Chapter 9

FEMTOSECOND LASERS FOR OPTICAL CLOCKS AND LOW NOISE FREQUENCY SYNTHESIS

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Abstract: In the late 1990's mode-locked femtosecond lasers were introduced as an important new tool for the synthesis and measurement of optical frequencies. The simplicity, robustness and improved precision of femtosecond lasers have now led to their prominence in the field of optical frequency metrology. In addition, their use is developing significant new time-domain applications based on the precise control of the carrier-envelope phase. It is anticipated that narrow linewidth lasers referenced to optical transitions in atoms and ions will soon be the best electromagnetic frequency references of any kind, with projected fractional frequency instability below $1\times10^{-15}\tau^{-1/2}$ and uncertainties approaching 1×10^{-18}. When used in conjunction with such ultraprecise frequency standards, the femtosecond laser serves as a broadband synthesizer that phase coherently converts the input optical frequency to an array of optical frequencies spanning hundreds of terahertz and to countable microwave frequencies. The excess fractional frequency noise introduced in the synthesis process can approach the level of 1×10^{-19}.

Key words: optical atomic clock, time and frequency metrology, femtosecond laser, optical frequency metrology, optical frequency comb, frequency synthesis

1. INTRODUCTION

Recent developments in time and frequency metrology have brought us to the point where we can begin to address the possibility of using atomic clocks "ticking" at optical frequencies (~500 THz) to measure time intervals with uncertainties approaching 1 part in 10^{18}. This represents a dramatic shift from the past fifty years of precision time keeping, where atomic clocks

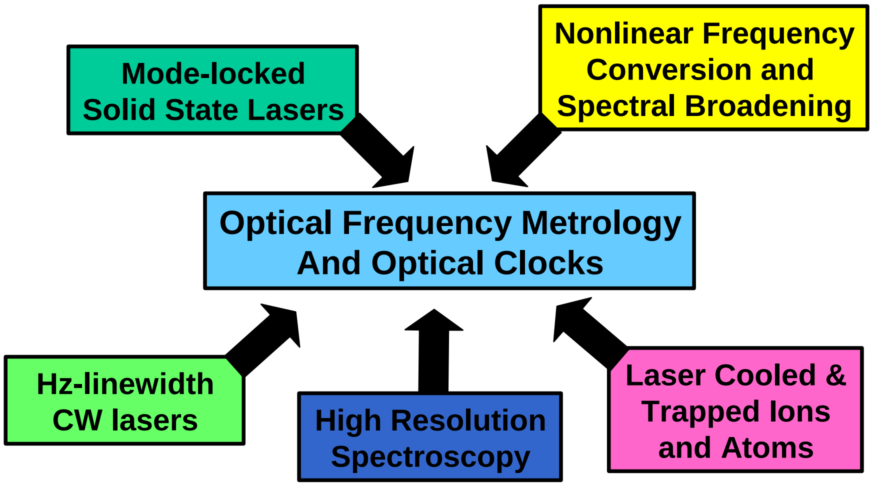

Figure 9-1. The development of optical clocks and optical frequency metrology has arisen out of many different sub-fields of atomic, molecular, and optical physics. While optical clocks have their roots in the precision optical-frequency spectroscopy of cooled and trapped ions and atoms, the emergence of robust mode-locked solid-state femtosecond lasers and nonlinear spectral broadening/conversion techniques has provided new opportunities and invigorated the field as a whole.

have exclusively operated at microwave frequencies, with the 9,192,631,770 Hz ground-state hyperfine transition in cesium-133 being the internationally recognized definition for the SI second. The jump from microwave to optical frequencies has been jointly fueled by advances in several branches of atomic and optical physics, including laser cooling and trapping, precision spectroscopy, frequency stabilized lasers, nonlinear fiber optics, spectral broadening and frequency conversion, and femtosecond mode-locked lasers (see Figure 9-1). Over the past 30–40 years, developments in precision optical frequency spectroscopy with highly stabilized lasers have formed a solid basis for the surge of activity in optical frequency metrology since 1999. Pioneers of this field [1-8] recognized that the spectroscopy of cooled and trapped atoms with narrow linewidth lasers would be valuable for precision measurements in general, and even 20–30 years ago, prototype "optical clocks" were being developed [6, 9]. While it was clear that clocks operating at optical frequencies had much potential for improved stability and accuracy, a longstanding problem in the development of such clocks has been a reliable and straightforward means of counting the extremely rapid optical oscillations. Solutions to this counting problem involved harmonic frequency chains [10, 11] at several national facilities or an alternative approach based on interval bisection [12]. However, both of these techniques required significant resources and were rather complicated, meaning it was unlikely they could ever become widespread and reliable. In 1998-99, this scenario was rapidly changed after the group of T. Hänsch demonstrated that mode-locked femtosecond lasers could play an important role in optical frequency metrology [13, 14]. With the subsequent introduction of highly nonlinear optical fibers that could broaden the femtosecond laser spectrum to bandwidths greater than one octave, all of the pieces rapidly came together for the implementation of a simple and elegant single-step phase-coherent means of counting optical frequencies [15, 16].

As illustrated by Figure 9-1 and this brief introduction, the science and technology that underlie optical clocks are rich and varied. Several excellent reviews and introductions to this topic already exist [17-22] and others are found in this volume itself. Instead of attempting to provide a comprehensive review, we will focus on the role that femtosecond lasers play in emerging optical clocks and in low noise frequency synthesis. We will begin by presenting the main components that make up an optical clock and highlight some of the most relevant detail of the atomic references. The bulk of this chapter will then describe in detail the role of the femtosecond laser in optical clocks and low-noise optical and microwave frequency synthesis. This description will include various methods by which the femtosecond laser can be controlled as well as some tests that have searched for inaccuracies in such laser systems when they are used in precision frequency measurements. An overview of the array of different femtosecond laser synthesizers that have been developed since 2000 will be presented. And in conclusion, we will discuss recent experiments focused on the distribution of optical clock signals through optical fiber networks using mode-locked lasers.

1.1 Basic components of optical clocks

Most common clocks consist of two major components: an oscillator that produces periodic events or “clock ticks,” and a counter for accumulating and displaying each tick [23]. For example, the swing of a pendulum provides the periodic events that are counted, accumulated, and displayed via a set of gears driving a pair of clock hands. Similarly, in a quartz watch, the mechanical vibrations of a small quartz crystal are electronically detected, accumulated, and displayed to generate time.

However, because of the influence of environmental fluctuations, the nature of an artifact-based standard makes it very difficult for a quartz oscillator to remain reproducible at a level of 1 μs per day. Atomic clocks add a third component that solves this problem: the resonance of a well-isolated atomic transition that is used to control the oscillator frequency. This is where precision spectroscopy becomes vital to ensure that the evolution of a clock signal is ultimately governed by fundamental quantum physics. If the frequency of the oscillator is controlled to match the transition frequency between two unperturbed atomic states (i.e., the oscillator is locked to the atomic transition frequency), then the time generated can have the desired long-term stability and accuracy. For an atomic clock based on a microwave transition, high-speed electronics count and accumulate a defined number of cycles of the reference oscillator to mark a second of time. As sketched in Figure 9-2, the basic concepts are the same for an atomic clock based on an optical transition at a much higher frequency. In this case, the oscillator is a stabilized laser with its frequency locked to an optical transition. However, no electronic device exists that can directly count the extremely rapid optical oscillations. Until very recently, devices that could divide an optical frequency down to a countable microwave (called "frequency chains") were complicated, large-scale devices requiring significant resources for operation [11, 24, 25]. However, as will be described in detail in Section 3, the advent of femtosecond-laser-based clockwork greatly simplifies this problem of directly counting the optical frequency.

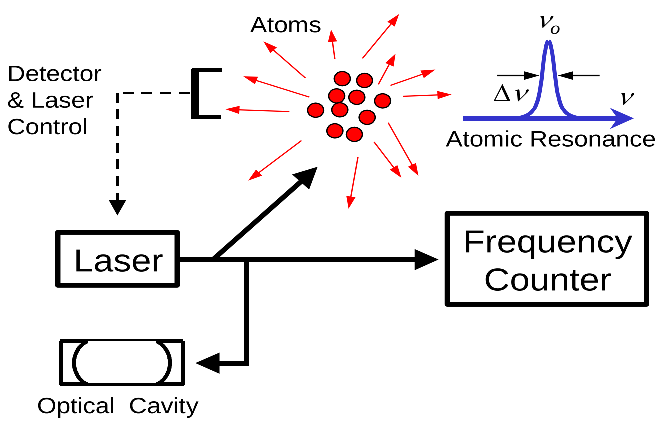

Figure 9-2. Schematic diagram of an optical clock. A laser is first stabilized to a Fabry-Perot optical cavity that provides a means to narrow the laser linewidth leading to good short-term stability. The center of a narrow resonance in an appropriate atomic sample then provides a stable reference to which the frequency of the laser can be steered. Once its frequency is locked to the center of the atomic resonance, a predetermined number of optical cycles are counted to mark a second of time.

