Optical Frequency Standards for Clocks of the Future

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Abstract

New scientific insight and technological developments of the past few years have stimulated renewed enthusiasm for the development of optical frequency standards. Long-standing problems have now been eliminated, and it appears that frequency standards using stable lasers and optical transitions may someday replace modern atomic clocks that are based on microwave transitions.

Keywords: atomic clocks, optical frequency standards, stable lasers, cold atoms

The excitement surrounding optical standards results from their potential to improve the accuracy and stability of atomic clocks by orders of magnitude. If so, they will profoundly effect areas of fundamental physics as well as advanced communication systems and navigation. The laser, atomic physics, and metrology communities from around the world are beginning to seriously explore the possibilities.

Since this session of the SPIE meeting is dedicated to the NIST centennial we will highlight some of the significant contributions from NIST scientists to the development of optical frequency standards. This limited and admittedly biased perspective cannot give the complete picture, and obviously will neglect many important contributions that originated in other institutions and laboratories throughout the world.

Historically, the measurement and calibration of time and frequency have relied upon precise astronomical observations combined with various types of mechanical oscillators. For the purpose of realizing a better clock, the first experimental demonstration of a frequency standard using an atomic transition was accomplished at NIST in 1949 by H. Lyons. This first "atomic clock" actually used molecules, and was based on the inversion transition in ammonia at 23.87 GHz in the microwave region. The ammonia atomic clock had a frequency instability of about 10⁻⁸. This was groundbreaking research that showed that it was indeed possible to construct a frequency standard referenced to a quantum transition. The ammonia frequency standard was known to have limitations, and the cesium clock as demonstrated in 1955 by L. Essen and J. Parry at NPL soon replaced it. For the past almost fifty years the Cs atomic clock has been the preferred choice for the highest accuracy. Serious research efforts and technological advances have improved the performance of Cs frequency standards significantly. We see in figure (1) that the progress has been steady, with the frequency uncertainty of Cs standards improving by a factor of ten every ten years or so.

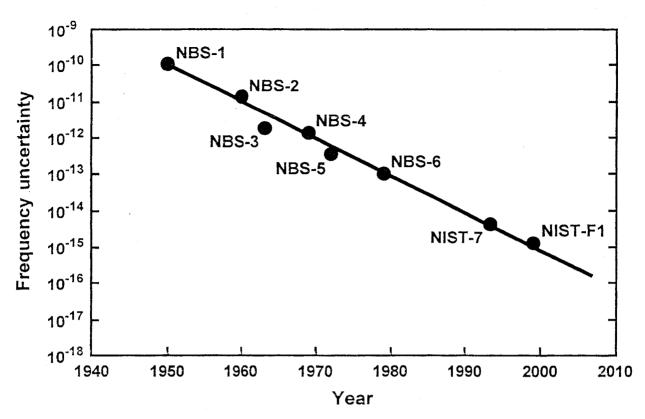


Fig. 1. This data shows the fractional frequency uncertainty of cesium atomic frequency standards at NIST over the past 50 years. Each result is characteristic of the state of the art at the corresponding time.

During the late 1950s and early 1960s it became clear that Cs atomic clocks were more precise, more stable, and more reproducible than the astronomical observations (combined with quartz crystals) that served as the time and frequency standards of the day. In addition, atomic clocks were more convenient to use; they could be reproduced in other laboratories, their signals could be easily distributed to users, and they were soon commercialized into compact reliable instruments. In 1967 the international standards organizations of the CGPM (Conférence Générale des Poids et Mesures) defined the unit of time (the second) in terms of the ¹³³Cs hyperfine frequency interval, 9 192 631 770 Hz, exactly. Some interesting general information on atomic clocks and their historical development can be found in references 1 and 2.

It was clear from the very beginning that there were significant advantages to using higher oscillation frequencies as the reference for precise time and frequency measurements. Higher operating frequencies would divide "time" into smaller units, thus giving higher precision and faster data rates. Even as microwave-based atomic clocks were being developed it was evident that using lasers and optical transitions could be advantageous. Unfortunately, there were also serious technical problems that made it extremely difficult to realize an atomic clock based on an optical transition. The main problems were: (1) that the coherent sources in the optical region (lasers) were not stable enough, (2) that the thermal motion of atoms caused Doppler broadening and shifts that resulted in significant systematic errors, and (3) that even when a laser was stabilized to an atomic transition there was no way to make a coherent connection between the frequency of the stable laser and the microwave and RF frequencies that are required for counting and distribution. Each of these problems has now been dealt with, as outlined below.

