The evolution of the Frequency Standards and Metrology symposium and its physics

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Abstract. In this paper I recall some of the earlier developments in the physics and measurement of atomic frequency standards and clocks, as revealed in presentations at past Frequency Standards and Metrology (FSM) symposia in this series. Coverage is not meant to be comprehensive; unfortunately, I can select only a relatively small number of representative examples of work that I felt stimulated many of the major areas of interest since the first meeting in 1971.

1. Introduction

Although I didn't attend the first Symposium on Frequency Standards and Metrology[1], I have been fortunate to attend all subsequent symposia, starting in 1976. I very much like this meeting, in part because it is held relatively infrequently and each new installment brings significant advances and renewed excitement in the field. This was the intention of the founders of the meeting, Claude Audoin (LHA), Helmut Hellwig (NBS), and Jacques Vanier (Laval University). It is interesting to highlight some of these advances; here, I will focus on the physics of high-performance frequency standards.

In recalling past conferences, I was struck by the fact that many of today's current high-interest topics have been with us for a long time, of course with dramatic advances. Also, some overriding themes have persisted such as efforts to: reduce first- and second-order Doppler shifts; increase resolution and Q through velocity selection, confinement, and cooling; improve the spectral purity of probe oscillators; reduce noise to the limits allowed by quantum mechanics; connect a broad range of frequencies in a coherent way; and search for new physics with precision frequency comparisons. I will attempt to highlight these topics with some illustrative examples taken from the earlier symposia; more recent results are much better covered in these proceedings. This approach is inherently flawed because it doesn't give proper credit to all the groups working on these subjects; moreover, it suffers from my inherently personal (and probably too-narrow) view. For this, I apologize.

[1] The first meeting was actually called a "Seminar."

2. 1971, University of Laval, Quebec; Chair, Jacques Vanier

Not surprisingly, there were a number of reports on Cs beam standards and H masers in this first meeting. For Cs beams, Dr. Gerhard Becker (PTB) set the benchmark, reporting $\sigma_y(\tau) \cong 3 \times 10^{-12} \tau^{-1/2}$ and an inaccuracy of 4×10^{-13}. This standard had a Ramsey cavity length L = 0.8 m and also featured hexapole state selection and detection magnets, in order to focus the beam and mitigate uncertainties from the distributed cavity phase shift. Interestingly, distributed cavity phase shifts have continued to be a concern even today. Hydrogen masers featured prominently with reports from NRC, Goddard, Harvard, Orsay, Laval, NPL, and SAO. Although the difficulty of detecting hydrogen atoms was well known, an interesting report was given by Harry Peters (Goddard), who described a hydrogen beam magnetic resonance apparatus, where detection downstream was provided by a Penning ion gauge.

One talk (J. DePrins, University of Brussels) described results with an ammonia beam maser. I found this interesting given the historical significance of ammonia being the first maser/laser type device, but it was the only talk on ammonia masers at FSM symposia. An interesting report was presented by Hans Schuessler (Texas A&M) on trapped ion spectroscopy. This was not long after the experiments he carried out on $^3He^+$ hyperfine spectroscopy (~ 8.67 GHz) in the group of Hans Dehmelt (University of Washington) [1]. This was a rather heroic experiment in which the ions were polarized through spin-exchange with a polarized Cs beam and detected through state-dependent charge transfer ($^3He^+ + Cs \rightarrow$ $^3He + Cs^+$) in conjunction with measuring the change in $^3He^+$ ion number by sensing ion currents in the trap electrodes. Along with the H-maser, this experiment showed a key advantage of confinement - that the atoms' velocity and first-order Doppler shift can average to zero. In this experiment however, the ions were rather hot, which limited the accuracy to about 1 part in 10^9 from the second-order Doppler or time-dilation shift.

Today, given the recent dramatic advances with optical clocks, some of our younger colleagues might think that the subject of optical clocks is fairly new. However, already by 1971, this area was being actively pursued. One report was presented by Dick Barger describing his work with Jan Hall [2] on stabilization of a standing wave, HeNe laser at 3.39 μm by saturated absorption of CH_4 contained in an intra-cavity cell. Here also the first-order Doppler shift is essentially eliminated because the saturated absorption feature is due to both forward and backward traveling waves interacting with the same group of atoms whose velocity along the axis of the laser cavity is zero. This particular work was significant because the reproducibility of the frequency was at the level of 10^{-11}, two orders of magnitude better than the wavelength standard based on a 606 nm line in Kr, which was limited in accuracy because the lineshape was asymmetric. If frequency measurements could be extended to the visible, then the wavelength standard could be eliminated by defining the speed of light. Other talks on stable lasers included those by Charles Freed, Tony Siegman, Helmut Hellwig, and Shaoul Ezekiel.