1.2 Uses of optical atomic clocks

While timekeeping is an important aspect of our everyday lives, most of us do not really care or need to know the exact time to much better than ~60 seconds at any point in our daily routines. Computer networks that record financial transactions and reservations may require timekeeping that is ~100 times better than this, but still at the modest level of millisecond accuracy. However, many technological applications, such as communication and navigation systems (e.g., the Global Positioning System, GPS [26]), are much more demanding, requiring clocks that are stable and accurate at the level of 10–100 nanoseconds per day. The most accurate clocks are ultimately referenced to natural resonances in atoms, and they provide a unique tool for the scientific exploration of basic atomic structure and the physical interactions between atoms and their environment. It is necessary for timekeeping purposes to have clocks that are accurate at the level of 100 picoseconds per day (1 part in 10^{15}), but it is also arguable that clocks with such performance are equally valuable as basic science research tools. The next generation optical clocks that are currently being developed will move beyond this already astounding level of performance, with projected uncertainties approaching 1 part in 10^{18}. In other words, if such a clock could run indefinitely, it would neither gain nor lose one second in the lifetime of our universe.

It is valid and important to ask why scientists, or society for that matter, might want still better clocks. A brief look back in the history of timekeeping provides some idea of how we might expect emerging optical clocks to benefit society in the coming decades. In the 1950s when microwave atomic clocks based on cesium (and other atoms and molecules) were first developed, the situation was not so different from where we find ourselves today. The first atomic clocks were quickly recognized as being significantly better than the existing mechanical and quartz-based clocks, yet at the same time, they were mainly a tool of scientific interest. At that point, few people would imagine that just 30 years into the future, a constellation of satellites containing cesium and rubidium atomic clocks would circumnavigate the globe providing accurate time and position to all people below. The GPS and its constituent atomic clocks are now an integral part of our lives. While it is difficult to extrapolate to the next 50 years, it is fully expected that optical clocks will find numerous applications in advanced communications and navigation systems. For example, the very stable optical clock ticks may be especially useful for tracking and communication between satellites and spacecraft in the much greater expanses beyond our planet. Indeed, stable and precise lasers are envisioned to provide both laser distance ranging (length metrology) and time keeping simultaneously for satellites [27].

For the near future, it is already clear that optical clocks and optical frequency metrology will provide interesting and new scientific avenues to study our universe—pushing the limits on tests of the most fundamental physical laws to new levels. Of particular interest is the continued application of optical frequency standards in spectroscopy and the improved determination of the fine structure constant α and the Rydberg constant R_∞ [14, 28]. As measurement accuracy improves, metrologists may find themselves in the unique position of being able to observe physical "constants" evolve in time [29]. Indeed, laboratory tests on the possible divergence of clocks based on different atomic transitions already provide some of the most stringent constraints of the variation of α [30, 31]. Experiments of fundamental importance for which precision clocks/oscillators are of value include searches for variations in the isotropy of space, a preferred reference frame, and Lorentz and Charge-Parity-Time (CPT) symmetry violation [32]. Following the recent trapping of cold antihydrogen at CERN [33], in the coming years it may be possible to compare optical clocks based on the 1S–2S transitions in both hydrogen and anti-hydrogen [34]. Such measurements would provide precisions tests of the fundamental symmetry between matter and antimatter [35].

1.3 A brief history of the development of optical clocks

In the decade following the invention of the laser, it became clear to many researchers that a stable optical oscillator had great potential for precision measurements including time keeping. The reader is referred to the references [24] and the contributions of Chapter 8 in this volume for a more detailed history of the development of optical frequency standards and optical clocks. Here, we present only a brief overview of this topic touching on a few of the most important advances.

Not long after the invention of the laser, efforts were in place to stabilize its frequency to the center of its Doppler-broadened gain profile. From that point forward, an increasingly sophisticated set of tools was used to improve the resolution and fidelity with which a laser's frequency could probe, and be stabilized to, a narrow spectroscopic feature in an atom, ion, or molecule. The proposal to use the divided-down output of a laser-based oscillator suitably locked to a quantum reference as a time and/or frequency standard was put forth by pioneers such as Javan [36] and Dehmelt [1]; however, the difficult task of counting optical cycles was a formidable challenge that could be addressed by only a few research groups and national laboratories with sufficient resources. This effort resulted in the so-called harmonic frequency chains that employed successive nonlinear steps to multiply the frequency of a cesium-based primary standard up to the infrared and then, ultimately, the optical domain. At NIST (then National Bureau of Standards), a cesium-referenced frequency measurement of the 88 THz methane-stabilized helium-neon laser was combined with a krypton-referenced wavelength measurement of the same stabilized laser to provide a hundredfold improvement to the measurement of the speed of light. This was a conclusive demonstration of the value of laser-based frequency standards, and soon thereafter, the speed of light was defined as a fixed value such that an optical frequency standard could also serve as an absolute length standard (e.g., $\lambda = c/f$). The result of this advance cannot be overstated. The ubiquitous 633 nm helium-neon laser, with its *frequency* stabilized to a known transition in iodine, has served for the past 20 years as the *length* standard for wide variety of industrial and scientific applications.

The widespread use of atomically stabilized lasers for time standards has been more difficult to realize. Beyond the serious problem of counting optical cycles, this is due, at least in part, to the continued improvements and excellent performance accessibility of cesium-based microwave frequency standards (the latest developments of cesium standards around the world can be found in Reference [37]). Nonetheless, in the 1980s at least two groups developed operational "optical clocks" based on the 88 THz methane-stabilized helium-neon laser and conventional frequency chains [6, 9]. The fractional short-term stability for these devices was already in the 10^{-12} regime where the limitation arose from the microwave standard against which the optical clock was compared. In the 1990s laser cooling and trapping techniques were introduced and refined frequency chains were developed to connect optical transitions to the microwave domain. Noteworthy in this respect were the efforts surrounding Ca (456 THz) [10] and Sr^+ (445 THz) [11] at the Physikalisch-Technische Bundesanstalt of Germany and the National Research Council of Canada, respectively.

2. THE ATOMIC REFERENCE

Every optical atomic clock needs a quantum reference, which is most commonly an electronic transition in an atom, molecule, or ion. As might be expected, many factors go into the choice of a specific reference, and atoms, molecules, and ions all have advantages and disadvantages relative to one another. In this section, we will first describe the benefit one gains by choosing an optical reference as opposed to a microwave reference. Then we will spend some time describing references based on single trapped ions, an ensemble of laser-cooled and trapped atoms, and molecular gases. Specific examples from laboratory systems will also be provided.

When the center-of-mass motion of the quantum reference for the clock is well controlled, the coherent interaction time in both optical and microwave domains can be the determining factor of the spectral resolution. However, the optical part of the electromagnetic spectrum provides ~100,000 times higher operating frequencies. Therefore, the resonance quality factor, $Q = \nu/\nu_0$, of an optical clock transition is expected to be significantly higher than that available in the microwave domain. (Here, ν_0 is the atomic transition frequency and $\Delta\nu$ is the transition linewidth). A superior Q factor provides a more stable frequency standard, and is essential for making a more accurate standard as well. This is seen in the Allan deviation $\sigma_y(\tau)$ which provides a convenient measure of the fractional frequency instability of a clock as a function of the averaging time τ [38]. For an oscillator locked to an atomic transition, the Allan deviation is given as

$$\sigma_y(\tau) \approx \left\langle \frac{\Delta\nu_{rms}}{\nu_0} \right\rangle_\tau \approx \frac{1}{\pi \cdot Q}\sqrt{\frac{T}{\tau N}}\,, \tag{1}$$

where $\Delta\nu_{rms}$ is the measured frequency fluctuation, N is the number of atoms, and T is the cycle time (i.e., the time required to make a determination of the line center) with $\tau > T$. This expression assumes that technical noise is reduced to a sufficiently small level that the quantum-mechanical atomic projection noise is the dominant stability limit [39]. In this limit, $\sigma_y(\tau)$ decreases as the square root of the averaging time for all clocks, so a tenfold decrease in the short-term instability leads to a hundredfold reduction in averaging time τ to reach a given stability and uncertainty. This point is particularly important if one ultimately hopes to reach a fractional frequency uncertainty of 10^{-18}, which is the anticipated level for ion-based optical clocks. In this case, an extremely small short-term instability, i.e., $\sigma_y(\tau) \leq 1\times10^{-15}\tau^{-1/2}$, is clearly desirable to avoid inordinately long averaging times (see Figure 9-3).

