Optical Frequency Standards : Progress and Applications

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A number of individual small advances, taken together, bring us to the beginning of a new domain of optical frequency stability, reproducibility, and measurement. Particular advances include the refinement of Noise-Immune Cavity-Enhanced Optical Heterodyne Molecular Spectroscopy (NICE-OHMS), especially by active control of the residual AM produced by a phase modulator, and by improvement of the PZT's HV amplifier noise and stability. Replacing a mirror of a Kurogi comb generator by a selective output filter provides a frequency-displacement capability that will be very helpful in realizing a "Divide and Conquer" optical frequency synthesis chain.

Optical frequency standards will offer us the highest resonance line Quality Factor, and so highest potential stability of frequency, making optical sources the interesting ones for atomic clocks and precision (null) tests of various fundamental physical principles. One thinks here of the symmetrization postulate, and the spectra of simple atomic systems (hydrogen, positronium, muonium … .) Helium calculations also approach this domain of accuracy. A new QED test, taking its place beside Lamb Shifts, is the predicted birefringence of the vacuum which could be induced by powerful magnets, such as the ones which have abruptly become available from the SSC. Stable frequency sources are ideal for study and measurement of important small effects, such as those induced by Parity Non-Conservation, or by Chiral Molecular structure, or even by gravitational shifts in the earth's field. The high precision makes possible interesting null tests, as in the anisotropy of space, breakdown of Local Lorentz invariance, or postulated interactions which might affect the Equivalence Principle, or its derivatives. As the stable laser technology becomes more robust and widely practiced, it also often offers the ideal channel for measurement of some particular physical quantity of interest, such as an atomic natural decay width or coherent motion of an ensemble of laser-cooled atoms.

Of course the precision available in the measurement would be degraded if all technical problems were not simultaneously solved, at some level. For example, we need first to be able to observe the narrow atomic resonance by itself, separated from the inevitable background. Tricks and concepts here are well known, and include modulation, especially making use of the first, second and third derivatives of the system's response. One can chop the saturating beam to identify the nonlinear

Doppler-free resonant term, or direct the modulated beam anti-parallel to the detection beam, relying on the molecular response to provide the sub-Doppler modulation transfer signal[1].

With an isolated resonance for locking the laser, it is easy to show that

$$\Delta\nu/\nu \sim (1/Q) \bullet (1/ (S/N)) \bullet 1/ \sqrt{\tau} \quad . \qquad \text{Eq. 1}$$

This shows the high premium placed on good S/N: it gives us better stability at some averaging time τ. Perhaps more important, it speeds up our checking of the systematic shifts -- which are always changing a little -- and so enables us to obtain better independent reproducibility, which could map into better accuracy capability if we are able to make accurate connection with the SI unit of frequency, at 9.2 .... GHz. Experience shows that the resonance linewidth may be meaningfully divided into 10 or even 100 sub-elements, if one can invest an afternoon in the experiment, and has suitable apparatus and experience. Getting to 3 $\times 10^{-4}$ is a 3 months investment for a skilled worker, while the 30^+ years invested by the Cs clock community has brought them to $1{:}10^6$ linesplitting accuracy. So narrow lines with high S/N are good! The paper by Ye et al. in these proceedings[2] documents the dramatic recent increase in sensitivity, to 5 $\times 10^{-13}$ integrated absorption. This has made possible very good locking performance even using a weak absorber such as overtone resonances in HCCD. Below we compare the performance with other schemes. Following these general introductory remarks, we turn to a few small, but important technical advances.

As explained elsewhere[3], the new high sensitivity method of NICE-OHMS relies on a high finesse cavity to increase the resonance's contrast, FM detection for approaching the quantum-limited sensitivity, and the accurate matching of the FM sidebands to the cavity's FSR to avoid discriminating the laser's residual frequency noise into the signal channel. With the development of the active lock of a Voltage-Controlled Oscillator to the cavity fsr (this rf source provides the FM sideband frequency), the noise associated with laser frequency-locking errors is effectively supressed. But the balance of the two FM sidebands at ±319 MHz is still critical to the locking accuracy. One initially might suppose that any baseline offset problem associated with the FM modulator could be suppressed by a dither of the cavity length, followed by 2-nd derivative signal recovery of the rf balanced mixer signal output. While this is indeed true, another issue concerns the recovered *lineshape* under the conditions of FM with a Residual AM part (RAM). The FM dispersion signal at line center, useful for locking the laser, has no even-symmetric absorption component: the two absorption components at line center are exactly cancelled when the modulation is pure FM. In the presence of AM, however, an even-symmetric absorption-phase signal does exist at line center. So the lineshape is changed by the AM contamination, and this causes a frequency shift, even with the cavity dither method.