First, there was the need for narrow-linewidth highly stabilized lasers that could be tuned into resonance with narrow atomic or molecular transitions. The stabilization of laser frequencies has now progressed to a fine art, and a variety of narrow-linewidth laser sources are available. Most of the stabilization techniques lock the frequency of the laser to a high-finesse Fabry-Perot cavity, which serves as the stable frequency reference for short times, and also provides the signal-to-noise ratio required to narrow the laser's linewidth. Preeminent among the frequency stabilization methods is the technique known as the Pound-Drever-Hall method introduced in 1983.³ This technique has been used to stabilize a laser to a reference cavity with relative frequency fluctuations of a few millihertz⁴, and achieved relative frequency fluctuations between two different systems of ≈ 0.2 Hz averaged over 20 seconds.⁵ A recent paper by Hall⁶ provides an excellent overview of the field of stabilized lasers and frequency measurements, as well as some perspective on how we got to where we are today.

Second, the problem of atomic motion has been addressed in a couple of different ways. The idea of using laser light to cool atoms as proposed by Wineland and Dehmelt⁷, and Hänsch and Schawlow,⁸ was a major accomplishment in the field of atomic physics, and provides a foundation for the realization of modern frequency standards (microwave, optical, ion and neutral). Laser-cooling is now being used in *all* of the highest-accuracy atomic frequency standards around the world.

For the case of ions, the atomic motion can be reduced or effectively eliminated by using electromagnetic ion traps to confine the atoms. The concept of using trapped ions as frequency standards was proposed by Dehmelt⁹, and further refined by Wineland and collaborators¹⁰ at NIST, and many other scientists. Combined with the methods of laser cooling, trapped ions provide a sample of atoms that can be probed with lasers at very high resolution; linewidths as narrow as 6 Hz on a 10¹⁵ Hz transition in a single Hg⁺ ion have been demonstrated recently by Rafac et al.¹¹

For neutral atoms, the development of magneto-optic traps and optical molasses has led to routine production of samples with millions of atoms at temperatures in the milli or microkelvin range. Probing these cold atoms with methods of Doppler-free spectroscopy, such as saturated absorption and two-photon spectroscopy, have further reduce the problems associated with atomic motion. The spectral resolution can also be enhanced by using Ramsey's method of separated-oscillatory-fields, but now in the optical domain as suggested by Baklanov¹² and Bordé,¹³ and first demonstrated experimentally by Bergquist et al.^{14,15} at JILA and NIST. Optical Bordé-Ramsey fringes with shelving detection have been used with cold calcium atoms to produce an optical frequency standard with a short-term instability of $4 \times 10^{-15} \tau^{-1/2}$, where τ is the averaging time in seconds.¹⁶

Third, the other missing link that impeded the development of optical frequency standards was the lack of a direct coherent connection between optical frequencies and the countable microwave region. One concept, explored since the 1960s, uses repeated frequency multiplication of coherent sources from the microwaves up to higher frequencies in the far-infrared and infrared. These harmonic frequency chains have been powerful tools that have allowed the precise measurements of laser frequency standards,¹⁷ and a large number of molecular lines in the infrared with sufficient redundancy to predict accurately over 30,000 transition frequencies as exemplified by the work of Maki and Wells.¹⁸ However, it wasn't until the 1980s that difficult technical challenges were overcome by heroic efforts of Jennings and collaborators at NIST, who produced the first optical frequency chain that reached to the visible region of the spectrum.¹⁹ Worldwide to the present day, only a few harmonic frequency standards. Harmonic chains that operated regularly to 88 THz (3.39 µm in the infrared), and on rare occasions to the visible, have been run at NPL in the UK, NRC in Canada, the PTB in Germany, LPTF in France, and in Russia. Over the years, the methane-stabilized HeNe laser at 88 THz has played the major role in laser-based frequency standards of high accuracy. High-resolution saturated absorption spectroscopy in this system

56

gave the first experimental demonstration of photon-induced atomic recoil.²⁰ And it was the 1972 measurement of the frequency and wavelength of this standard at NIST that gave a precise value for the speed of light.²¹ Combined with measurements from other laboratories, and the subsequent measurement to the visible¹⁹ this led to the *definition* of the speed of light (c = 299792458 m/s exactly) and the result that the unit of length, the meter, is to be realized as the distance light travels in 1/(299792458) of a second, where the second is defined in terms of the Cs frequency standard.