Since $\sigma_y(\tau)$ scales as $1/Q$, all else being equal, the shift from microwave to optical frequencies should improve the short-term stability by a factor of 10^5. Thus, with a linewidth of about 1 Hz and 10^6 atoms detected every 0.5 seconds, theoretically, these systems could support an instability $\sigma_y(\tau) \approx 5 \times 10^{-19}\tau^{-1/2}$. This simplistic estimate ignores significant complications that will degrade the performance. Nonetheless, it promises that in the years ahead there will be plenty of room for improvement using optical frequency standards. The current status and near-term prospects of the Allan deviation for optical and microwave frequency standards is shown in Figure 9-3

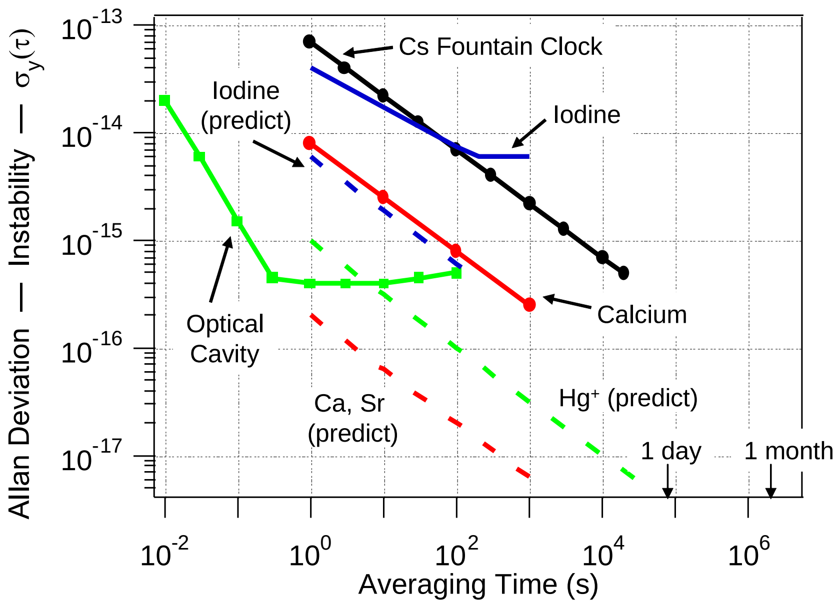

Figure 9-3. Fractional frequency instability as characterized by the Allan deviation of a microwave frequency standard (cesium fountain clock) and several promising optical frequency standards. The plotted curves are based on measured data, except where predicted curves are given as dashed lines.

2.1 Single ion references

Trapped ions, particularly single laser-cooled ions, have numerous advantages as optical frequency standards and clocks [1, 40]. Perhaps one of their most important attributes is the nearly ideal independence between the trapping potential for the center-of-mass motion and the internal atomic level structure that provides the clock reference. Importantly, ions can be confined in an rf trap and laser-cooled so that the amplitude of the residual motion is much less than the optical wavelength of the probe radiation (the Lamb-Dicke limit). This nearly eliminates the velocity-dependent Doppler broadening and shifts associated with motion of the ion relative to the probing radiation. In a cryogenic environment, the ion is nearly unperturbed by atomic collisions, and the effects of blackbody radiation are also very small. The storage time of a single ion in a trap can be months; hence, the probe interaction time is usually constrained by other technical issues such as the laser coherence time or, fundamentally, by the natural lifetime of the transition under study. This permits extremely high-Q resonances to be observed. All of these factors are critically important for the achievement of the highest accuracy.

The technical challenges of making an optical frequency standard based on a single ion are formidable but single-ion standards have now been achieved in a handful of laboratories around the world. At NIST, an optical-frequency standard based on a single trapped $^{199}Hg^{+}$ ion is being developed. The performance of this standard is immediately competitive with the performance of the best microwave standards and has the potential to surpass those standards in terms of stability, frequency reproducibility, and accuracy. For this standard, a single $^{199}Hg^{+}$ ion is trapped in a small rf Paul trap (≈1 mm internal dimensions) and laser cooled to a few milliKelvins using 194 nm radiation. A highly stabilized dye laser at 563 nm with a linewidth of less than 1 Hz is frequency doubled to 282 nm (1,064 THz) to probe the clock transition [41]. Measured linewidths as narrow as 6.7 Hz on the 282 nm transition have been reported [42]. For an averaging time τ in seconds, the projected instability of an optical frequency standard using a single Hg^{+} ion is $1 \times 10^{-15}\tau^{-1/2}$, and fractional frequency uncertainties approaching $1\text{x}10^{-18}$ seem feasible.

One apparent difficulty of modern optical standards is the requirement that suitable transitions for laser cooling, fluorescence detection, and the clock itself be present in the same atom or ion. While some atoms have good clock transitions, their cooling and detection transitions might be less than ideal. Wineland et al. have proposed an efficient solution to this problem in the case of ion-based clocks that involves simultaneously trapping a “clock” ion and a “logic” ion [43]. Using techniques developed for quantum computation applications [44], the logic ion would provide the functions of sympathetic cooling and detection, leaving more flexibility in choosing the best clock ion with a narrow, unperturbed and accessible transition. This approach is currently be implemented at NIST using $^{27}Al^{+}$ as the clock ion and $^{9}Be^{+}$ as the logic ion.

2.2 Neutral atom references

Some neutral atoms also have narrow optical transitions that are relatively insensitive to external perturbations and are thus attractive as optical frequency standards. Neutral atoms have some advantages and disadvantages relative to ions. Using the well-established techniques of laser cooling and trapping, they are fairly easy to confine and cool to low temperatures. However, in contrast to ions, the trapping methods for neutrals perturb the atomic energy levels, which is unacceptable for use in a frequency standard. To avoid the broadening and shifts associated with the trap, neutral atoms are released from the trap before the clock transition is probed. The atoms fall from the trap under the influence of gravity and expand with low thermal velocities (typically a few cm/s). The resulting atomic motion brings with it limitations in accuracy (and even stability) that are associated with velocity-dependent frequency shifts. Two of the more troublesome effects are the limited observation time and the incomplete cancellation of the first-order Doppler shift associated with wave-front curvature and wave-vector mismatch. Reduced observation times limit the line Q, the stability, and the accuracy. However, neutral atoms do have at least one significant advantage: large numbers of atoms can be used, producing a large signal-to-noise ratio (*S/N*) in a short time and the potential for exceptional short-term stability.

The atomic Ca optical frequency standard [45, 46] is one of the most promising and extensively studied cases. It has a 400 Hz wide clock transition at 657 nm ($^1S_0 – {}^3P_1$) that is reasonably immune from external perturbations. It is readily laser cooled and trapped, and it is experimentally convenient because the relevant transitions are accessible with tunable diode lasers. Cooling and trapping of about 10^7 Ca atoms to mK temperatures can be accomplished in a magneto-optic trap (MOT) with frequency doubled diode laser tuned to the 423 nm $^1S_0 – {}^1P_1$ transition. With the cooling radiation turned off, an injection-locked and stabilized diode laser at 657 nm (456 THz) then probes the clock transition with the separated excitation method of optical Ramsey-Bordé spectroscopy. Optical fringes with high signal-to-noise ratio are observed using shelving detection on the cooling transition. With this technique, the present Ca standard can provide short-term fractional frequency instability of about 4×10^{-15} in 1 s of averaging. Second stage cooling on the narrow $^1S_0 – {}^3P_1$ to temperatures ~10 μK has been achieved with the aid of 552 nm light that quenches (i.e., depopulates) the long-lived $^3P_1$ state [47]. The 10-μK temperatures reduce velocity-related systematic shifts, and it appears that uncertainties at or below 10^{-15} will be attainable with such a system.

Ultimate neutral atom-based systems with high accuracy will demand a stringent separation between the external degrees of freedom (controlled by the trapping potential) and internal level structure (clock transitions), similar to that obtained with single trapped ions. A crucial step towards high reproducibility and accuracy is to confine neutral atoms in the Lamb-Dicke regime, while at the same time limiting the effect of the confining potential to only the external degrees of freedom [48]. The fermionic isotope of ^{87}Sr has a nonzero nuclear magnetic moment (I = 9/2) that gives rise to magnetic substructure in both the ground and excited states [49]. Moreover, ^{87}Sr possesses a doubly forbidden J = 0 to J = 0 clock transition, $^1S_0$ (F = 9/2) – $^3P_0$ (F' =9/2), which has a ~1mHz linewidth (corresponding to an intrinsic line Q of $4.3x10^{17}$) and is expected to be highly insensitive to external electromagnetic fields and collisional shifts [50]. The Sr atoms are first cooled in two stages to temperatures < 1 μK [51]. These pre-cooled atoms can then be efficiently transferred into a far-off-resonance optical lattice trap where sideband cooling can bring the atoms to the motional ground state. Using a magic wavelength (~800 nm) for the latter trap ensures that both the ground and excited ($^3P_0$) state experience the same energy shift anywhere inside the trap. The sensitivity to the light polarization is reduced because of the fact that J = 0 for both clock states. The ^{87}Sr $^1S_0$ (F = 9/2) – $^3P_0$ (F' = 9/2) transition can then be interrogated free from broadening and shifts due to Doppler, recoil, and trapping potential related shifts.

2.3 Molecular references

The abundance and relatively convenient gas-cell spectroscopy of molecular resonances make them attractive references for optical clocks as well. Indeed, infrared transitions in molecules such as CH_4, CO_2, and OsO_4 were some of the first references explored [25, 52] and continue to be used in several research programs around the world [53]. Their natural linewidths range from a few megahertz to below a kilohertz, limited by molecular fluorescent decay. Useable linewidths are usually ≥10 kHz due to the transit of molecules through the light beam. Transitions to higher levels of these fundamental rovibrational states, usually termed overtone bands, extend these rovibrational spectra well into the visible with similar ~ kHz potential linewidths. Until recently, the rich spectra of the molecular overtone bands have not been adopted as suitable frequency references in the visible due to their small transition strengths. However, with one of the most sensitive absorption techniques, which combines frequency modulation with cavity enhancement, an excellent *S/N* for these weak but narrow overtone lines can be achieved, enabling the use of molecular overtones as standards in the visible [54].

