

SPECTROSCOPY

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Absolute Optical Frequency Metrology

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Introduction

Visible light occupies a portion of the electromagnetic spectrum between frequencies of approximately 400 terahertz (THz) to 750 THz, where 1 THz is 10^{12} oscillations per second. Optical radiation, which includes adjacent infrared and ultraviolet portions of the spectrum, covers a frequency range of approximately 15–1500 THz. The techniques required for measuring frequencies in this range are qualitatively different from those currently employed in the radio frequency (RF) and microwave portions of the spectrum, where high-speed electronic counters make a direct measurement possible.

The frequency of an oscillatory phenomenon is the number of oscillations that occurs in unit time, or alternatively, the inverse of the period of one oscillation. Thus, to understand frequency, we must understand time, which is intrinsically related to phase. Only a time interval, the time elapsed between two events, has fundamental physical meaning. The 'time' that we are most familiar with, the time of day (including the date), is based on an arbitrarily chosen

starting point. The starting point chosen in current international agreements can be traced to noon on December 31st, 1899. Furthermore, the time defined by those agreements, Universal Coordinated Time (UTC), is periodically adjusted relative to atomic time (discussed below) to compensate for irregularities in the rotation of the Earth.

Knowledge of the transition frequencies (energies) in simple atoms gives detailed information on the structure of the atoms. This can be used to test our understanding of the fundamental interactions in nature by comparing the measured frequencies with predictions of fundamental physical theories, in this case quantum electrodynamics (QED). Beautiful results along these lines are exemplified by precision measurements of transition frequencies in hydrogen and helium. In addition, the ability to measure and precisely control laser frequencies promises tremendous advances in the performance of the next generation of atomic clocks and frequency standards. These have potential applications to navigation and communication systems.

The measurement of frequency and time intervals also involves a choice, that of the unit of time, i.e., the definition of the second. The original definition of the second was based on the rotation of the Earth. However, not only is it irregular, but it is slowing down. Both of these facts limit its utility as a frequency standard. Once atomic clocks were highly developed, the second was redefined in 1967 to be the time it takes for the $F = 4, m_F = 0 \rightarrow F = 3, m_F = 0$ transition in the hyperfine structure of the ground state of ^{133}Cs to undergo 9192 631 770 oscillations. Thus an 'absolute' measurement of time interval, and hence frequency, must be directly connected to this defining frequency. The basic structure of an atomic clock is shown in Figure 1.

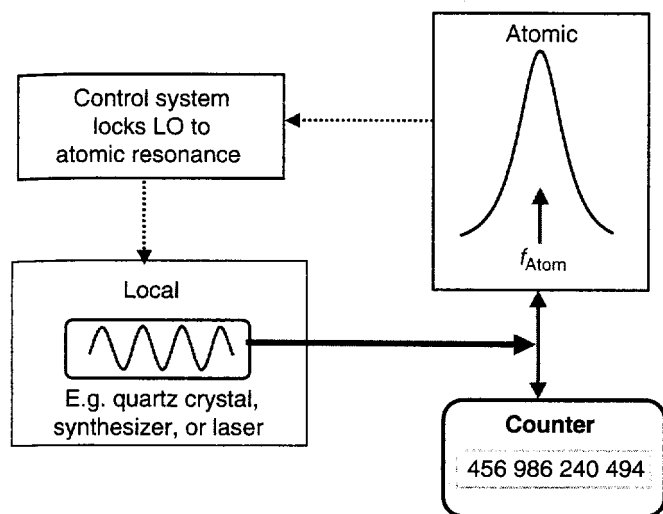


Figure 1 An atomic clock has three basic components: an atomic resonance, an oscillator to probe and lock to the atomic resonance, and a counter that records the number of oscillations and thus displays the time interval since some chosen starting time.

Frequency standards (or clocks) are characterized by three properties: accuracy, stability, and reproducibility. Accuracy describes how well the natural and fundamental atomic frequency is produced by the atomic frequency standard. Stability describes the frequency fluctuations on short time-scales (an accurate but unstable standard produces a frequency that fluctuates around a constant mean that is the atomic frequency). Reproducibility between standards addresses whether or not two implementations of the standard produce exactly the same frequency. Atomic clocks are intrinsically accurate because the frequency of the transitions they are based on is determined by identical atoms in nature. The largest inaccuracies are usually due to environmental perturbations such as tiny variations in the local magnetic field.