Active control of residual AM was explored earlier[4] with reasonable results, but the ring-gap resonator design used for the 320 MHz modulation does not lend

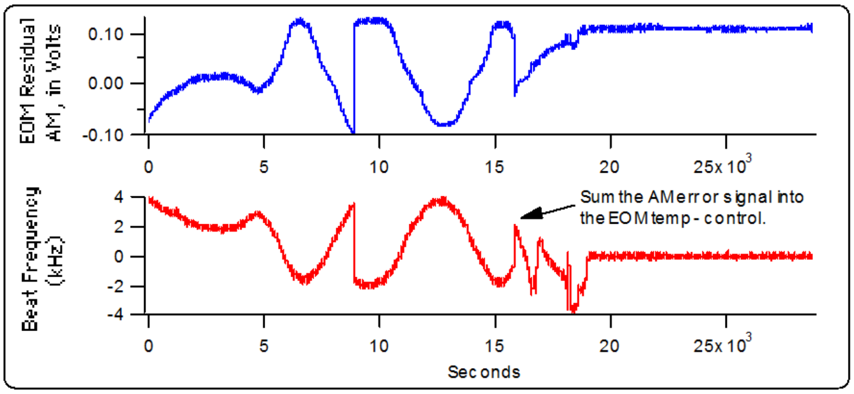

Fig. 1. Stabilization of Residual AM produced by Phase Modulator. RAM is caused in part by residual interference of waves reflected from the faces. Temperature control affects the optical index and so the RAM. No electronic integrator was used in this test, so output RAM is stable, but not zero.

itself immediately to applying a dc voltage onto the crystal. Instead we are using the temperature-dependent index of refraction to tune the interference between the two reflections on the modulator crystal's faces. The transmitted, modulated beam is sampled with a beamsplitter and an auxiliary rf detector/balanced mixer system to measure the fractional spurious AM which is synchronous with the desired ΦM. The resulting dc output changes sign at zero Residual AM, and so is filtered and used as an offset in a thermal control loop based on Peltier cooling of the Ring Resonator and its $LiTaO_3$ modulator crystal. Fig. 1 shows the improvement.

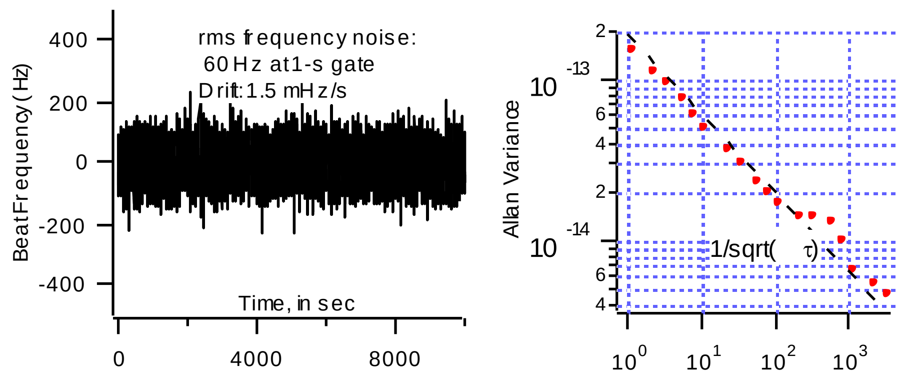

Figure 2. Stability of beat between I_2 stabilized and HCCD-stabilized lasers. The 532 nm-stabilized laser has a stability ~5 x10^{-14} at 1 s, from beating experiments with two I_2-stabilized systems.

We improved the HCCD cavity's HV PZT driver amplifier, providing 50 µV rms noise in 60 Hz control BW, with a 1.2 kV dynamic range. With these improvements, the long-term stability achievable with absolutely-stabilized visible sources is unprecedented. Remarkably, it is similar for the I_2 and HCCD stabilization resonances, in spite of the nearly 10^6 x ratio of absorption coefficients. See Fig. 2.

Perhaps one can do much better with I_2 … ? For example, the line excited by the 501.7 nm Ar^+ laser has a lifetime rumored to be > 25µs, with a corresponding potential linewidth of ~12 kHz. Following the modern trend to solid-state laser technology, it would be interesting to try Iodine, NH_3, HCCH, and HCCD with a frequency-doubled 1.03 µm Yb:YAG, pumped of course with a diode laser !

In the direction of 'much better, but still simple,' one has to notice laser-trapped atomic samples, since the density is near optimum for sharp resonances, and the milliK temperature guarantees second-order Doppler shift ~10^{-17} or below. Calcium and Strontium recommend themselves because of the narrow intercombination "clock" transitions. We are pursuing Sr because our modelling suggests we may be able to enjoy a "second-stage" of cooling and trapping, based on this E1 intercombination line (width ~ 7 kHz.) This would be interesting because the recoil steps would be far larger than the Doppler cooling limit: discreteness of the cooling process would surely be evident. At present our simple trap can capture ~ 5 x 10^6 Sr atoms, which can be increased to 10^8 Sr atoms by use of diode lasers at 707 nm to repump $^3P_2$ and at 679 nm to repump atoms lost into $^3P_0$. Both connect to $^3S_1$. We find a nice density increase, 20x, but also see clear evidence of enhanced cold-atom collision processes involving a groundstate Sr interacting with an excited $^1P_1$ Sr atom. A possible target for this work is a "fountain" interaction on the M2 transition at 671.2 nm connecting to the $^3P_2$ state. At NIST in Boulder, Hollberg's group is already measuring Ramsey resonances in their Ca trap. Of course the PTB group deserves everyone's enthusiastic praise for their pioneering Ca measurements[5]!