Perhaps the most widespread example of a molecular reference is the iodine molecule. The narrow-linewidth I_2 transitions in the visible wavelength region have provided excellent cell-based optical frequency references for laser stabilization. Frequency-doubled Nd:YAG/$^{127}I_2$ at 532 nm has proved to be one of the best portable optical frequency standards with compact size, reliability, and high stability (< 5×10^{-14} at 1 s) [55]. To reach a better frequency stability, it is useful to explore I_2 transitions towards the dissociation limit at wavelengths below 532 nm where the natural linewidths decrease at a faster rate than the line strengths. The systematic variation of the I_2 transition linewidths within the range of 498–532 nm has been measured, with the linewidth decreasing by nearly sixfold when the wavelength is decreased from 532 nm to near the dissociation limit [56]. The high *S/N* results indicate that I_2 transitions in the wavelength range 501–532 nm hold great promise for future development of optical frequency standards, especially with the advent of all-solid-state Yb:YAG lasers. One exciting candidate is the 514.67 nm standard that has a projected instability $<10^{-14}$ at 1 s [57].

Table 9-1. Some of the promising optical frequency references for emerging and future optical clocks. Presently achieved values of instability and uncertainty are listed, while the projected values are given in parentheses. The research institute is given in the last column.

	Frequency (THz)	1-s Fractional Instability	Fractional Uncertainty	Institute
Ions				
Hg^+	1064	3×10^{-15} (1×10^{-15})	1×10^{-14} ($<1\times10^{-17}$)	NIST
In^+	1267		2×10^{-13}	MPQ
Sr^+	445		5×10^{-15}	NRC, NPL
Al^+	1124			NIST
Yb^+	688		9×10^{-15} ($<1\times10^{-17}$)	PTB
Yb^+	642		2×10^{-12}	NPL
Neutral Atoms				
H	2466		2×10^{-14}	MPQ
Ca	456	4×10^{-15} ($<2\times10^{-16}$)	2×10^{-14} ($<1\times10^{-15}$)	PTB, NIST
Sr				U. Tokyo, JILA, SYRTE, LENS
Yb				KRISS, NIST
Molecules				
OsO_4	29		3×10^{-13}	Paris Nord
CH_4	88			Lebedev, NPL, PTB, Novosibirsk
I_2	563	4×10^{-14}	9×10^{-12}	JILA, PTB, NMIJ, BIPM

2.4 Local oscillator requirements

While it is necessary to start with a suitable atomic transition, whether it be in an ion, atom, or molecule, as diagramed in Figure 9-2, another important element of an optical clock is the local oscillator. For an optical clock, the local oscillator is comprised of a continuous wave (cw) laser that has its emission spectrum narrowed and stabilized to an isolated high-Q Fabry-Perot optical cavity. The optical cavity must have sufficiently good short-term stability to permit interrogation of a narrow spectroscopic feature in the quantum reference (i.e., ion, atom, or molecule) with a good *S/N*. For example, in the case of the single Hg^+ ion, the optical transition linewidth is just a few hertz wide, which requires a local oscillator with a sub-hertz linewidth. Significant effort has been invested for the past few decades in the reduction of the laser linewidth down to this sub-hertz level [41, 58]. A key aspect in this development has been improved techniques for the mechanical and thermal isolation of the Fabry-Perot optical cavity from the surrounding laboratory environment [41]. A more recent advance in this evolution focuses on reducing the acceleration sensitivity of the cavity through geometrical design and by careful choice of how the cavity is supported.

3. FEMTOSECOND LASER-BASED OPTICAL FREQUENCY SYNTHESIZERS

The possibility of using a mode-locked laser as a tool for optical frequency metrology was first demonstrated with picosecond lasers by Hänsch and co-workers in the late 1970s [59]. The essence of this original idea was to use the comb of frequencies emitted from a mode-locked laser as a precise “optical frequency ruler.” The spacing of the tick marks of such an optical frequency ruler is given by the repetition rate f_r at which pulses are emitted from the mode-locked laser, while the differential phase shift $\Delta\phi_{ce}$ between the pulse carrier and the pulse envelope each round trip determines the overall offset of the comb elements $f_0 = f_r\,(\Delta\phi_{ce}/2\pi)$. The relationship between these two parameters and the nth element of the optical frequency comb is given by the simple expression

$$\nu_n = nf_r + f_0 \qquad (2)$$

This technique lay largely dormant until experiments in 1998 demonstrated that the frequency comb associated with a femtosecond mode-locked laser could be readily controlled and provided a more versatile and precise method for counting optical frequencies than existing technologies [13]. In fact, in just a matter of years, the new femtosecond comb technology has fully replaced laboratory efforts that existed for decades; it is now widely accepted that mode-locked femtosecond lasers will play a critical role in the next generation of atomic clocks based on optical frequencies [46, 60, 61]. Moreover, due to the simplicity and relatively low cost, the techniques and tools described here can be implemented in university and industrial research labs, which has resulted in a variety of interesting and exciting new avenues of research [62-64].

The absence of robust solid-state femtosecond lasers likely contributed to the twenty-year delay between the first conceptual experiments and practical implementation of the frequency comb concept. Although the femtosecond dye laser was developed throughout the 1970s and 1980s, it was not until the 1990s that robust and high-power femtosecond lasers based on titanium-doped sapphire (Ti:sapphire) were perfected [65, 66]. (Chapter 2 of this volume presents a detailed account of development of solid-state mode-locked laser technology.) For comb generation, it was natural that femtosecond Ti:sapphire would become the source of choice. When combined with nonlinear microstructure and tapered fibers [67], octave-spanning spectra could be generated. A few representative Ti:sapphire laser configurations and resulting spectra are shown in Figure 9-4. It is important to realize that the array of frequency modes given by Equation (2) reside beneath the broad spectral envelopes shown in this figure (which were recorded with a low resolution grating spectrometer). As will be described in further detail below, such a simple means for generating very broad spectra allows for the straightforward measurement and control of f_0 in addition to providing a network of useful comb lines spanning the visible and near-infrared.

In this section, we will not dwell on the actual generation of the frequency comb from a femtosecond laser. The reader is referred to several excellent review articles on this topic [17, 18, 21], in addition to the other chapters in this book. Instead, we will explain the details of how a femtosecond laser is actually used in an optical clock and for low noise frequency synthesis. We first spell out some of the specific metrological considerations that arise in the use and choice of a femtosecond laser for optical clocks and synthesis applications. Next, we discuss the “mechanical” details of actually using and controlling a femtosecond laser. We will then present what is currently known about the achievable noise properties of the laser itself. Finally, while Ti:sapphire femtosecond lasers continue to be the most widely used options for optical clocks and frequency metrology, fiber-based femtosecond laser sources and alternative solid-state lasers that can be efficiently pumped offer significant advantages in terms of size and cost. We will spend the last part of this section looking at these sources and the benefits they might have for future optical clocks.

3.1 Considerations in designing a femtosecond comb for use in an optical clock

The first experiments [13, 15, 16] verifying the valuable contribution of femtosecond lasers to optical frequency metrology all employed femtosecond lasers that had been developed over the previous decade for experiments in ultrafast science [66]. While such lasers worked very well for the initial proof-of-principle experiments, and continue to be used in many cases, they lacked other desirable qualities for frequency metrology. The ultimate femtosecond laser for optical frequency metrology and optical clocks is likely to be a continuously evolving device, but nonetheless, in this section we attempt to lay out some of the desirable characteristics of these devices as they relate to their use in frequency metrology and optical clocks.

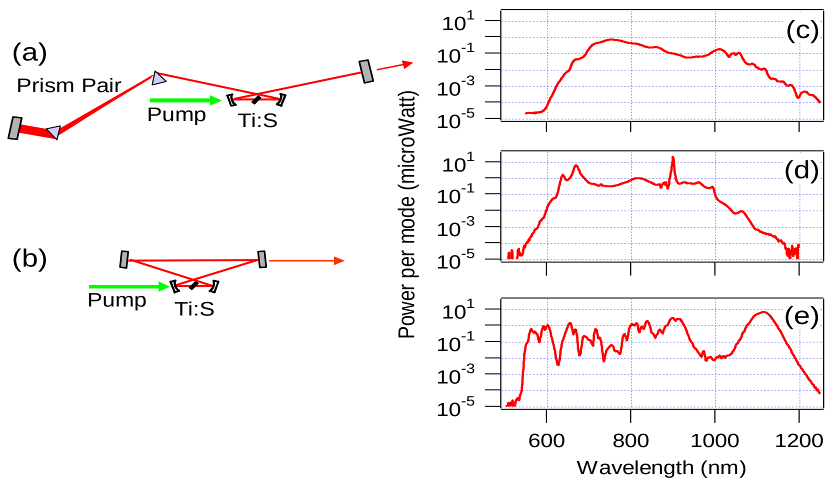

Figure 9-4. Common Ti:sapphire femtosecond lasers used for frequency metrology and their output spectra. (a) Conventional linear cavity design employing a combination of chirped mirrors and prisms produces the broadband spectrum shown in (c). The ring laser shown in (b) uses only chirped mirrors and can produce the spectrum show in (d). The spectrum shown in (e) is produced using either a laser (a) or (b) in combination with nonlinear microstructure optical fiber.