A careful analysis of all of these issues, together with technical considerations, yielded the choice of cesium for the current definition of the second. Other common frequency standards include quartz crystal oscillators, hydrogen masers, and rubidium vapor cell standards. Quartz oscillators are inexpensive and have good stability, but in comparison to atomic standards they suffer from accuracy and reproducibility issues. Hydrogen masers have the best stability, but also poor accuracy and reproducibility compared to cesium. Vapor cell rubidium standards can be small and inexpensive but have poorer accuracy than cesium beam standards due to cell and buffer gas effects.

There are several motivations for using optical sources rather than RF or microwaves to measure

time intervals and frequencies. Obviously, a higher oscillation frequency can divide time into smaller units and this can provide higher measurement precision. It is thus natural to use the highest oscillation frequency that can be precisely counted to measure time and frequency. Not long after lasers were invented, suggestions were made that it would be possible to make optical frequency standards using lasers locked to narrow atomic resonances.

A basic feature of optical transitions is that they have a very high Q factor (the ratio of the center frequency to the linewidth). This allows them to be measured to very high precision (current measurements are good to a few parts in 10^{15}). It may be possible to use this high precision to build improved atomic clocks that use optical transitions instead of the microwave transition currently being used. Candidates for optical standards include both trapped ions (Hg^+ , Yb^+ , Sr^+ , and In^+ are under investigation) and laser-cooled neutral atoms (Ca , Sr , Mg , Ba , and Ag). The high precision is also utilized for fundamental tests of quantum electrodynamics by comparing transition frequencies (usually of hydrogen or helium atoms) to first-principle calculations. For comparison Figure 2 shows the relevant transitions and energy levels in the Cs microwave standard and the Hg^+ optical frequency. It is even possible to use the precision of atomic clocks for high-sensitivity tests of variation in the fundamental constants with time. Finally, because the speed of light is constant length measurements based on the wavelength of light with a known frequency, the precision of length metrology ultimately rests on optical frequency metrology.

An obvious question, which might occur to many in the field of optics, is why a standard spectrometer does not provide absolute frequency measurements. All spectrometers measure wavelength, not frequency, based on interference between light traveling two different path lengths. Thus, knowledge of the path length is needed to determine the wavelength. However, since length is now defined using the speed of light, the definitions become circular unless a source of light with known absolute frequency is available. In addition, frequency measurement is intrinsically more precise and reliable than mechanical length measurement.

Recently, there has been a significant breakthrough in optical-frequency metrology and optical clocks by using mode-locked lasers that generate optical pulses with durations of a few femtoseconds. Before discussing optical frequency metrology based on mode-locked lasers, we will review earlier techniques to provide the necessary background. After describing

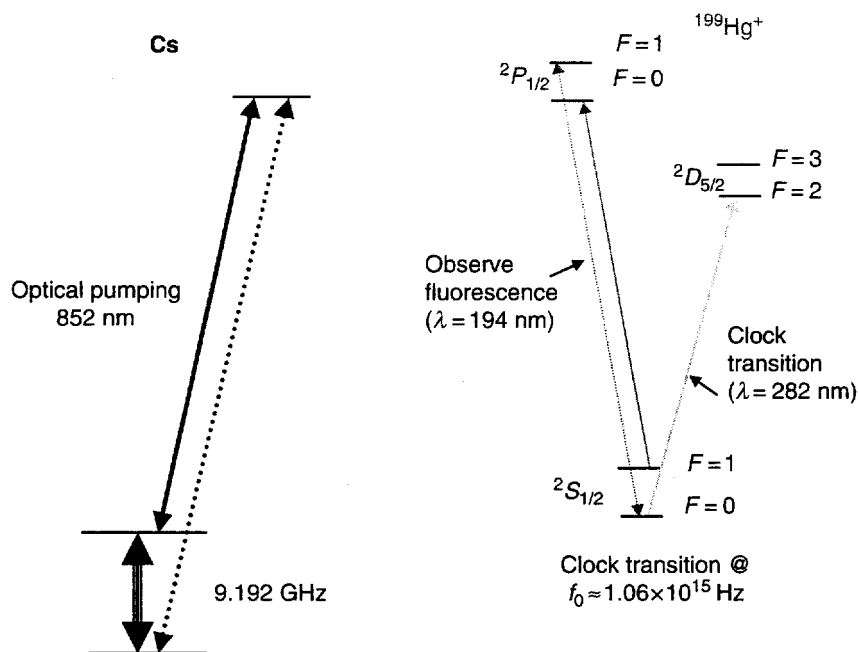


Figure 2 Simplified energy level diagram of Hg^+ and Cs.