Even being able to prove that one has a highly stable and reproducible optical source is not quite enough: what *IS* actually the absolute frequency of this stable laser? There has been a huge amount of work on the frequency synthesis/measure–ment problem over the years, beginning with work at standards labs such as NBS, NPL, etc, even in the late 1960's epoch. Finally it was possible to directly measure 88 THz, the frequency of the 3.39 µm CH_4-stabilized HeNe laser. Following Chebotayev's suggestion, Ne atoms were used by the NBS team as the nonlinear frequency mixer to reach the HeNe red line in 1983, and in 1984 it was possible to redefine the meter in terms of the speed of light. Subsequently there are many, many schemes for using nonlinear optics to synthesize optical frequencies. As noted above, a recent spectacular triumph is the work at PTB to measure the Ca 657 nm intercombination line, with an inaccuracy below 1 kHz!

A powerful new concept was introduced by Telle, Hänsch and Meschede[6] in which one could phase-lock a laser to the arithmetic average frequency of two reference lasers. Iteration of this "frequency-interval bisection" method, also called the "Divide and Conquer" method, leads from optical frequencies to microwave ones in some 13 or 14 stages. A beautiful and tremendously promising simplification arises from the work of Kurogi, Nakagawa, and Ohtsu (their Optical Frequency Comb Generator[7]), which allows the microwave/optical difference frequency connection to be in the few Thz range. Now the number of stages is 8 or fewer, particularly with the wider comb widths reported by Brothers et al.[8] and by Kurogi et al.[9]

A broadband optical frequency comb generator consists of an efficient electro-optic phase modulator within an optical cavity. With appropriate choice of cavity length, as shown by Kurogi et al., an optical comb extending over several TeraHertz is produced. This apparatus was improved[10] by Ye, Ma and Hall who added a closely-spaced additional mirror on the output side to resonantly out-couple a selected order of sideband. This modification increases the strength of the selected spectral comb component by several orders of magnitude. This makes possible simple and unambiguous beat frequency measurements using the ~20 dB spectral purity of the output-coupled selected comb line. We show the measured single frequency shifted output vs shift frequency in Fig 3.

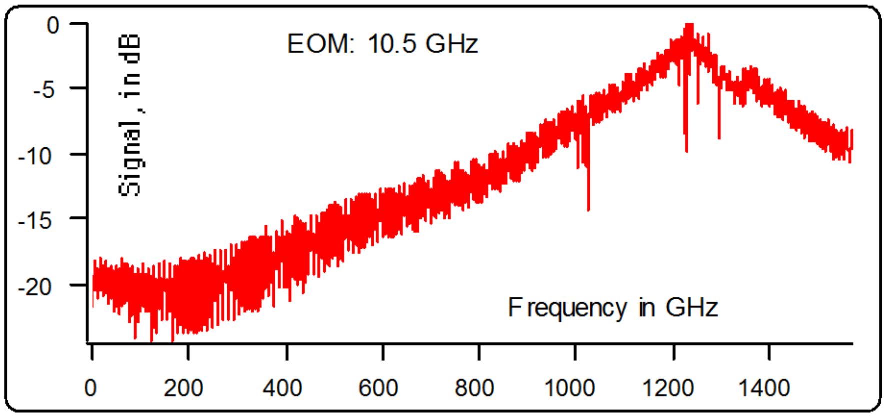

Fig 3. Successive single frequency outputs as output filter is tuned. Input frequency is at ~1250 Ghz on this axis. Downward spikes arise from unlock events.

It is exciting to consider the possible near-term reality of a "Divide and Conquer" frequency synthesis from Microwaves to the visible: with due attention to dispersion compensation using techniques known in the femptosecond laser field, the useful spectral width of the associated frequency-displacement operation then becomes limited only by optical attenuation within the modulator cavity. It seems sure that a 10% shift will be reported by the next Conference! More light can be admitted to the cavity using a filter-type input mirror, as demonstrated by Macfarlane, Bell, Riis, and Ferguson[11]. We can enter with tens of milliWatts, and could live with tens of picoWatts in the shifted beam. With the increasing availability of the "designer" nonlinear optical material, Periodically-Poled Lithium Niobate, only 4 auxiliary diode lasers would be needed with that dream modulator to reach the stabilized Nd:YAG laser at 1.064 µm from a microwave source!

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