Repetition rate: There are a few factors that drive the consideration of the best repetition rate. The most obvious feature is that for a constant average output power from the laser, the mode comb of a high repetition rate laser will have a proportionately higher power per mode. This is a clear advantage for frequency domain applications where we rely on a good *S/N* in the heterodyne beats between a cw laser and an individual tooth of the comb itself. However, one cannot take this point to the extreme. For example, if we again assume a constant output power, as the repetition rate increases, the power per pulse decreases. This is an important consideration if one is to rely on nonlinear processes to broaden the optical spectrum to an octave. As a rule of thumb, with the typical microstructure fibers used with Ti:sapphire lasers around 800 nm, one needs roughly a few hundred picojoules of energy in a ~50 fs pulse to obtain an octave of spectral broadening. This can be achieved with a 1 GHz repetition rate laser, but given the same average power, it is not likely to be possible if the laser could operate with a 10 GHz repetition rate.

Photodetectors & electronics: Since many uses of the femtosecond laser comb ultimately involve a connection to the rf/microwave domain, the photodetectors that provide this connection as well as the subsequent electronics that are used must also be a consideration. While photodetectors and microwave electronics with bandwidths up to ~50 GHz are commercially available, devices that operate much above 10 GHz tend to be more costly, more difficult to use, and less sensitive. This last factor can actually result in a loss of *S/N* that one had hoped to achieve precisely by choosing a higher repetition rate in the first place. This can be particularly true with the photodetector that is used to detect the optical clock output at f_r, where extremely high *S/N* is required to generate stable electrical signals [68]. Another important electronics-related consideration surrounding the choice of repetition rate is the frequency one might ultimately choose for connecting the f_r output of the femtosecond laser to existing rf/microwave sources and standards. The best choice of low-noise electronic synthesizers that would divide or multiply f_r up or down to common frequencies of 5, 100 or 9,192.631770 MHz will certainly be a consideration for the most demanding applications.

Size, simplicity & robustness: While size, simplicity, and robustness are not necessarily synonymous, it is often true that a smaller device of simple construction can be more robust. For example, small size lends itself to improved temperature stability, which directly impacts the operation of any femtosecond laser and the required dynamic range for the various servo controls. This is a point in favor of higher repetition rate systems, or at least systems with smaller footprints. When it comes down to the actual design of the laser, this factor would also tend to favor the use of chirped mirrors or fully integrated optics (such as a fiber laser) over the use of prisms for dispersion control. The reliable generation of octave-spanning spectra directly from the laser is also a significant simplification as it allows one to eliminate sensitive microstructured nonlinear fibers from the apparatus.

Spectral coverage: A study of the different atoms, ions, and molecules used in emerging optical-frequency standards (see Table 9-1) reveals that many of the atomic transitions of interest fall in the 500–1100 nm range (or harmonics thereof). Thus, Ti:sapphire-based femtosecond laser combs centered at 800 nm are a good starting place and direct continuum generation or subsequent broadening in nonlinear microstructure fibers provides overlap with most of the wavelengths that are presently of interest. While nonlinear microstructure fibers have performed amazingly well in all experiments to date, they present challenging technical limitations on the ability to control the femtosecond laser over long periods. Coupling of the light into their ~1.7 μm diameter cores is difficult to maintain for long times at a level where an octave of spectrum is attained. An attractive alternative is recently developed femtosecond lasers that directly emit octave-spanning spectra suitable for self-referencing [69-71]. These lasers have slightly less bandwidth than is typically achieved from nonlinear microstructure fibers; however, initial experiments indicate the broadband lasers can be much more robust [72, 73]. A full understanding and optimization of these extremely broadband lasers is yet to emerge, but we anticipate they will play a very important role in optical clocks, perhaps reducing the need for nonlinear microstructure fibers.

Noise: Perhaps the most important consideration for any synthesizer is its residual or excess noise. More specific details on the presently achieved noise levels will be presented below, but from a general design point of view, there are important considerations that should be pointed out with regards to systems that employ nonlinear microstructure fibers. First, it is important to construct a femtosecond laser with robust and stable mechanical configurations and environmental shielding. Understanding the laser dynamics and noise sources is also fundamental to design and implementation of stabilization feedback loops [74, 75]. Because of its highly nonlinear nature, microstructure fiber can significantly amplify both technical and fundamental shot noise on the input light [76, 77]. With careful attention, the most significant aspects of the technical noise can be reduced; however, the shot noise on the laser output will always be present and potentially represents a more formidable problem. Of particular significance is the finding that longer pulses and chirped pulses generate more noise when launched into nonlinear microstructure fibers [77, 78]. For example, 75 fs pulses containing a few nJ of energy can make a stunning continuum stretching from 450 to 1400 nm, but excess broadband noise makes it nearly useless for precision frequency metrology.

3.2 Frequency synthesis with a femtosecond laser

The basic function of the femtosecond laser in an optical clock is to provide a phase-coherent link between the uncountable optical reference frequency and the more accessible microwave domain. Using a mechanical analogy, we can imagine the femtosecond laser to be a one-step reduction, or multiplication, gear. Alternatively, one can discuss the femtosecond laser as being an extremely broadband optical frequency synthesizer, with general properties not so different from commonly used rf and microwave synthesizers. While the mechanical analogy is straightforward conceptually, the synthesizer picture is the more useful one as it allows us to employ rigorous characterization techniques developed in the rf and microwave domains.

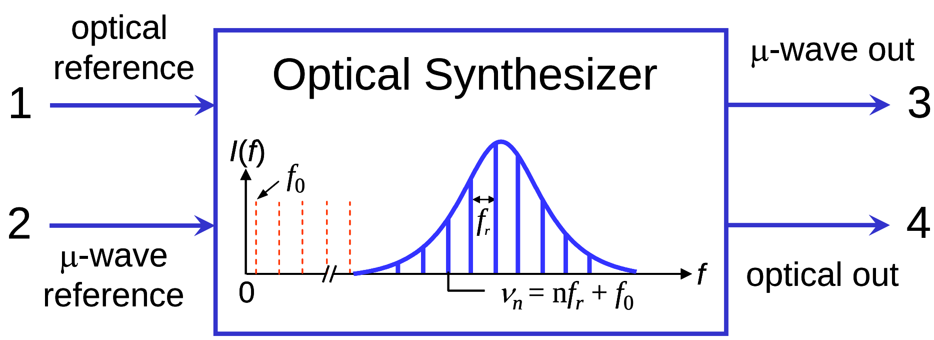

Figure 9-5. The femtosecond laser-based optical frequency synthesizer.

As shown in Figure 9-5, the femtosecond laser-based synthesizer can employ either a microwave (μ-wave) or optical input reference that is phase coherently linked to an optical or microwave frequency at the output. These operations can all be carried out and understood by considering the underlying frequency comb structure associated with the femtosecond laser. The first uses of the femtosecond laser-based synthesizer involved supplying a microwave reference at port 2 and obtaining coherently related optical frequencies in the form of the optical comb of Equation (2) at port 4 [15, 16, 79]. This path relies on the femtosecond laser to effectively multiply f_r by approximately a factor of 10^6 to reach the optical domain. In principle, the multiplicative factor could be a much smaller value, on the order of 10^2 or 10^3, to instead yield microwave frequencies in the hundreds of gigahertz (or even terahertz range). This potentially interesting use of the synthesizer (ports 2 $\rightarrow$ 3) has so far received less attention. When used to down-convert an optical frequency input to a microwave output (ports 1$\rightarrow$ 3), the frequency of the output is given by the repetition rate f_r or one of its harmonics [46, 60, 61]. The synthesizer can also function to effectively synthesize new optical frequencies that are displaced from the input optical reference by a freely chosen offset plus-or-minus harmonics of f_r (ports 1→ 4) [80].

3.2.1 Methods of control

Control of f_0: To accomplish all of these operations, it is required that at least two of the three variables given in Equation (2), in addition to n, be measured and controlled. For example, one might control f_r and f_0 and use ν_n as an output. Alternatively, Equation (2) can be inverted such that one tooth (n = N) of the comb ν_N and f_0 are controlled and f_r is then the output. Whereas it is relatively straightforward to measure the repetition rate f_r by monitoring the pulse train with a sufficiently fast photodetector, it is more challenging to measure the offset frequency f_0 of the comb. In principle, once f_r is measured, it is sufficient to use a cw laser with well-known frequency to measure and stabilize f_0. While this might be practical in some situations, it doubles the number of required frequency standards (one for f_0 and a second for f_r) required for using the synthesizer and potentially limits the performance.

A more elegant means of measuring and controlling f_0 involves using nonlinear frequency generation to compare different regions of the frequency comb [81]. As shown in Figure 9-6(a), if the laser spectrum covers more than one octave, then the comb elements at the low-frequency end of the spectrum can be doubled in a nonlinear crystal and subsequently heterodyned against the high-frequency components of the comb to yield $2(nf_r + f_0) - (mf_r + f_0) = f_0$ when n = m/2. Spectral broadening of low power Ti:sapphire lasers to more than an octave is accomplished through self-phase modulation (SPM) in microstructure fibers [67]. Such an approach was first demonstrated by Jones et al. [16] and continues to be the most common manner in which to measure f_0. This so-called "ν-to-2ν" self-referencing scheme has also been demonstrated with low-power Cr:LiSAF [82] and Er-doped fiber lasers [83]. It has also been shown that "ν-to-2ν" self-referencing can be accomplished with Ti:sapphire lasers that directly emit an octave-spanning spectrum [71, 84]. As summarized by Telle et al. [81], it is not necessary to have an octave-spanning spectrum from the femtosecond laser to measure f_0. The price to be paid, however, is that extra steps of nonlinear conversion must be employed. For example, with a spectrum spanning 2/3 of an octave, f_0 can be obtained through the comparison of the second harmonic and third harmonic of separated portions of the optical spectrum, e.g., $3(nf_r + f_0) - 2(mf_r + f_0) = f_0$ when n = 2m/3 [72, 84, 85].