methods that use mode-locked lasers, we will then provide a summary of current measurements and standards. Finally we will briefly describe the outlook for the future of absolute optical frequency metrology.

Background

To imagine being able to measure the frequency of light we need to have laser sources with high coherence, like that of radio waves, where the phase is stable enough to be measured. Even though the first beat-note between two lasers was demonstrated in the 1960s, most lasers were not coherent (stable) enough that the optical phase could be tracked for much longer than about 10 to 100 ns. After many years of research and inspiring technological achievements, Hall, Chebotayev, Bergquist and others were able to stabilize the frequency of tunable laser sources to the point that lasers could indeed be considered coherent sources, with the phase continuously measurable and even predictable for times as long as seconds. Once the lasers were stable enough to be counted, there was still the essential problem that no counter was fast enough to actually count optical frequencies.

With spectrally pure laser sources it was possible to conceive of measuring optical frequencies by extending the nonlinear methods that are used to generate and measure RF and microwave frequencies. By using simple nonlinear components such as diodes, it is easy to generate high harmonics of an input frequency in the RF and microwave regions of the spectrum. At least in principle, and by direct analogy, it seemed that it should be possible to multiply coherent sine

waves from radio frequencies up through the microwave region to reach the THz frequencies of far-infrared lasers, and then to use nonlinear optical methods to multiply on up to the infrared (IR) and eventually to the visible region of the spectrum. The concept is simple enough, $f \rightarrow 100 \times f \rightarrow 1000 \times f \rightarrow 2000 \times f \rightarrow 4000 \times f \dots$ until you reach an optical frequency. Multiplication up to the microwave region works quite well, but unfortunately the nonlinear optical processes in the far-infrared, IR, and optical regions are typically quite inefficient; the power at the second harmonic is typically only 10^{-5} times the square of the fundamental power in watts. Just multiplying a single-frequency laser by a factor of two can be a technological challenge. After reaching a microwave frequency of about 50 GHz we still require a multiplication factor of 10^4 to reach the visible at 500 THz. Since we are usually forced to multiply in the IR and optical by factors of 2, we have $2^N = 10^4$, which means we require $N \approx 13$ multiplication stages. This is approximately the number of stabilized laser oscillators that are required to span the frequency gap from the microwave to the visible. Notwithstanding these technological limitations, Evenson and collaborators did a groundbreaking demonstration of a harmonic frequency chain in 1972 that connected a microwave atomic frequency standard and a stabilized HeNe laser in the IR at 88 THz. With a concerted multiperson effort, more lasers, and 10 years of research, Jennings and co-workers finally succeeded in extending the harmonic optical frequency chain to the visible. That original optical frequency chain from the RF to the

visible is diagrammed in Figure 3. In the succeeding 18 years, only three other harmonic frequency chains were built world-wide that connected the microwave standards to optical frequencies.

An alternative scheme for measuring optical frequencies is the optical bisection method, a conceptually simple technique proposed by Telle, Meschede and Hänsch, for dividing down from

