups are now working in the optical frequency standards field, and the rate of progress has been remarkable. Table I shows many of the results, from the first high accuracy frequency measurement of Ca, made with a traditional coherent multiplication chain in 1996. Comb-based measurements also began in 1996, with microwave modulators. The floodgates opened in 2000 as many groups acquired stabilized fs lasers and MicroStructured Fibers for the comb bandwidth expansion.

<table>
<thead>
<tr>
<th>Quantum Absorber</th>
<th>Clock Wavelength</th>
<th>Author/Group</th>
<th>Reference / Date</th>
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<td>Ca</td>
<td>657 nm</td>
<td>Schnatz – PTB</td>
<td>PRL 1 Jan '96</td>
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<tr>
<td>C2H2</td>
<td>1500 nm</td>
<td>Nakagawa – NRLM</td>
<td>JOSA B Dec. '96</td>
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<tr>
<td>Sr</td>
<td>674 nm</td>
<td>Bernard – NRC</td>
<td>PRL 19 Apr '99</td>
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<td>H</td>
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<td>Reichert – MPQ</td>
<td>PRL 10 Apr. '00</td>
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<td>D. Jones – JILA</td>
<td>PRL 28 Apr. '00</td>
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<td>PRA 7 July '00</td>
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<td>I2</td>
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<td>PRL 30 Oct. '00</td>
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<td>In+</td>
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<td>v. Zanthier – MPQ</td>
<td>Opt. Lett. 1 Dec. '00</td>
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<td>Stenger – PTB</td>
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<td>PRL 28 May '01</td>
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<td>Opt. Lett. 15 Oct. '01</td>
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<tr>
<td>I2</td>
<td>514.7 nm</td>
<td>R. Jones – JILA</td>
<td>Appl. Phys. B Apr '02</td>
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COMPARING DIFFERENT FREQUENCIES AND IN DIFFERENT PLACES

Beginning with a fs comb system well-stabilized to a stable optical reference, one has the prospect to measure the optical frequency of any other accessible candidate reference system. The comb covers an octave and made it possible for a NIST group to compare their Ca and Hg+ optical standards with their H-maser. This was most instructive in that the optical beats had noise levels vastly less than the repetition rate signal had compared with the H-maser.13. To broaden the intercomparison further, light at 1.06 μm from our I2-stabilized system was sent to NIST by optical fiber (the Boulder Regional Area Network). After the 3.4 km trip, a portion was frequency offset and returned to JILA in the same fiber. The results14 are shown in Fig. 4 where a ~2 kHz broadening can be seen. Using our fiber noise canceller concept, one can find basically perfect restoration of the phase-stable signal back at JILA after the double trip. Next, the measured phase error was divided by two so that the phase-stable cancellation would be produced over in the NIST location. To measure accurate optical frequencies using our comb at JILA, a 1.3 μm source with 1 GHz modulation derived from the H-maser signal relayed UTC-NIST to us using another fiber strand from NIST. At the same time our 1.06 μm light from the JILA-W standard could be measured at NIST using their comb system. Fig. 5 shows these Allan deviation results.
**FIGURE 4.** Optical Frequency Noise Generated by Fiber Transit from JILA to NIST and Back. In the uncompensated case, about 2 kHz linewidth is added by the fiber’s perturbations. Narrow feature shows the resolution-limited optical carrier recovered with the aid of the active noise-cancellation system. High resolution in inset shows bright optical line of 0.05 Hz width, still resolution limited. We estimate national coverage could be obtained with about a dozen phase regeneration stations.

In Fig. 5 the top trace shows the Cs clock specification, while the next trace shows the performance we observed. The third trace shows measurements of the repetition rate of the Iodine-stabilized fs comb, compared with the NIST H-maser, while the next (extended curve) shows the NIST maser noise model. The close agreement of noise model and laser-based source data shows the maser is making the dominant noise contribution. This is convincingly confirmed by the two bottom curves which show the stability of the beat between two I$_2$-stabilized systems at JILA (filled squares), while the overlapping trace (open circles) shows the optical frequency stability of the JILA-W IR light, received at NIST via the compensated fiber link, as measured against the NIST comb stabilized with the high stability laser reference$^{15}$ developed for the Hg$^+$ clock studies. The clear conclusion is that the optical sources are more stable than the “gold standard” for RF stability, the H-Maser. A second clear conclusion is that the transfer of stability to the comb systems is better than the stability of the JILA-W reference laser. NIST experiments using a local Cs standard push this comparison further, to $\sim 3 \times 10^{-15}$ at 1s. Still, at longer times, the relative immaturity of the optical systems becomes evident: averaging times $\sim 10^3$ or $10^4$ s are adequate to allow some drift processes to become apparent. Commercial Cs, on the other hand, is still improving as $1/(\sqrt{\tau})$ at 1 week!
FIGURE 5. Comparing Frequency Stability of Optical and Microwave Sources. A fs laser comb at JILA was stabilized to the JILA-W I$_2$-stabilized optical reference. The specs of a local Cs standard are the top line, the next being the fs laser repetition rate, compared to it. Third trace is comparison with H-maser standard, relayed to JILA by optical fiber, compared to NIST maser noise model (extended, fourth curve). Two lowest curves show optical frequency stability, between two JILA I$_2$-stabilized systems (filled squares) and against the NIST comb, based on the ultrastable laser source developed at NIST for the Hg$^+$ measurements. The close agreement of the last two measurements shows the full laser stability is transferable to the combs.