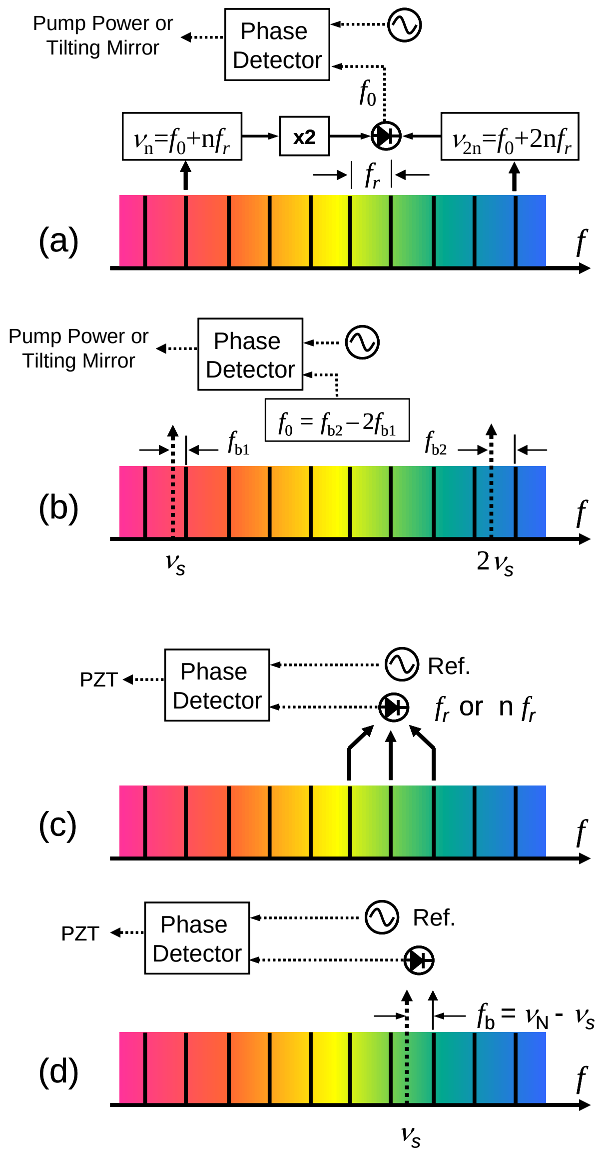

Figure 9-6. Techniques for measuring and controlling f_0 (a, b) and f_r (c,d). See text for details.

Once f_0 has been measured, it is typically phase-locked to a stable rf source or to a rational fraction of f_r itself. Such a phase-lock requires an actuator inside the laser cavity that provides a differential change to the group and phase velocities of the circulating field. Two common modes of control have so far been used. The first employs modulation of intracavity laser power [79, 86], and the second (which is applicable in linear cavities employing prism pairs) involves the tipping of one of the cavity mirrors [18, 87]. While the merits and physical mechanisms of these differing techniques remain a topic of discussion, both have been successfully implemented to reduce the residual fluctuations of f_0 to the millihertz level relative to a stable reference. When f_0 is phase-locked to D.C. or a harmonic of f_r, precise stabilization has the consequence of maintaining the relative carrier-envelope phase at a fixed value for times approaching one hour [88]. This has enabled the subsequent amplification and use of such lasers for experiments in phase-sensitive extreme nonlinear optics experiments (see, for example, Reference [64] and Chapter 10 of this volume).

Control of f_r: As already mentioned, f_r can be detected with a sufficiently fast photodetector. The rf spectrum of the resulting photocurrent consists of a series of harmonics of f_r, one of which can be filtered and phase-locked to a low-noise rf source (Figure 9-6(c)). In this case, it is common to use a piezo-mounted cavity mirror as an actuator for changing f_r. A more careful consideration of Equation (2) highlights some of the difficulties and disadvantages of controlling f_r in the rf/microwave domain as just described. Because the optical frequency ν_n scales as the multiplicative mode number n, the spectral density of phase noise on the rf reference source to which f_r is phase-locked is multiplied by a factor of n^2. For a mode in the optical domain with $n \approx 500{,}000$ (f_r = 1 GHz), this implies that the phase noise of the rf reference is increased by 115 dB when it is effectively multiplied up to the optical domain using the femtosecond laser comb. A high-quality quartz-based rf reference at 1 GHz might have a typical noise floor of –110 dBc/Hz, but when it serves as the reference for f_r, there will be nothing remaining of the phase-coherent carrier in the optical mode ν_n. This is similar to the well-known problem of "carrier collapse," which places extreme demands on the rf reference (as well as all intermediate electronic components) for f_r.

There are, however, a few ways to minimize this phase-noise multiplication problem. First of all, one can rely on the relatively good short-term stability of the femtosecond laser itself. On time scales less than ~1ms, the noise of a typical Ti:sapphire laser has been measured to fall below that of most high quality microwave sources [20, 68]. This means that if one controls f_r relative to a microwave reference, a control bandwidth of ≤1 kHz is all that is required to remove the low-frequency thermal and acoustic noise of the mode-locked laser. Use of higher bandwidth will simply transfer the noise of the microwave reference to f_r, which will subsequently be multiplied up to the optical comb elements. Although the observed optical linewidth may still be on the order of 0.1–1 MHz, one generally finds that the fractional stability of the optical comb elements can be equal to the fractional stability of the microwave reference. Of course, if a microwave reference for f_r with lower phase noise is available (such as a cryogenic sapphire microwave oscillator), then one can potentially narrow the optical linewidth further.

However, in the near future it is likely that the lowest-noise electromagnetic oscillators of any kind will be based on cw lasers referenced to stable Fabry-Perot optical cavities and/or atomic or molecular resonances. In fact, cw lasers with sub-hertz linewidths have recently been demonstrated [41], allowing the use of such a source to control f_r in the optical domain.

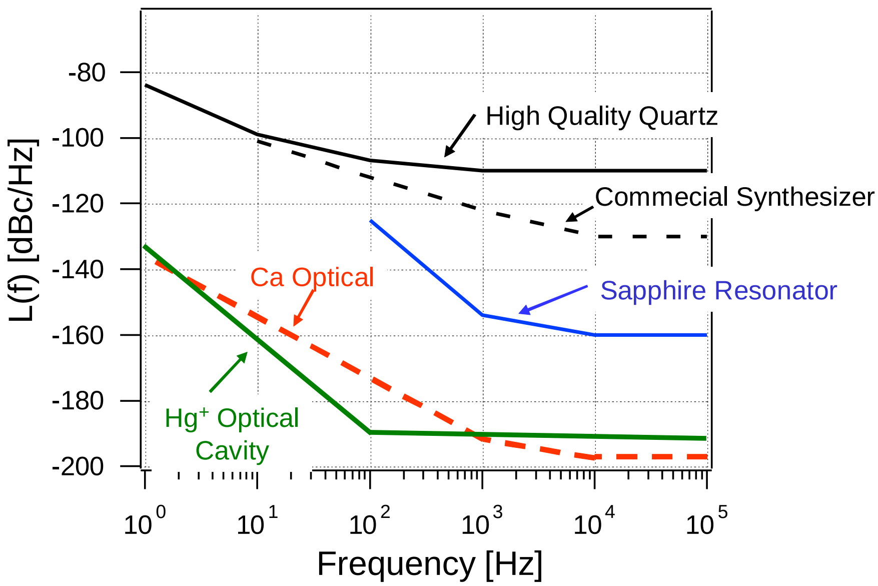

Figure 9-7. Phase noise of various oscillators and synthesizers at 1 GHz. The quartz curve corresponds to a high-quality 5 MHz oscillator and assumes noise-free multiplication to 1 GHz. The commercial synthesizer is residual synthesizer noise that would be added to the reference oscillator (typically quartz). The sapphire resonator curve represents a state-of-the-art commercial product with its frequency divided down from 10 GHz to 1 GHz. The dotted Ca curve is a projection of what should be achievable, and the Hg^+ optical cavity is estimated from experimental data [41].

As seen in Figure 9-6(d), this is typically done by making an optical-offset phase-lock between one tooth (mode N) of the femtosecond comb (ν_N) and the stable cw reference laser ν_S. The resulting expression for f_r is

$$f_r = (\nu_S - f_0 + f_b)/N\,, \qquad (3)$$

where f_b is the arbitrary (yet phase-locked) beat frequency between ν_S and ν_N. This expression illustrates that any frequency fluctuations in ν_S are divided by N before appearing in f_r (spectral density of phase fluctuations are divided by N^2). Since both f_0 and f_b are phase-locked microwave frequencies <1 GHz having fluctuations well below 1 Hz, the fluctuations of ν_S will likely dominate the noise of f_r. Assuming a perfect frequency divider, the anticipated limits to the phase noise spectral density of a sub-hertz laser (Hg^+ optical cavity) and the Ca optical standard are compared to other low-phase-noise sources in Figure 9-7. Clearly, the phase noise of the optical standards offers significant potential in this regard. At present, the noise from the femtosecond laser and the optical detection of f_r (1 GHz) has been measured to be -120 dBc/Hz at 1 Hz offset, decreasing to ~ -155 dBc/Hz at 1 kHz [89]. It has been shown that the phase noise on the optical comb of the femtosecond laser itself can be significantly less than this level [90], leading to the conclusion that the dominant noise sources for optical-to-microwave conversion with the femtosecond laser synthesizer are introduced in photodetection of f_r and subsequent electronic processing and measurement.