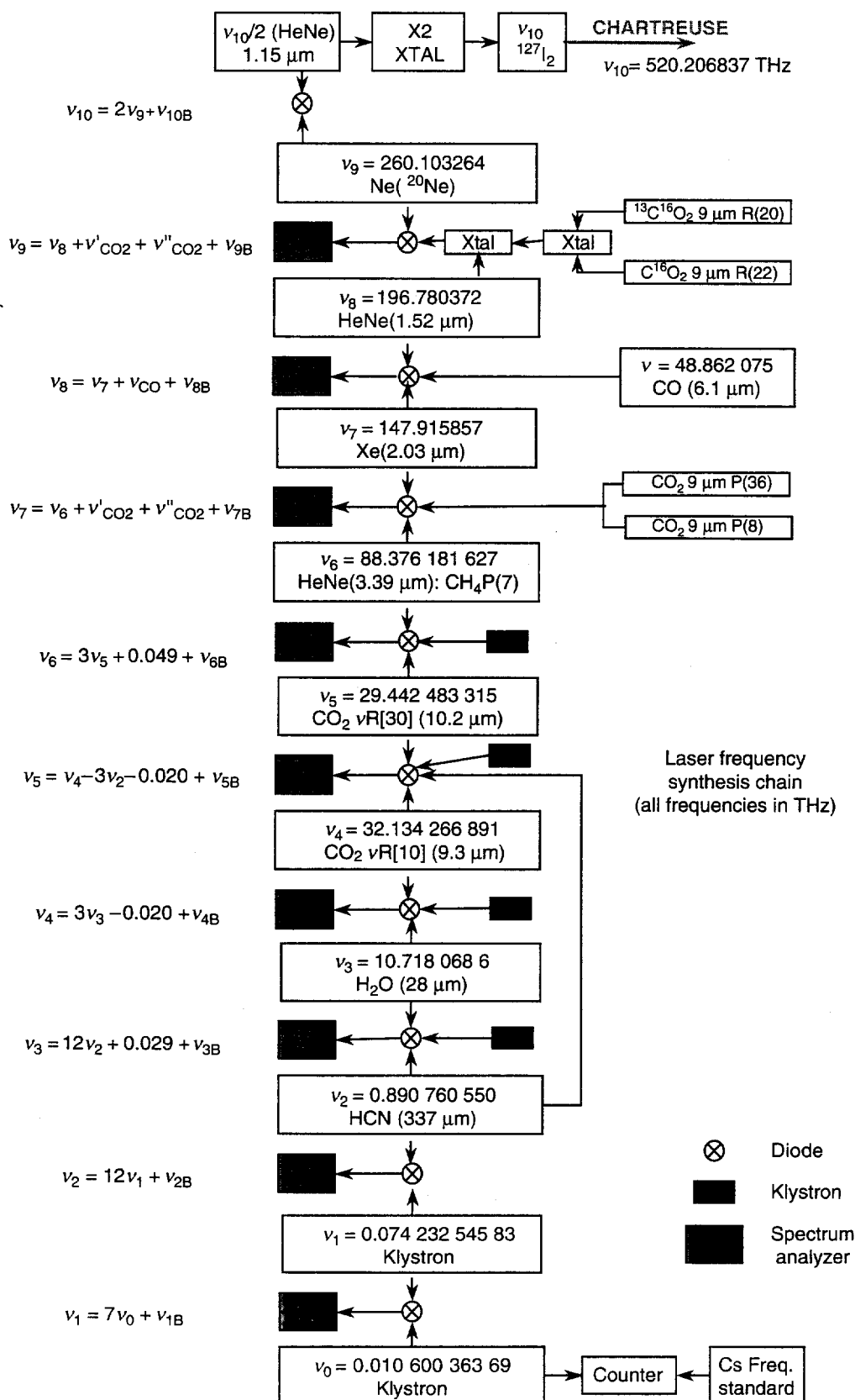


Figure 3 Schematic of the first frequency chain used to measure optical frequencies relative to the Cs standard.

optical frequencies to microwaves. In fact, the bisection method divides an arbitrary optical frequency interval in half by utilizing second-harmonic generation and sum-frequency-mixing, and forcing the condition that $2f_3 = f_1 + f_2$. Thus, the laser at frequency $f_3 = (f_1 + f_2)/2$ bisects the frequency interval between the lasers at f_1 and f_2 . The optical bisection method has the following two significant advantages over the multiplication method: the phase noise is decreased in successive divisions (rather than increased by multiplications), and the system could be constructed out of one convenient and reproducible laser technology, say diode lasers in the near-visible. In principle, any optical frequency could be measured by successive operations of the optical bisector. Optical frequency intervals as large as 8 THz have been measured with this method, but even though it is feasible, no bisection system has been built to date that connects the optical region all the way to countable microwave frequencies. The main limitation is the same as that facing the multiplication scheme: it still requires lots of stabilized laser sources to divide by factors of 2 from 10^{15} to 5×10^{10} Hz. The problem can be reduced somewhat by taking advantage of an optical-frequency comb generator based on an electro-optic modulator inside a resonant optical cavity. These systems, developed by Korougi and others, generate many coherent microwave sidebands on a laser so that optical frequency intervals on the order of 5 THz can be measured.

