Optical Frequency Metrology with Mode-Locked Laser Frequency Combs

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We review the field of frequency comb generation with mode-locked lasers and its application to optical frequency metrology. Essential physics of mode-locked lasers, frequency combs, and non-linear pulse broadening in microstructure crystal fiber is reviewed, and we discuss some landmark experiments.

INTRODUCTION

While light has always been an object of serious study in physics, in the past few decades it has become an increasingly important tool in experimental physics and in technological applications. The ability to make accurate frequency measurements is central to our ability to test fundamental physical theories such as those describing interactions between atoms and photons, and it has enabled us to rely on atomic transitions for our fundamental measurement standards. Arthur Schawlow's advice to his students — "Never measure anything but frequency!" — was predicated on the fact that frequency amends itself to very accurate measurements, as it is much easier to count discrete cycles than measure continuous variable such as length or time [1]. These facts are all predicated on our ability to resolve these cycles, which until recently could only be done outside the realm of electronic frequency counters (good up to $\sim$100 GHz) using very cumbersome frequency chains and frequency interval dividers [2]. Chains and interval dividers, consisting of many non-linear frequency conversion processes, were costly and often quite large, and therefore were only ever assembled at well-funded, stable national laboratories. Nevertheless, they worked well enough to demonstrate the potential of optical frequency measurements, allowing accurate spectroscopy of the hydrogen $1S$-$2S$ transition frequency and giving the most accurate (at the time) measurement of the speed of light in vacuum in 1972, which subsequently allowed the redefinition of the meter in 1983 relative to the speed of light, i.e. c=299792458 m/s exactly [3–5].

The use of atomic clocks relying on microwave transitions, such as the cesium microwave standard, can provide measurements of time with fractional uncertainties below 10^{-15}, but avenues for improving the accuracy of these clocks are nearly exhausted [6]. This kind of fractional uncertainty is certainly adequate for many applications, and is comical overkill in defining a national standard time. However, these uncertainties naturally manifest themselves in any measurements of frequency, time, or length (in view of the above) that we make, which does impede our ability to make optical spectroscopic measurements and test certain theories such as general relativity and those describing slow changes in fundamental constants like α. The linewidths of optical atomic transitions are generated through similar mechanisms and are of similar magnitudes, but the absolute frequencies involved are hundreds of thousands of times greater, yielding far lower fractional uncertainties. Thus measurements of optical frequencies allow us to make measurements of far greater stability than that provided by microwave frequencies, and this presents one of many motivations for developing a more reliable, accurate, and convenient tool for measuring frequencies in the tera- to petahertz range.

In the late 1990's, it became apparent that optical frequency combs provide just this tool. While the first use of mode-locked lasers to make frequency measurements was performed in 1978, the bandwidth of these lasers at the time was not sufficient to allow this method to gain traction [7]. It was the coincidence of the development of femtosecond Ti:sapphire lasers and the ability to broaden and fix the absolute spectrum of mode-locked laser light, both of which occurred in the 1990's, that finally allowed for convenient application of mode-locked lasers in optical frequency metrology. Optical frequency combs generated by spectrally broadened mode-locked lasers allowed a series of landmark experiments to be performed in spectroscopy and frequency synthesis. In this paper we cover the beginnings of the field of frequency comb generation in the late 1990's and early 2000's, discuss the essential physical ideas as they become chronologically relevant, and end with a review of some of the significant experimental results that mode-locked laser frequency combs allowed. We begin with basic mode-locking theory.

MODE-LOCKED LASERS

A laser cavity with broad gain bandwidth can support many oscillating modes — for a typical Ti:sapphire laser cavity, on the order of hundreds of thousands [8]. Typically there will be no relationship between the oscillations in each mode. They will oscillate independently, and the laser output will be broadband, continuous wave light. If some mechanism is put in place to induce phase coupling or mode-locking between the cavity modes, the modes can be made to oscillate phase-coherently, that is, with some relationship between the phases so that the laser output is a series of short pulses rather than continuous wave light that is incoherent across the spectrum. The origin of these pulses is the fact that the electric fields of the phase-locked modes periodically constructively interfere.