Long-Term Frequency Measurements (Reproducibility)

In Fig. 6 we show the measured frequency of JILA-W over the several years of the “comb era.” The most significant feature is the complete absence of long-term drift. Data after day 700 are quieter because they are referenced to the NIST H-maser. Considering the H-maser performance shown in Fig. 5, in one day something like 50 measurements could be made, which could in principle bring our results solidly into the $10^{-15}$ domain. Before making a week of such continuous measurements, several technical issues need to be addressed, including automation of the offset corrections of JILA-W, and compensation of the propagation-time variations in the 1 GHz fiber-based reference pulse train. Further, a recent study$^{16}$ of various frequency standards systems based on intracavity spectroscopy leads us to believe that major performance improvements are possible in principle. We considered intracavity spectroscopy using the NICE-OHMS dual modulation concept, with full attention to saturation and time-of-flight broadening, cavity contrast enhancement, optimum finesse and input power, detection shot noise and a number of other concerns. A real system also needs attention to the required gas pressure stability. The reference system for comparison is our JILA-(West) spectrometer which uses a 1.3 m Iodine cell, probed by modulation transfer spectroscopy. This works well, as may be seen from Fig. 6.
FIGURE 6. Long-Term Frequency Stability of JILA-W I₂-stabilized Optical Standard. Frequencies shown are offset from CIPM-adopted frequency. After day 700 measurements were made using a new 375 MHz fs laser and a fiber of different design. Noise reduction is mainly due to reference to the NIST's H-Maser.

POTENTIAL IMPROVEMENTS OF OPTICAL STANDARDS

One major frustration with present cell-based optical standards is that, while the stability is good at short times, drifts of various systematic offsets compromise the long-term stability and reproducibility. One major cause is the usual presence of undesired amplitude modulation produced synchronously with the desired optical phase modulation used for locking. It has finally been possible to build modulators devoid of wavelength-based interference ripples, coupled mechanical resonances, synchronous spatial position modulation and, most recently, synchronous angular modulation of the output beam. Assuming a phase modulation of 1 radian is needed, the Residual Amplitude Modulation can now be reduced well below 1 ppm by trimming out the “Electro-Optic Prism Deflector Effect” arising from non-uniform RF modulation fields across the optical aperture. For our usual 532 nm I₂ line, the implied drift due to this cause would now be below 1 Hz/30 minutes.

More compact and simple gas cell systems suggest themselves. For example the 514.6 nm I₂ transition, already recognized as a wavelength standard, has a ~100 kHz linewidth, ~5-fold reduced relative to that of the usual 532 nm line. Good signals have been obtained with a frequency-doubled Yb:YAG laser. The second-order Doppler shift of these lines is ~5 x10⁻¹³ and so will provide a small and sufficiently stable offset. Going to the narrower lines offered by molecular overtone transitions, some work has been done with CH₄ and especially with C₂H₂, where overtone resonances at 1.6 μm, 1.03 μm, and 790 nm have been observed with cavity-enhanced sensitivities. For long-lived absorbers, saturation-based velocity-selection will shift the effective transverse temperature to a lower value and probably will require active control.