Even combining all of these ideas and techniques, a convenient system to count optical frequencies remained elusive. This has now changed with the revolutionary new optical frequency combs based on mode-locked femtosecond lasers.

Optical Frequency Metrology with Femtosecond Combs

Recently, the introduction of mode-locked lasers into the field of absolute optical frequency metrology has resulted in an important advance. The extremely complex frequency chains described above can be replaced by a single mode-locked laser if it produces pulses with sufficiently large bandwidth. Although the underlying ideas are not new (they were originally discussed by Hänsch and Chebotayaev in the 1970s) it is only recently that mode-locked lasers with the required characteristics were developed and the concept carefully tested.

The dramatic simplification of a frequency chain that uses a mode-locked laser means that absolute optical-frequency measurements can now be made by a single person as compared to the team of

approximately 10 highly trained scientists required to run the previous chains. This has resulted in a large number of measurements being reported in the last 2 years. The simplicity also allows longer averaging times and a greater number of measurements to be used in determining the final reported frequency. In addition, since a simpler system has fewer sources of error, it has improved the quality of measurements made in a given amount of time.

Mode-locked lasers produce ultrashort pulses of light. For a typical modern high-quality mode-locked laser, the pulse duration is around 10 fs ($1 \text{ fs} = 10^{-15}$ second), corresponding to three or four optical cycles in the near-infrared, where these lasers typically operate. The current best mode-locked lasers can produce pulses that are shorter than two optical cycles in duration (5–6 fs). Since the spectrum of these lasers is complicated, the bandwidth is significantly larger than it would be for a smooth pulse such as one with a Gaussian or hyperbolic-secant temporal intensity profile.

At first sight, using mode-locked lasers for optical-frequency metrology seems counter-intuitive, because optical-frequency metrology clearly requires very well-defined frequencies in contrast to the broad frequency spectrum of ultrashort pulses. This apparent conundrum is resolved by the fact that a mode-locked laser actually produces a very regularly spaced train of ultrashort pulses. The ultrashort pulses emitted by a mode-locked laser are replicas of a steady-state pulse that circulates inside the cavity of the laser. Every time the internal pulse impinges on the output coupler, which is a partially reflective mirror, a portion of it is transmitted, producing the output. The timing between these output pulses is determined purely by the time, τ_c , for the intracavity pulse to make one round trip, which is typically between 1 and 20 ns. The spectrum of a train of pulses is a regularly spaced 'comb' of frequencies spaced by the repetition rate of the train, $f_{\text{rep}} = 1/\tau_c$. If all of the pulses are identical, then these comb lines are just integer multiples of f_{rep} . However, due to dispersion, the group and phase velocities are different inside the cavity of a mode-locked laser. This leads to a pulse-to-pulse shift in the phase between the carrier and envelope (which we call the carrier-envelope phase; ϕ_{ce} , the pulse-to-pulse shift of ϕ_{ce} is designated by $\Delta\phi_{\text{ce}}$). The presence of $\Delta\phi_{\text{ce}}$ results in a rigid shift of all the comb lines by an amount $\delta = 2\pi\Delta\phi_{\text{ce}}/f_{\text{rep}}$. Thus the frequencies of the comb lines are given by $\nu_n = nf_{\text{rep}} + \delta$, where n is a large integer of order 10^6 . The essential point of this equation is that it gives optical frequencies in terms of RF frequencies (f_{rep} and δ) that can easily be measured with conventional electronics and