The revolutionary development of the methods of femtosecond optical frequency metrology over the past three years by the groups lead by Hänsch at MPQ-Garching, and Hall at JILA-NIST now solves the problems associated with the optical frequency chains.^{22,23} This technique uses femtosecond mode-locked lasers to produce a spectrally broad comb of frequencies, where the frequency interval between modes of the comb is precisely equal to the repetition rate of the laser. A comb covering more than an optical octave can be generated by sending the output of a Kerr-lens-modelocked Ti-sapphire laser through ≈ 20 cm of special microstructure fiber. If the comb covers a full octave, and if the repetition rate is referenced to a Cs atomic clock we can measure the absolute frequency of an unknown laser in a straight forward manner. Detailed descriptions of how the femtosecond frequency measurement systems operate can be found in references[24,25]. We now have access to a practical, compact "optical clockwork" that coherently connects the visible to the microwave region. In figure 2 we see the historical improvement in absolute optical frequency measurements and standards.

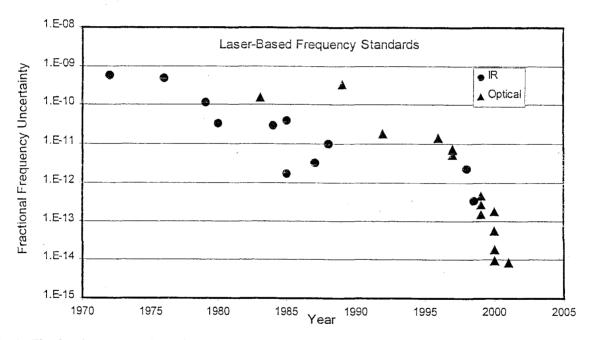


Fig. 2. The fractional uncertainty of some of the most accurate absolute frequency measurements of laser standards displayed as a function of date. The round symbols indicated measurements of IR standards [CH₄ stabilized HeNe lasers at 88 THz.(3.39 μ m), and OsO₄ stabilized CO₂ lasers at 29 THz (10 μ m)], while the triangular symbols represent various "optical" frequency standards (somewhat arbitrarily chosen as f > 300 THz, λ <1000 nm). Harmonic frequency chains were used prior to 1999, but since that date femtosecond optical combs have dominated.

The high accuracy that is achievable with femtosecond optical frequency combs was the key discovery that altered the direction of optical frequency measurements. Because of this success, and also because of the dramatic simplification in optical frequency synthesis, many groups are building mode-locked laser systems for precise measurements frequency and time interval. Combined with advances in stabilized lasers, and laser cooling and trapping, optical frequency standards are becoming a very viable

option. The dramatic improvements that have occurred in the past three years are apparent in the data plotted in figure 2. Projections for the future of optical frequency standards include: frequency uncertainties as low as 1×10^{-18} for trapped ions, and instabilities $< 1 \times 10^{-16} \tau^{-1/2}$ for neutrals. The next few years will provide the opportunity to test these predictions, and to explore totally new regimes of atomic frequency standards and clocks.

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References

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⁵ B. C. Young, F. C. Cruz, W. M. Itano, and J. C. Bergquist, "Visible Lasers with SubHertz Linewidths", Phys. Rev. Lett., **82**, 3799-3802 (1999).

⁶ J.L. Hall, "Optical Frequency Measurement: 40 Years of Technology Revolutions," IEEE J. Selected Topics in Quant. Electron., 6, 1136-1144 (2000).

⁷ D.J. Wineland and H.G. Dehmelt, "Proposed $10^{14}\Delta\nu < \nu$ Laser Fluorescence Spectroscopy on Tl⁺ Mono-Ion Oscillator III," Bull. Am. Phys. Soc. 20, 637 (1975).

⁸ T.W. Hänsch and A.L. Schawlow, Cooling of Gases by Laser Radiation," Opt. Comm., 13, 68- (1975).

⁹ H.G. Dehmelt, "Mono-Ion Oscillator as Potential Ultimate Laser Frequency Standard," IEEE Trans. Instrum. Meas. IM-**31**, 83-87 (1982).

¹⁰ D.J. Wineland, J.C. Bergquist, J.J. Bollinger, W.M. Itano, D.J. Heinzen, S.L. Gilbert, C.H. Manney, and M.G. Raizen, "Progress at NIST Toward Absolute Frequency Standards Using Stored Ions," IEEE Trans. Ultrason. Ferroelect. and Freq. Control, **37**, 515-523 (1990).

¹¹ R. J. Rafac, B. C. Young, J. A. Beall, W. M. Itano, D. J. Wineland, and J. C. Bergquist, "Sub-dekahertz Ultraviolet Spectroscopy of 199Hg+", Phys. Rev. Lett., **85**, 2462-2465 (2000).

¹² Ye.V. Baklanov, B. Ya. Dubetsky, and V.P. Chebotayev, "Non-Linear Ramsey Resonance in the Optical Region," Appl. Phys. 9, 171-173 (1976).