An interesting talk was given by Judah Levine who reported a strain meter based on a 30 m reference cavity (at 3.39 μm) where the mirrors were mounted to bedrock in an abandoned mine near Boulder, Colorado. One goal of the project was to measure earth vibrations and tides but the device was also sensitive to strains induced by underground nuclear explosions. Data was shown for such an event from the underground test site in Nevada. The signature of these signals could be distinguished from naturally occurring earthquakes, an important factor in insuring the viability of the future nuclear test ban treaty. The possible application to detection of gravity waves was also noted, and although the sensitivity was well below that of today's gravitational wave detectors, the required technology was already being developed in these early experiments.

3. 1976 Copper Mountain, Colorado; chair: Helmut Hellwig

Cesium standard inaccuracy had steadily improved (to around 10^{-13}) and there were several reports on advances in hydrogen masers. One of the most interesting of these was a preliminary report by Bob Vessot on the results of a sub-orbital rocket flight ("Gravity Probe A") that carried a maser [3]. During the approximately 2 hour, 10,000 km high flight, the frequency of this maser was compared to one on the earth's surface. Because of the different time dependences of the relativistic frequency shifts caused by changes in the gravitational potential and time dilation, both effects could be confirmed with an uncertainty less than 10^{-4}. This was a pioneering experiment showing the viability of carrying precise atomic clocks into space, a topic of ever increasing interest up to this day. Moreover, from a basic physics standpoint, it is still the most accurate direct measurement of the gravitational potential red shift. As a side note, the transponder technique used to determine and correct for the first-order Doppler shift between masers is now commonly used to stabilize the phase between separate sites in optical clock and frequency standard comparisons [4].

Optical frequency standards continued to improve. An interesting effect in saturated-absorption spectroscopy was the observation [5] of the splitting of the feature due to photon recoil [6] where the position of the two lines is governed by the condition where the transverse velocity of either the upper or lower state in the transition is at zero velocity. For many years, Christian Bordé, who has attended these symposia, has been a key person in explaining how the effects of recoil must be accounted for in atomic clocks, at all ranges of frequency. Recoil effects are of course an integral part of the atom interferometer experiments whose dramatic advances are reported at the current Symposium. To increase resolution in saturated absorption experiments, some laboratories started pushing towards laser beams with larger waists and heavier absorbers such as SF_6 and OsO_4, for increased interaction times. In the early 70's, the dye laser was becoming an extremely useful tool in AMO physics; for optical frequency standards, it's wide tunability meant that atomic or molecular reference transitions need not be in near coincidence with an existing fixed frequency laser line and one could now focus on choosing a reference that had particularly low systematic offsets. Ted Hänsch described his early very impressive experiments on the 1S – 2S two-photon Doppler-free transition in hydrogen, using a pulsed dye laser at 486 nm (peak power ~ 50 kW) doubled to 243 nm in a lithium-formate crystal.

Frequency chains were featured by a number of laboratories. Here, through a sequence of lasers of increasing frequency referenced to each other with harmonic mixers, phase coherence could be maintained. At the highest frequencies, metal-on-metal diodes were used but these lost efficiency as one approached the visible range of the spectrum. Therefore, a difficult step was to go from frequencies corresponding to wavelengths of a few micrometers to the visible range. As possible solutions to this problem, Veniamin Chebotayev discussed 4-wave mixing experiments in Ne to sum three infrared laser beams to the visible ($\lambda \cong 0.65$ μm) and also the use of nonlinear crystals (e.g. $LiNbO_3$). In any case, with the Cs to 3.39 μm frequency chain, it was now possible to connect the ^{86}Kr wave length standard to Cesium via a comparison of the ^{86}Kr wavelength to the 3.39 μm wavelength in a shared reference cavity [7]. This led to a measurement of the speed of light to about 3 parts in 10^9 prior to the second Symposium [8] (see also [9]).

Even at this relatively early stage in the Symposium series, the benefits of low temperature reference cavities were demonstrated. Sam Stein presented results of a microwave parametric oscillator whose signal component was stabilized to a Nb reference cavity that had a Q of around 10^{10}, yielding an instability $\sigma_y(100\ \text{s}) \cong 6 \times 10^{-16}$. Of course cold, cavity-stabilized microwave and optical oscillators continue to be intensely studied today.