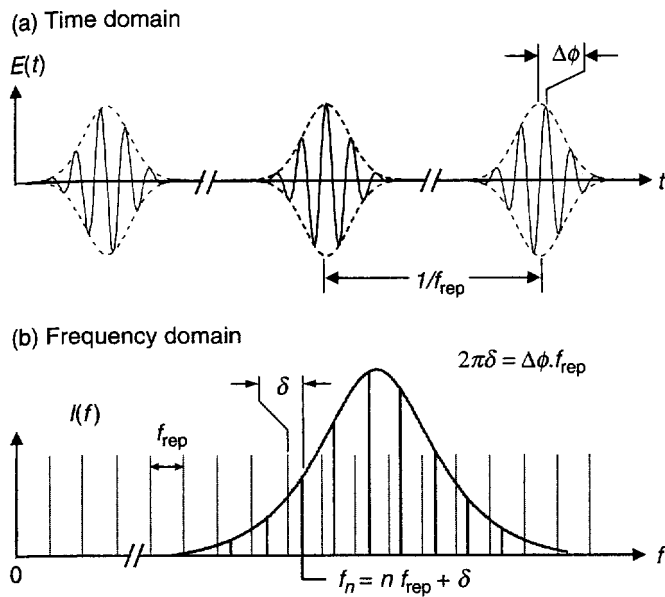


Figure 4 Time-frequency correspondence and relationship between $\Delta\phi$ and δ . (a) In the time domain, the relative phase between the carrier and the envelope evolves from pulse to pulse by the amount $\Delta\phi$. (b) In the frequency domain, the elements of the frequency comb of a mode-locked pulse train are spaced by f_{rep} . The entire comb is offset from integer multiples of f_{rep} by an offset frequency $\delta = \Delta\phi f_{\text{rep}}/2\pi$.

compared to the microwave cesium frequency standard. The relationship between the time and frequency domains is shown in Figure 4.

Given such a comb of frequencies, a heterodyne measurement readily yields the frequency difference between the unknown optical frequency of a narrow-band source (typically a single frequency laser) and the optical frequency of a nearby comb line. Thus, the absolute unknown frequency can be determined if the absolute frequencies of the comb lines are known. This requires measurement of f_{rep} and δ . The repetition rate, f_{rep} , is easily measured with a fast photodiode. Measurement of δ requires somewhat greater effort.

The development of a 'self-referencing' method to easily measure δ is the key enabling breakthrough that has brought about the recent revolution in optical-frequency metrology. It is called self-referencing because it provides a direct measure of δ with no other optical frequencies as input. It works by comparing frequencies that differ by a factor of two (typically these lie in the wings of the spectrum) using second-harmonic generation. Specifically, the frequency difference between the second harmonic of a comb line n and the comb line $2n$ is given by $2f_n - f_{2n} = 2(nf_{\text{rep}} - \delta) - (2nf_{\text{rep}} - \delta) = \delta$. Such a frequency difference is easily measured using a heterodyne beat. This technique is shown schematically in Figure 5. Other variations on this basic idea are possible; for example, rather than frequency doubling

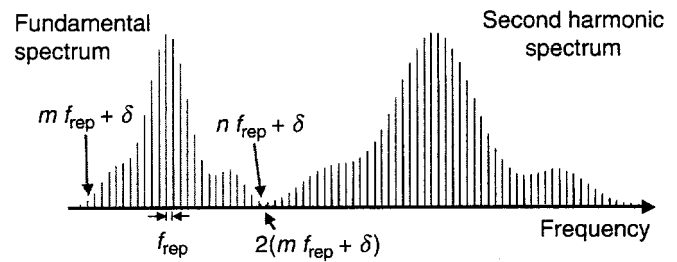


Figure 5 Schematic of the self-referencing technique for determining the offset frequency of the frequency comb produced by a femtosecond mode-locked laser. The frequency difference between the fundamental comb and its second harmonic is the offset frequency. In the region where overlap occurs, this is easily detected as a heterodyne beat.

the comb itself, it is also possible to extract the same information by comparing the comb lines to a single-frequency laser and its second harmonic.

Implementation of the self-referencing technique requires a pulse spectrum that spans a factor of two in frequency (an optical octave). Although a laser has recently been demonstrated that produces such a broad spectrum, they are not commonly available. However, the development of microstructured fiber has made it easy to broaden the spectrum of ordinary mode-locked lasers so that it spans an octave, as was first demonstrated by Ranka and co-workers at Bell Labs. The broadening occurs because optical nonlinearity in the fiber results in self-phase modulation of the laser pulse. Microstructure fiber achieves guiding by surrounding the core region with microscopic air holes, as compared to regular fiber that uses doping to produce a difference in the index of refraction. The much larger difference in the index of refraction in microstructure fiber allows a smaller core region to be used, which increases the effective nonlinearity. It also makes it possible to modify the dispersion such that the group velocity dispersion goes through zero for light with a wavelength close to 800 nm. This is crucial because group-velocity dispersion in ordinary fiber causes ultrashort pulses to temporally spread, which lowers the peak power and hence nonlinearity. It is possible to implement self-referencing techniques using less bandwidth if higher orders of nonlinearity are used; for example, if the difference between the second harmonic of the high-frequency end and third harmonic of the low-frequency end of the comb are used, only a half-octave bandwidth is required.