Alternative control methods: Instead of frequency-doubling a portion of the octave spanning comb spectrum as described above, one can use a cw laser and its second harmonic to access and control f_0 and f_r. [91]. This is diagramed in Figure 9-6(b). Measurement of the heterodyne beat between the cw laser frequency, ν_s, and the comb line n gives $f_{b1} = \nu_s - (nf_r + f_0)$ and between the second harmonic of the CW laser and comb line $2n$ gives $f_{b2} = 2\nu_s - (2nf_r + f_0)$. Mixing the beats with appropriate weighting factors gives $f_{b2} - 2f_{b1} = 2\nu_s - (2nf_r + f_0) - (2\nu_s - 2(nf_r + f_0)) = f_0$, which represents another detection scheme of f_0. On the other hand, it is now also possible to establish a direct phase coherent link between optical and microwave frequencies without the need of stabilizing f_0. The following expression, $f_{b2} - f_{b1} = 2\nu_s - (2nf_r + f_0) - (\nu_s - (nf_r + f_0)) = \nu_s - nf_r$, illustrates this principle. After appropriate processing, this error signal is used to stabilize the phase of f_r coherently to ν_s, thereby producing an output clock signal in the rf domain derived from ν_s [61].

There are several other methods for removing the dependence of the frequency comb on f_0. For example, by using difference-frequency generation (DFG) between different spectral portions of the same frequency comb, a DFG comb independent of f_0 in the infrared spectral region with excellent accuracy and stability can be generated [92, 93]. Furthermore, a DFG comb (again, independent of f_0) tuned to 800 nm might also pave the way to an all-optical carrier-envelope phase-stabilization scheme by re-injecting this DFG comb into the femtosecond laser cavity. Equivalent to the DFG approach, sum-frequency generation (SFG) between an infrared optical frequency standard (e.g., a methane-stabilized He-Ne laser at 3.39 μm) with the long-wavelength portion of a frequency comb yields an SFG comb that can be tuned to spectrally overlap with the short-wavelength portion of the original frequency comb. The resultant heterodyne beat between the two combs of the same origin leads again to a carrier-envelope frequency-independent optical clockwork. We have recently implemented such an optical clockwork based on SFG between a cw optical parametric oscillator (OPO) at 3.39 µm idler wavelength and a custom-tailored mode-locked Ti:sapphire laser with two dominant spectral peaks at 834 and 670 nm. No additional single-frequency lasers phase-locked to the Ti:sapphire comb are necessary.

Finally, it is important to note the clever "transfer oscillator" [94, 95] concept in which a judicious choice of frequency mixings effectively eliminates the noise properties of the *unstabilized* femtosecond laser when it is used to determine the ratio of widely separated optical frequencies or even optical and microwave frequencies. An advantage of this technique is that it allows one to replace the more difficult and relatively slow (<100 kHz) control of the femtosecond laser with more straightforward and faster (~1 MHz bandwidth) phase-tracking oscillators on the various heterodyne beats.

3.3 Testing the synthesizer

As might be expected, the introduction of mode-locked femtosecond lasers into the field of optical frequency metrology was met with some initial questions about how well such a new technology could perform relative to what existed. Such healthy skepticism has led to some interesting and valuable tests of Equation (2). Table 9-2 provides a summary of some of the relevant experimental tests of the femtosecond laser synthesizer. It is not always straightforward to understand precisely what is being tested in some of these experiments, but in general they can all be viewed as placing simultaneous constraints on f_r and f_0.

The most obvious question one can ask about Equation 2 is whether or not the comb of optical frequencies is indeed uniform or evenly spaced. The time domain perspective of how the mode-locked laser functions leads one to the conclusion that this must be the case. An uneven spacing of the modes would imply that different portions of the spectrum experience different roundtrip delays in the cavity. Were this the case, the pulse would rapidly spread and break apart, which is not consistent with the solitonlike operation of mode-locked lasers. The first frequency-domain verification of this operation was offered by the group of T. Hänsch, by comparing a 44 THz wide comb from a femtosecond laser with an optical interval divider, thus confirming the uniformity at the level of a few parts in 10^{18} [13, 96].

While these impressive experiments can also be viewed as a test of the uniformity of f_0, one apparent weakness is that they employed only a single femtosecond laser and did not verify that the actual frequency position of the modes of the comb could be controlled or reproduced at this level.

Table 9-2. Summary of various tests of the femtosecond laser synthesizers

What was tested	**Fractional Uncertainty**	**Systems Used**	**Reference**
Uniformity of f_r	3×10^{-18}	fs laser broadened to 44 THz in standard fiber	[13] [96]
Microwave to optical frequency synthesis	5.1×10^{-16}	ν-to-2ν fs synthesizer with 3.5ν-4ν frequency chain	[79]
Microwave to optical frequency synthesis	1.6×10^{-12}	ν-to-2ν fs synthesizer and harmonic frequency chain	[97]
Optical-to-optical frequency synthesis	4×10^{-17}	Two ν-to-2ν fs synthesizers	[98]
Optical-to-optical ratio and SHG	7×10^{-19}	Single fs laser and cw laser +SHG	[94]
Microwave to optical frequency synthesis	1×10^{-15}	Two ν-to-2ν fs synthesizers	[99]
Uncertainty of DFG relative to fundamental comb	6.6×10^{-21}	Single fs laser and DFG and SFG	[92]
Optical-to-optical frequency synthesis	1.4×10^{-19}	Two ν-to-2ν and two 2ν-to-3ν fs synthesizers	[100]

To minimize the possibility of unknown systematic effects, a better test of the mode-locked laser frequency comb is the comparison of several independent devices. A few tests have been performed using microwave standards to reference both a femtosecond laser-based synthesizer and a more traditional frequency chain or interval divider, which were subsequently compared in the optical domain [79, 97, 99]. The best result in this case was an uncertainty limit as low as 5×10^{-16} [79]. In general, the limit in these cases was imposed by the short-term instability and associated long averaging times [79, 99] or by the uncertainty of the optical standard that was measured [97]. Significantly improved short-term instability can be obtained when the femtosecond laser-based synthesizer is referenced to an optical standard. In an earlier test, two such devices were compared and agreement was found at the level of 4×10^{-17} [98]. More recently, this same test was repeated with four systems, two of which employed octave-spanning spectra generated in nonlinear microstructure fiber and two which generated broad spectra directly from the laser [100]. With improved short-term instability and longer averaging times, a fractional frequency uncertainty limit of near 1×10^{-19} was achieved. Using the "transfer oscillator" technique already discussed above [94], Stenger et al. tested a femtosecond laser comb by measuring the ratio of the frequency of a cw laser to its second harmonic with an uncertainty of 7×10^{-19}.

3.4 Alternatives to Ti:sapphire

In the few years since the introduction of femtosecond lasers into field of frequency metrology, the palette of available sources has already begun to broaden beyond that of Ti:sapphire lasers (see Figure 9-7). The reliability of the mature Ti:sapphire laser has made it the natural place to begin this exciting field, and it is likely that femtosecond laser-based synthesizers employing Ti:sapphire will continue to be used in many applications. However, the present size and cost of the pump laser for Ti:sapphire-based systems has motivated the search for alternative systems. In the future, it would not be surprising to find that femtosecond laser-based synthesizers and optical clocks will be widely used in science and technology and will be commercially available in compact packages similar to today's microwave synthesizers and clocks. The availability of robust, low-priced femtosecond-laser synthesizers will be particularly important for applications in air- or space-borne platforms or widespread applications (such as communications systems) for which cost and rugged packaging are of greatest importance.