Cliff Will and Robert Pound gave very interesting lectures proposing various fundamental investigations. These included tests of the equivalence principle for clocks whose frequencies depended in different ways on the fundamental forces, a laboratory test of frame dragging due to a nearby rotating object, space-born detection of gravitational waves, and searches for a time rate of change in the ratios of the fundamental constants. These topics endure today, benefitting from ever-increasing sensitivity. Prof. Pound pointed out the high Q's available in Mössbauer spectroscopy and gave an example of the 92 keV transition in ^{67}Zn whose Q could be measured using one ^{67}Zn sample as the source and another ^{67}Zn sample as the absorber, a direct comparison of two essentially identical samples. Here, the relative frequencies of the two samples could be tuned via the first-order Doppler shift by moving one of the samples relative to the other. The linewidth measured in this way was 0.8 μm/s corresponding to a Q of around 3×10^{14}. As a personal aside, when I was a graduate student at Harvard working on masers in Norman Ramsey's group, Prof. Pound would tease us in a light-hearted way about our meager Q's of only around 10^9. I had always hoped we might compete using atoms, and finally in 2011 our lab at NIST was able to compare the $^1S_0$ – $^3P_0$ transitions of two $^{27}Al^+$ ions in a similar way to Mössbauer spectroscopy and demonstrate a Q of 3.4×10^{16} [10].

For atomic ions at this symposium, Guenter Werth reported on laser optical pumping in Ba^+ ions; however, more interesting to me was that between this and the previous symposium, he and Fouad Major had measured the 40.5 GHz hyperfine transition in $^{199}Hg^+$, where optical pumping and state detection could be accomplished with fluorescence from a $^{202}Hg^+$ resonance lamp [11]. This work was extended by Len Cutler and colleagues at HP and John Prestage, Lute Maleki, and colleagues at JPL, where it has undergone dramatic improvements, continuing today. Hans Dehmelt described his "shelved electron amplifier" detection idea [12] where state-dependent fluorescence on an allowed transition can be used to discriminate between two levels of a clock transition with essentially 100 % efficiency. In this case, noise in the measurement of transitions reduces to the fundamental quantum "projection" noise associated with the fluctuations in the measured state for a superposition of two clock states, the so-called "standard quantum limit" for uncorrelated atoms. Impressively, with the current precise control of technical noise in the clock experiments, the quantum noise can still dominate on even large numbers of atoms ($N \sim 10^6$) where the quantum noise-to-signal ratio scales as $N^{-1/2}$. Dehmelt also described early laser-cooling proposals [13, 14].

4. 1981, Aussois, France; chair: Claude Audoin

Cesium beam standards started appearing where state selection and detection could now be accomplished with compact lasers rather than large magnets. New cavity designs were explored with the goal of further suppressing distributed cavity phase shifts. As inaccuracies became smaller, one now needed to include Stark shifts from blackbody radiation (-1.7×10^{-14} at 300 K); an effect that has become increasingly important over the years for both Cs standards and the precise optical clocks of today. Walter Hardy (UBC) gave a very interesting talk proposing H-masers operated below 1 K where the temperature dependences of collision shifts due to Helium in the gas phase and liquid Helium on the glass bulb would cancel at a certain temperature. Hans Dehmelt described the initial laser cooling experiments on trapped ions by groups at Heidelberg and NIST. The cooling was important for suppressing time dilation, and the Heidelberg group showed the first pictures of individual atoms in a trap [15].

Optical frequency standards continued to advance with the use of significantly expanded laser beams and heavy molecules, to increase interaction times. These improvements exposed the difficulty of dealing with shifts due to wavefront curvature; however, some experiments were now using separated optical beams to observe Ramsey fringes, where loss of signal due to atomic/molecular beam divergence could be suppressed with multiple optical beam paths. Progress on frequency chains had been dramatic since the last Symposium with reports from IEN (E. Bava), NRC (K. M. Baird), NPL (D. J. E. Knight), Novosibirsk (V. Chebotayev) and NBS (K. Evenson). Despite the many stages required in the chains, this was a triumph of frequency metrology since it was now possible to relate optical frequencies to the frequency of Cesium in a phase coherent way.