Given the ability to measure both f_{rep} and δ , and as long as its frequency lies within the comb spectrum, it is possible to measure the optical frequency of any optical source by measuring the heterodyne beat

Figure 7 Actual frequency measurements of the beat-note between a hydrogen maser stabilized optical frequency comb (as shown in **Figure 6**) and the NIST Hg^+ optical frequency standard. The scatter in the data for counter gate time of 10 seconds corresponds to a fractional frequency uncertainty of about 4×10^{-14} , consistent within the instability in the maser. The inset shows the results from numerous datasets of measurement of the frequency of the Hg^+ optical standard relative to the cesium primary standard at NIST. The frequency data are plotted relative to 1064 721 609 899 142.6 Hz. The dotted lines give estimates of systematic uncertainties in lieu of a complete accuracy evaluation.

Table 1 Summary of optical frequency measurements of a selection of molecular, atomic, and ionic transitions. Recent measurements listed for all and additional measurements are given for a few to indicate the level of agreement

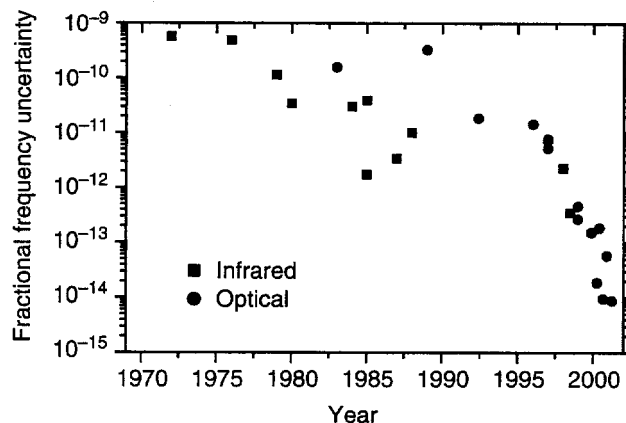
Atom/molecule/ion	Wavelength (nm)	Frequency (THz)	Uncertainty		Year	First author and institution
			\pm Hz	Frac.		
O ₃ O ₄	10318.0	29.054 057 446 660	50	2×10^{-12}	1985	Clarion, LPTF
		29.054 057 446 579	10	3×10^{-13}	1999	Ducos, LPTF
CH ₄	3392.2	88.376 181 627 000	50 000	6×10^{-10}	1972	Evenson, NBS
		88.373 149 028 553	200	2×10^{-11}	1998	Ering, PTB
Rb-2 photon	778.11	385.285 142 367 000	8000	2×10^{-11}	1993	Nez, ENS
		385.285 142 374 800	3000	8×10^{-12}	2000	Diddams, JILA
Sr ⁺	674.03	444.779 904 409 540	200	4×10^{-13}	1999	Bernard, NRC
Ca	657.45	455.986 240 495 150	8	10×10^{-14}	2003	Helmcke, PTB
		455.986 240 494 158	26	6×10^{-14}	2000	Udem, NIST
I ₂ a ₁₅ of R(127) 11-5	632.99	473.612 340 492 000	74 000	2×10^{-10}	1983	Jennings, NBS
I ₂ a ₁₆ of R(127) 11-5	632.99	473.612 353 604 800	1200	3×10^{-12}	2000	Ye, JILA
I ₂ a ₁₀ of R(56) 32-0	532.24	563.260 223 480 000	70 000	1×10^{-10}	1995	Jungner, JILA
		563.260 223 514 000	5000	9×10^{-12}	2000	Diddams, JILA
Yb ⁺	435.51	688.358 979 230 931	6	9×10^{-15}	2001	Stenger, PTB
H 2S-8S	389.01	770.649 561 581 100	5900	8×10^{-12}	1997	de Bouvoir, ENS
Hg ⁺	281.57	1064.721 609 899 140	10	9×10^{-15}	2000	Udem, NIST
In ⁺	236.54	1267.402 452 899 920	230	2×10^{-13}	2000	von Zanthier, MPQ
H 1S-2S	121.57	2466.061 413 187 100	46	2×10^{-14}	2000	Holzwarth, MPQ