To date, some of the promising femtosecond lasers that have produced octave-spanning spectra include diode-pumped Cr:LiSAF [82], a fiber-laser-pumped Cr:Forsterite [101], and an Er-doped fiber laser [83, 85, 102]. Each of these has some advantages and disadvantages relative to Ti:sapphire. For example, both Cr:LiSAF and Cr:Forsterite have more convenient and compact pumping schemes either directly with diode lasers or with a Yb-doped fiber laser. One of the trade-offs here is that these laser hosts are not as broadband and efficient as Ti:sapphire and tend to have worse thermal properties. An Er-fiber laser-based comb generator has a number of advantages over Ti:Sapphire. It can be much more compact, robust, lighter, and power-efficient than a bulk optic solid-state laser system, and would require less alignment. Additionally, it can be easily integrated into a

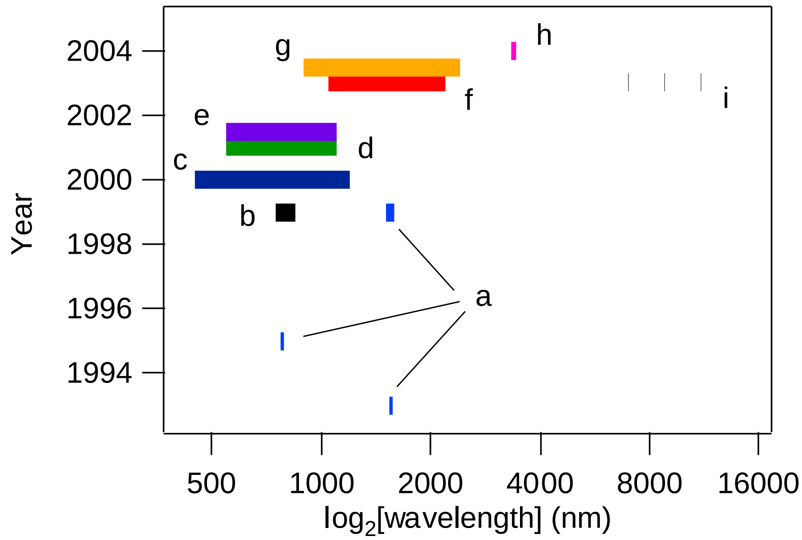

Figure 9-7. Spectral extent of some sources used as combs in frequency metrology. (a) Electro-optic modulator-based comb developed by Kourogi et al. [103]; (b) Ti:sapphire femtosecond laser comb [13, 14]; (c) Octave-spanning femtosecond laser comb generated using microstructure fiber [15, 16]; (d) Broadband spectrum generated directly from a Ti:sapphire laser [69]; (e) Cr:LiSAF femtosecond laser plus microstructure fiber [82]; (f) Octave-spanning comb generated with femtosecond Cr:forsterite and nonlinear optical fiber [104]; (g) Octave-spanning comb generated with Er-fiber laser and nonlinear optical fiber [83]; (h) Offset-frequency-free comb near 3400 nm generated via difference-frequency generation [93]; (i) Tunable frequency comb generated via difference-frequency generation between synchronized Ti:sapphire lasers [105].

telecommunication system in the important 1300–1600 nm regime. However, at this point, the Er-fiber-based systems only operate at repetition rates of 50–100 MHz and can have excess noise that is not fully understood [85].

If wavelength coverage is a concern, one can take advantage of nonlinear frequency conversion external to the femtosecond laser itself. This has been demonstrated for 2ν-to-3ν with Cr:forsterite and fiber laser sources [85, 106]. Another interesting option is to use DFG between two extremes of a Ti:sapphire laser comb to generate a frequency comb further in the infrared [92, 93]. While this provides extra wavelength tunability for the femtosecond source, it also is a means of generating a frequency comb that is independent of the offset frequency f_0. As also shown in Figure 9-7, wide tunability between 7 and 10 microns can also be accomplished with independent Ti:sapphire lasers that are synchronized and potentially phase-locked [105].

4. SIGNAL TRANSMISSION AND CROSS-LINKING

With the advent of optical atomic clocks and the associated superior short-term frequency stability, transfer of signals linked to such clock/frequency standards over an appreciable distance with a minimal loss of stability has become an active subject for research [107, 108]. The stability of the best microwave and optical frequency standards, for times less than ~ one week, can now exceed the capabilities of the traditional transmission systems (i.e., GPS, Two-Way Time transfer [109]) used to distribute these time/frequency reference signals to remote sites. The instabilities in transmission channels can contribute a significant fraction of the overall uncertainty in the comparison of high-performance standards that are not co-located. While improvement in the transfer process over large-scale signal paths remains challenging, researchers have started experimenting with optical fibers as effective means for local networks of dissemination or comparison of time and frequency standards, both in the microwave and optical domains. The attractiveness of this approach lies in the fact that an environmentally isolated fiber can be considerably more stable than open-air paths, especially at short time scales. Furthermore, the same advantages enjoyed by communication systems in optical fiber (e.g., low loss, scalability, etc.) can be realized in a time/frequency distribution system. Active stabilization of fiber-optic channels for the distribution of reference frequencies can also be employed to improve the stability of the transmitted standard. Besides the obvious benefit of more precise time/frequency transfer, an actively stabilized optical fiber network can play a critical role in the implementation of long-baseline coherent interferometry or ultralow timing jitter in particle-accelerator-based novel light sources.

An rf signal can be transferred in an optical fiber network by amplitude modulating a suitable optical carrier (for example at 1.55 μm) used for transmission and then recovering the modulation frequency at the remote end as the transferred reference signal. However, the instability of this rf modulation-based frequency transfer protocol seems to rise to a few parts in 10^{13} at 1-s over a 6.9 km long fiber linking JILA to NIST. Jet Propulsion Laboratory colleagues have demonstrated the rf transfer instability of parts in 10^{14} at 1 s for a 16 km-long fiber under active noise cancellation control [110]. In comparison, direct transfer of a cw laser-based optical frequency standard through the same 6.9 km fiber suffered an instability of a few parts in 10^{14} at 1 s. This instability is further reduced to 3×10^{-15} at 1 s after active optical-phase-noise cancellation is implemented [107]. A femtosecond laser located at the remote end can be phase locked to this incoming cw laser carrier, essentially redistributing the optical frequency to the microwave domain via the mode-locked laser's repetition frequency.

With the development of precision femtosecond comb technology, it appears natural to use a mode-locked source to transfer both optical and microwave reference signals at the highest level of stability. Of course, the first logical step is to extend the precision optical-comb spectral coverage into the 1.5 μm wavelength region, where compact, reliable, and efficient mode-locked lasers exist for fiber distribution networks. This step has been accomplished by tight synchronization between the repetition rates and coherent phase locking of the optical carriers of the 1.5 μm mode-locked laser sources and a Ti:Sapphire-based femtosecond frequency comb, which is used as the clockwork for an optical atomic clock based on the molecular iodine transition. A phase-coherent link between mode-locked lasers requires two distinct conditions to be met [63, 111]. First, the comb spacing of the 1550 nm source ($f_{r,1550}$) must be stabilized to the optical clock's comb spacing ($f_{r,775}$). Second, the two combs' offset frequencies ($f_{0,775}$ and $f_{0,1550}$) must be phase locked together. This latter step requires spectral overlap between the two combs. The optical comb of the 1550 nm source is frequency doubled and compared against the Ti:sapphire comb at a mutually accessible spectral region to generate a heterodyne beat.

We have investigated a number of different types of passively mode-locked lasers in the 1550 nm region, including an erbium/ytterbium-doped waveguide laser [112], an erbium-doped fiber laser [108, 113], and a mode-locked laser diode (MLLD) [75, 114]. Each laser offers at least two control parameters to obtain simultaneous time synchronization and carrier phase-locking. For example, stabilization through the cavity length is a common feature for all these lasers. The carrier-envelope-offset frequency ($f_{0,1550}$) can be tuned in both waveguide and fiber lasers through control of the intensity of their pump lasers. A MLLD, on the other hand, can be tuned with both the injection current and the reverse bias voltage on the saturable absorber. Among the three mode-locked laser sources, the waveguide laser achieved the lowest residual timing jitter with a record-low, root-mean-square relative timing jitter of 14.4 fs integrated from 10 Hz to 375 MHz (the Nyquist frequency), owing to the laser's high-Q cavity and overall gain dynamics. Although the MLLD has a larger rms timing jitter of ~22 fs within the bandwidth of 1 Hz – 100 MHz, it does offer the advantage of a compact size, robust operation, and completely electrical control, along with the potential in device improvements to lower the jitter at high frequencies.

Transfer of optical frequency standards is then implemented by using the stabilized mode-locked laser sources at 1.5 μm that are transmitted through an optical fiber network. We have explored the dual transfer process for both the optical carrier frequency and the rf signal represented by the laser's repetition frequency and to compare the transfer of such an rf signal with that of a modulated optical carrier. When the 1.5 μm mode-locked laser is phase coherently connected to a Ti:sapphire mode-locked laser serving as the clockwork for an optical atomic clock, both the optical carrier frequency and the repetition frequency of the 1.5 μm mode-locked laser are connected to a single optical frequency standard. Therefore a single fiber link allows a user at the remote end to either simply use a fast photodiode to recover the RF reference signal or establish a more elaborate, direct connection to the optical carrier. Simultaneous existence of both the optical phase and the pulse-repetition-rate information would also enable novel measurement capabilities on material dispersion and length determination. The experimental results confirm that with the use of a mode-locked laser at 1.5 μm, the instability of the transfer process for the repetition frequency (rf reference) over the 6.9 km long fiber is nearly the same as that of optical carrier transfer of a cw laser discussed earlier, and is almost an order of magnitude better than that of transfer of a RF-modulated cw laser carrier. Furthermore, it is clear that a pulsed transfer process preserves the stability of the optical carrier as in the cw optical transfer process. Preliminary results on pulsed transfer under active noise cancellation indicate rf transfer instability reaching a few parts in 10^{15} at 1 s. We are currently exploring the utility of such ultrastable frequency transfer for time-domain experiments where the goal is to deliver extremely low-jitter timing signals throughout a fiber network, permitting ultrafast lasers located at remote areas be synchronized together at a precision level of a few femtoseconds within a multi-megahertz bandwidth.

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