5. 1988, Ancona, Italy; chair Andrea De Marchi

The Ancona meeting again saw many improvements across all areas – and was particularly memorable for some of us because Andrea De Marchi provided an excellent wine that came from the vinyards of his family. For Cesium, the use of lasers for state preparation and detection was becoming more common place. Bill Phillips reported on "sub-Doppler" laser cooling of Na atoms. This cooling was significantly better than that predicted by Doppler cooling, an effect Bill referred to as "a strong violation of Murphy's law." It wasn't that the simple theory of Doppler cooling was wrong but it applied only to two-level systems. As explained by Claude Cohen-Tannoudji, Steve Chu, and their colleagues, in sub-Doppler cooling, dynamic optical pumping of the ground state hyperfine levels was important, leading to the so-called "Sisyphus effect" cooling and much lower temperatures. The achievement of much lower temperatures than those provided by Doppler cooling was crucial for the enormous improvement in performance of current fountains, to prevent the atomic sample from expanding too much during free fall.

For hydrogen masers, the cryogenic masers proposed by Walter Hardy at the previous symposium had been subsequently built and successfully operated at UBC and SAO. These devices involved wonderful new physics but ultimately for frequency standards applications, the collision shifts proved to be too difficult to control.

In the area of stable lasers, Dieter Hils and Jan Hall reported locking separate HeNe lasers beams (633 nm) to adjacent fringes of the same cavity. By observing the beat frequency between these two beams, a relative linewidth of 30 mHz was observed. Although not providing absolute frequency stability, this experiment showed that state-of-the art electronics was sufficient to achieve extremely narrow laser beam linewidths, providing the optical cavities were stable. Therefore, the problem of providing precise oscillators was transferred to providing stable reference cavities, which of course occupies the efforts of many groups in our field to this day.

A number of advances were made in optical frequency standards. As one example, the Novosibirsk group (S. N. Bagayev, R. P. Chebotayev et al.) described a CH_4 saturated absorption cell for the 3.39 µm He-Ne laser that had a length of about 8 m, was cooled by LN_2, and was probed by a beam waist w_0 = 30 cm, giving a linewidth of 250 Hz. Similar experiments were carried out in Paris and Boulder. As another example, a PTB/NRC team (H. Helmke, J. Ishikawa, F. Riehle) reported a four-travelling-wave Ramsey/Bordé interferometer for the dye-laser excited 657 $^1S_0$ – $^3P_1$ transition in a Calcium beam, achieving a linewidth of around 20 kHz.

For trapped ions, groups at the University of Washington, (H. Dehmelt et al.), Heidelberg (P. Toschek et al.) and NIST (Ion Storage group) had been able to clearly observe quantum jumps between two states, as indicated by the presence or absence of fluorescence from a strong transition that involved one of the states. This demonstrated Dehmelt's electron-shelving amplifier scheme and showed that clock transitions could be detected, limited only by quantum projection noise. Using Dehmelt's detection scheme, Jim Bergquist showed spectra on the $^2S_{1/2} - {}^2D_{5/2}$ clock transition from a single trapped Doppler-cooled $^{198}Hg^+$ ion which consisted of the Doppler-free "recoilless" central feature (optical version of the Mössbauer effect) and first-order Doppler-effect generated motional sidebands. John Bollinger discussed the evaluation of the first laser-cooled clock, based on a ~ 303 MHz transition in $^9Be^+$ ions probed with a 550 s Ramsey time leading to a linewidth under 1 mHz. (Subsequently Peter Fisk (CSIRO) used a 600 s Ramsey time on the 12.6 GHz transition in $^{171}Yb^+$ to achieve a Q of 1.5×10^{13} [16]). I now find it amusing that in his introduction to the trapped ion session, Hans Dehmelt, in discussing a path to 10^{-18} uncertainties had written: "All this is no wonder, because high resolution spectroscopy on anything else but on an individual isolated metastable atomic particle at rest in the laboratory will always be affected by shifts of various kinds." Of course, these days, the neutral atom lattice-clock experiments seem to do quite well overcoming this apparent difficulty.

6. 1995, Woods Hole, Massachusetts, Chair: Jim Bergquist
2001, St. Andrews, Scotland; Chair: Patrick Gill
2008, Asilomar, California; Chair: Lute Maleki

By 1995, the first atomic Cesium fountains by the groups of Steve Chu and Andre Clairon had been demonstrated and at the meeting, Andre Clairon reported a Cesium uncertainty of 3×10^{-15}. Ideas of surpassing the standard quantum limit in signal-to-noise ratio emerged (e.g., spin squeezing), and although the basic effects have subsequently been observed, they are yet to be incorporated in a high precision clock. M. Kourogi presented his work on "Optical Frequency Comb Generation," in which an intercavity phase modulator is modulated at the free spectral range of the cavity to dramatically increase the range of sidebands (to ~ 7.5 THz at ~ 785 nm). Although impressive, this was to be dramatically superseded by the advent of optical combs.