Summary of Measurements/Standards

Starting with the first measurement of the frequency of the methane-stabilized HeNe laser in 1972, a new laser frequency measurement was reported every year or so using harmonic frequency chains. These are summarized in Table 1. An indication of the history of frequency measurements of stable laser references is given in Figure 8. The accuracy, in terms of fractional frequency uncertainty, improved from 10^{-10} in 1972 to 10^{-12} in 1999, at which time the femtosecond optical frequency combs came on line and there was a dramatic improvement in the precision as well as the number of measurements that were completed. It is impossible to predict with certainty but some have projected that optical standards can ultimately reach uncertainties as small as 10^{-18} .

Outlook

Three separate technologies have now reached a level of maturity that it is possible to build high-performance optical-frequency standards and clocks. The essential achievements are: laser cooling and trapping of atoms (first proposed by Wineland and Dehmelt, and Hänsch and Schawlow), highly stabilized narrow-linewidth cw lasers, and femtosecond optical frequency combs. Combining these key ingredients, we can construct an optical atomic clock as shown schematically in Figure 9.

**Figure 8** Progress in the accuracy of optical frequency measurements.

Here, for comparison with Figure 1, the cold atoms or single ion provide the narrow atomic resonance, the cw laser serves as the local oscillator to probe the resonance, and the femto-comb serves as the counter. Optical frequency standards of the future are expected to provide orders of magnitude better stability and improved accuracy over the existing atomic frequency standards that now use microwave transitions in atoms. As described in the previous section, frequency combs produced by femtosecond lasers can directly measure the frequency of a stable laser locked to an atomic transition relative to a known microwave frequency standard. This gives fundamental information about the atomic energy levels, and structure, and allows comparisons between different elements. However, to take

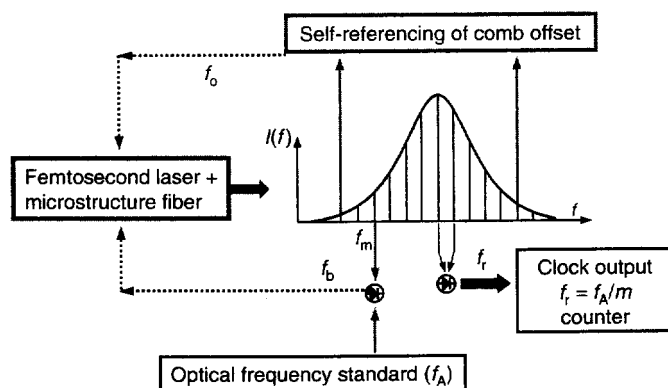


Figure 9 Simplified schematic of an optical clock.

advantage of the high stability of the optical references we run the system as an optical clock (Figure 9) where the stable laser and the femto-comb are locked to the atomic resonance, and the clock output comes as pulses at the repetition rate (e.g., 1 GHz) of the femtosecond mode-locked laser. With a judicious choice of control parameters it is possible to have the pulse repetition frequency, i.e., the clock output, at an exact subharmonic of the optical transition frequency. It is intriguing to note that a portable optical clock could measure time and length at the same time.

See also

Instrumentation: Spectrometers. **Quantum Electrodynamics:** Quantum Theory of the Electromagnetic Field.

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Fourier Transform Spectroscopy

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Introduction

Fourier transform spectroscopy (FTS) has emerged as one of the most powerful spectroscopic techniques since the first instruments became commercially available in the late 1960s. With this method, high

spectral resolution, high wavenumber accuracy, broad spectral range, high optical throughput, and high signal to noise ratio can be achieved simultaneously. Because of these combined advantages that will be discussed in more detail throughout this article, especially in the far- ($10\text{--}500\text{ cm}^{-1}$) and mid-infrared (FIR and MIR) spectral region ($500\text{--}5000\text{ cm}^{-1}$), FTS has become the method of choice for the most sensitive spectral investigations. In this article, an overview of the setup, the working principle, and the computation of spectra from the