A clear direction for improvement is the use of narrower transitions, as was suggested “at the beginning” by Professor Dehmelt. Still, some of us would like to use many responsive quantum systems – not just one trapped ion. So we come to laser-
cooled Group II metals, including Mg, Ca, and Sr. Spectacular results have recently been obtained by the PTB group\textsuperscript{17} by two-stage cooling of Ca to a velocity $<0.5$ cm/s. At this level the very troublesome influence of local curvature of the interrogating fields is basically eliminated. It still is a challenge to produce the 4 Ramsey pulses with stable, known relative phase shifts, although a measurement protocol involving several reversals can trade stability for reproducibility. At JILA we are looking at the attractive prospects of $^{87}$Sr, where the ultimate clock will be on the $^{1}$S$_{0}$ $\rightarrow$ $^{3}$P$_{0}$ doubly-forbidden transition (lifetime $\sim$100 s). For cooling also, $^{87}$Sr is attractive as its associated hyperfine structure leads to a form of Polarization Gradient cooling, even on the strongly allowed $^{1}$S$_{0}$ $\rightarrow$ $^{1}$P$_{1}$ transition at 461 nm. Temperatures below 300 $\mu$K have been obtained\textsuperscript{18}. The next step will be capture and Raman cooling in a Far Off-Resonant Optical Trap, formed with the magic wavelength\textsuperscript{19} ($\sim$800 nm) for which optical Stark shifts of ground and excited clock states would be equal. The $\gg$ kHz vibrational sideband structure should be well-resolved against the super-narrow linewidth. If atom collection efficiency needed to be improved, one could instead employ second-stage cooling\textsuperscript{20} on the $^{1}$S$_{0}$ $\rightarrow$ $^{3}$P$_{1}$ intercombination line. Using $^{88}$Sr instead of $^{87}$Sr would be problematical in that, without the hyperfine-induced mixing, the $^{1}$S$_{0}$ $\rightarrow$ $^{3}$P$_{0}$ transition will be exceedingly weak. Of course, the $^{1}$S$_{0}$ $\rightarrow$ $^{3}$P$_{2}$ transition could also be used in $^{88}$Sr, with the possibility of closed-cycle optical cooling upward to $^{3}$D$_{3}$ from $^{3}$P$_{2}$. We prefer to work first on the magnetic field-insensitive J=0 case.

**TIME DOMAIN ISSUES AND APPLICATIONS**

While frequency standards offer ideally-stable sinewave outputs, clocks produce trains of narrow pulses with jitter-free inter-pulse intervals. In fact the present competition for the best free-running source is represented by Cryogenically-cooled Sapphire Resonator microwave frequency sources and by free-running fs self-mode-locked Ti:Sapphire lasers. After the vibration problems have been suppressed, both sources offer short-term stability in the $\sim 1 \times 10^{-15}$ or better domain at 1 s. With one control loop, the fs laser can have its carrier-envelope phase stabilized, so that its coherence extends from the RF range, represented by the pulse repetition rate and its harmonics, up through the optical region where the comb structure is also manifest. In this attractive case, if any single comb line is stabilized, for example to the sharp resonance absorption of a trapped ion's spectrum, the entire comb becomes stable. A corollary is that the time-domain characteristics, particularly the pulse-pulse time intervals, become correspondingly stable. Basically ideal delta-function pulses in time are delivered with a delta-function repetition rate spectrum!

The interesting capability to precisely transfer time-domain stability has recently been demonstrated by Ye et al.\textsuperscript{21}, whereby one fs laser has been stabilized to a reference fs laser such that the relative jitter between their output pulses is at the sub-fs scale (1 ms averaging). Since the timing control is implemented by frequency-domain electronic phase comparison, a controlled deliberate phase shift manifests itself as a temporal delay of the controlled laser's pulses. Nothing requires the two lasers to have similar wavelengths but, if they do have a spectral overlap, the optical heterodyne beats can be used to phase-lock the two lasers' optical frequency combs together. The
strict synchronization insures the equivalence of the comb spacing of the two comb systems. Thus by simple laser beam combination optics, one produces a synthetic fs source with bandwidth, spectrum, and peak power characteristics unattainable from a single laser.

An interesting application of two-laser synchronization is spectrally-resolved Coherent AntiStokes Raman-Scattering Microscopy, (CARS Microscopy) where the two pulse systems differ in their average optical frequency by an amount chosen to excite a certain molecular vibration, with the resulting up-shifted light thus giving rise to a species-labeled image\textsuperscript{22}. This application requires only the secure synchronization via repetition-rate locking. On the other hand, generation of a coherent comb in the near IR by Difference Frequency Generation in a suitable nonlinear crystal requires optical phase-locking as well\textsuperscript{23}. The extreme ratio of peak-to-average power makes a wide variety of nonlinear mixing processes readily available. As a general summary, we can expect to see further rapid and fundamental progress in related fields as these new frequency-metrology tools are applied more widely. In view of the now-easy communication between RF and optical domains, we also can expect individual advances to have a broader applicability than was heretofore possible. If the "explosion" of progress in the last four years is the guide, we can look forward to a remarkable future.

ACKNOWLEDGMENTS

Professor L.-S. Ma is thanked for his many contributions to this work in its earlier stages. This research is supported in part by NASA and by NSF, while NIST provides continuing support for research in the possible new realizations of the fundamental standards.

REFERENCES