The last two meetings brought us close to where we are today. Cs (and Rb) fountains were operated at the projection noise limit reaching inaccuracies of ~ 10^{-15}. For optical frequency standards, Ted Hänsch, Jan Hall, and their colleagues had achieved optical combs that spanned an octave. This now meant that optical frequencies could be tied to microwave frequencies in a single step - the "frequency chain" had been simplified in dramatic fashion and within a year or so of the initial demonstrations, many labs were able to build and use these devices to make precise measurements of optical frequencies. We've seen a gradual but strong shift to optical clocks based on trapped atoms. The development of laser-cooled neutral atom clocks (Sr, Ca, Yb, Hg...) where the atoms are trapped in optical-dipole-force lattices, strongly suppresses all Doppler shifts. For ions, a number of candidates are being studied including In^+, Yb^+, Sr^+, Ca^+, Hg^+, and Al^+. Stable reference cavities are now operated at low temperatures and use crystalline materials such as silicon. Interestingly, mirror substrates and coatings cause significant noise which has led to the development of crystalline coatings. The state-of-the-art for optical clocks and frequency standards has recently been reviewed in [17].

Many other topics including searches for time variation of the fundamental constants, tests of Local Lorentz invariance and the equivalence principle are actively pursued. The performance of atom interferometers has increased dramatically in very large and small sizes. Advanced clocks are scheduled for space, and long-distance frequency transfer over fibers is now possible. Miniature clocks based on MEMS technology have found interesting applications. From the level of interest and excitement at the 2015 Symposium, we can look forward to an equally interesting next Symposium.

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References

[1] Schuessler H A, Fortson E N and Dehmelt H G 1969 *Phys. Rev.* A**187** 5
[2] Barger R L and Hall J L 1969 *Phys. Rev. Lett.* **22** 4
[3] Vessot R F C, Levine M W, Mattison E M, Blomberg E L, Hoffman T E, Nystrom G U, Farrel B F, Decher R, Eby P B, Baugher D R, Watts J W, Teuber D L and Wills F D 1980 *Phys. Rev. Lett.* **45**, 2081
[4] Bergquist J C, Itano W M and Wineland D J 1994 in *Frontiiers in Laser Spectroscopy, Proc. Int. School of Physics <<Enrico Fermi>>* CXX, ed. by T. W. Hänsch and M. Inguscio (North Holland, 1994) p. 359
[5] Hall J L, Bordé C J and Uehara K 1976 *Phys. Rev. Lett.* **37**, 1339
[6] Kol'chenko A P, Rautian S G and Sokolovskii R I 1968 *Sov. Phys. JETP* **28**, 986
[7] Barger R L and Hall J L 1973 *Appl. Phys. Lett.* **22** 196
[8] Evenson K M, Wells J S, Petersen F R, Danielson B L, Day G W, Barger R L and Hall J L 1972 *Phys. Rev. Lett.* **29** 1346
[9] Bay Z, Luther G G and White J A 1972 *Phys. Rev. Lett.* **29**, 189
[10] Chou C W, Hume D B, Thorpe M J, Wineland D J and Rosenband T 2011 *Phys. Rev. Lett.* **106** 160801
[11] Major F and Werth G 1973 *Phys. Rev. Lett.* **30**
[12] Dehmelt H G 1975 *Bulletin Am. Phys. Soc.* **20** 60
[13] Hänsch T W and Schawlow 1975 *Opt. Commun.* **13** 68
[14] Wineland D J and Dehmelt H G 1975 *Bulletin Am. Phys. Soc.* **20** 637
[15] Neuhauser W, Hohenstatt M, Toschek P E and Dehmelt H 1980 *Phys. Rev.* A **22** 1137
[16] Fisk P T H, Sellars M J, Lawn M A, Coles C, Mann A G and Blair D G 1995 *IEEE Trans. Instrum. Meas.* **44**, 113
[17] Ludlow A D, Boyd M M, Ye J, Peik E and Schmidt P O 2015 *Rev. Mod. Phys.* **87**, 637