¹³ Ch. J. Bordé, Ch. Salomon, S. Avrillier, A. Van Lerberghe, Ch. Bréant, D. Bassi, G. Scoles, "Optical Ramsey Fringes with Traveling Waves," Phys. Rev. A, **30**, 1836-1848 (1984).

¹⁴ J.C. Bergquist, S.A. Lee, and J.L. Hall, "Saturated Absorption with Spatially Separated Laser Fields: Observation of Optical Ramsey Fringes," Phys. Rev. Lett., **38**, 159-162 (1977).

¹⁵ R.L. Barger, J.C. Bergquist, T.C. English, and D.J. Glaze, "Resolution of the Photon-Recoil Structure of the 6573-Å Calcium Line in an Atomic Beam with Optical Ramsey Fringes, Appl. Phys. Lett. **34**, 850-852 (1979).

¹⁶ C.W. Oates, F. Bondu, R.W. Fox and L. Hollberg, "A Diode-Laser Optical Frequency Standard Based on Laser-Cooled Ca Atoms: Sub-kilohertz Spectroscopy by Optical Shelving Detection, Eur. Phys. J. **D**7, 449-460 (1999); C. W. Oates, E. A. Curtis, and L. Hollberg, "Improved short-term stability of optical frequency standards: approaching 1 Hz in 1s with the Ca standard at 657 nm," Opt. Lett., **25**, 1603-1605 (2000).

¹⁷ D.A. Jennings, K.M. Evenson, and D. J.E. Knight, "Optical Frequency Measurements", Proc. IEEE, 74, 168-179 (1986).
¹⁸ A.G. Maki and J.S. Wells, "Wavenumber Calibration Tables from Heterodyne Frequency Measurements," NIST

¹⁸ A.G. Maki and J.S. Wells, "Wavenumber Calibration Tables from Heterodyne Frequency Measurements," NIST Special Publication 821, U.S. Gov. printing office, Washington (1991).

¹⁹ D.A. Jennings, C.R. Pollock, F.R. Petersen, R.E. Drullinger, K.M. Evenson, J.S. Wells, J.L. Hall, and H.P.Layer, "Direct frequency measurement of the I2-stabilized He-Ne 473 THz (633 nm) laser", Optics Lett. 8, 136-138 (1983).

58

¹ T. Jones, Splitting the Second – The Story of Atomic Time, IOP, London (2000).

² D.W. Allan and C.C. Hodge, "The Science of Time Keeping," Agilent AN1289, Agilent Technologies.

³ R.W.P. Drever, J.L.Hall, F.V. Kowalski, J. Hough, G.M. Ford, A.J. Munley, and H. Ward, Laser Phase and Frequency Stabilization Using an Optical Resonator," Appl. Phys B, **31**, 97-105 (1983).

⁴ C. Salomon, D. Hils, and J.L. Hall, "Laser Stabilization at the milliHertz Level", J. Opt. Soc. Am. B, 5, 1576-1587 (1988).

²⁰ J.L. Hall, C.J. Borde, K. Uehara, "Direct Observation of the Recoil Effect Using Saturated Absorption Spectroscopy," Phys. Rev. Lett. **37**, 1339-1342 (1976).

²¹ K.M. Evenson, J.S. Wells, F.R. Peterson, B.L. Danielson, and G.W. Day, R.L. Barger, and J.L. Hall, Speed of Light from Direct Frequency and Wavelength Measurements of the Methane-Stabilized Laser, Phys. Lett. 29, 1346-1349 (1972).

²² Th. Udem, J. Reichert, R. Holzwarth, and T.W. Hänsch, "Accurate Measurement of Large Optical Frequency Differences with a Mode-Locked Laser," Optics Lett., **24**, 881-883 (1999).

²³ S.A. Diddams, D.J. Jones, J. Ye, S.T. Cundiff, J.L. Hall, J.K. Ranka, R.S. Windeler, R. Holzwarth, T. Udem and T.W. Hänsch, "Direct Link between Microwave and Optical Frequencies with a 300 THz Femtosecond Laser Comb," Phys. Rev. Lett., 84, 5102-5105 (2000).

²⁴ D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall, and S. T. Cundiff, "Carrierenvelope phase control of femtosecond mode-locked lasers and direct optical frequency synthesis," Science, **288**, 635-639 (2000).

²⁵ S. A. Diddams, Th. Udem, K. R. Vogel, C. W. Oates, E. A. Curtis, R. S. Windeler, A. Bartels, J. C. Bergquist, and L. Hollberg, , "A Compact Femtosecond-Laser-Based Optical Clockwork", Proc. SPIE vol. **4269**, Laser Frequency Stabilization Standards, Measurement and Applications, San Jose, CA (2001).