Lasers may be mode-locked actively or passively [10]. Active mode-locking is achieved through the introduction of some external signal into the cavity. A simple example of active mode-locking is the use of an amplitude modulator such as a Mach-Zehnder interferometer with an oscillating phase

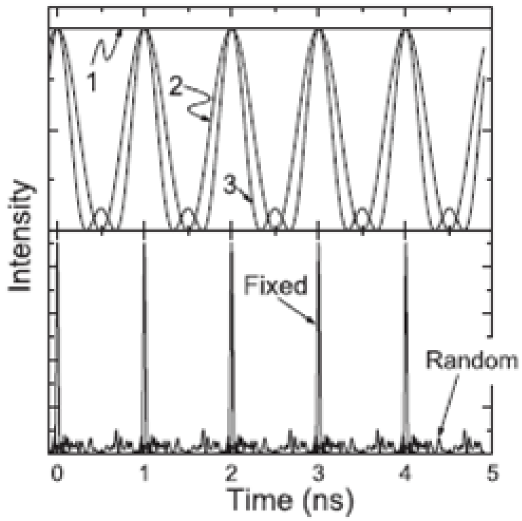

FIG. 1. A simulation of the intensity of light emitted by a laser cavity with one, two, three (top) and thirty (bottom) oscillating modes. In the mode-locked case, the intensity profile is short, high peak pulses, while for the case of random phases, the output is steady CW light that is not coherent from mode to mode. Image [9].

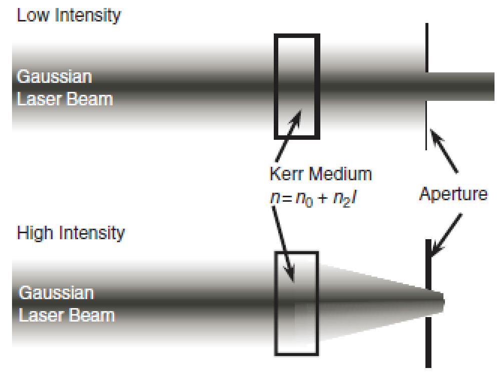

FIG. 3. Kerr-lens mode-locking. Low peak intensity, continuous wave operation experiences higher loss at the aperture. The aperture shown is hard, but soft apertures are also possible, *e.g.* stronger pumping of the gain medium along the optical axis of the cavity. Image [9].

delay on one leg. The phase delay is made to oscillate sinusoidally about zero with a frequency equal to the free spectral range $\Delta\nu$ of the cavity. In the simple picture of one oscillating cavity mode, this oscillating phase gives an oscillating intensity $I(t) = \frac{1}{2} I_o(1 + \cos(\sin(2\pi\Delta\nu\, t)))$ which is equivalent to the generation of sidebands in the spectrum of the laser. These sidebands are equally spaced in units of $\Delta\nu$ about the initial frequency, and their amplitudes decrease with distance from the initial frequency according to the Jacobi-Anger expansion. This leads to many modes oscillating phase coherently. In the realistic case of hundreds of thousands of initially incoherent modes oscillating above gain threshold, this process leads to the modes adjacent to the mode at the center of the gain curve becoming injection locked to that mode, and the injection locking then spills out into the rest of the modes oscillating in the cavity.

Lasers may also be mode-locked passively through the introduction of some element in the cavity that encourages short pulses via intensity-dependent gain or loss. An example of such an element is a saturable absorber, typically a liquid organic dye, which has transmission that increases with intensity. A laser cavity containing a saturable absorber preferentially amplifies short, high peak-power pulses over continuous wave light.

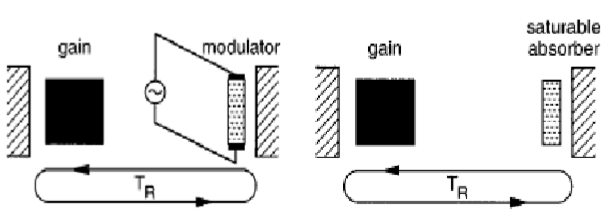

FIG. 2. Schematics of active and passive methods of mode-locking a laser cavity. Images [10].

Another very important implementation of passive mode-locking is Kerr-lens mode-locking (KLM). Kerr-lens mode-locked Ti:sapphire lasers were ideally suited to the generation of the first useful frequency combs because of their broad gain bandwidths and their stable phases. Kerr-lens mode-locking employs a Kerr medium having an intensity dependent index of refraction $n(I) = n_o + n_2 I$. Gaussian cavity modes then experience a Gaussian index profile in the Kerr medium which serves as a lens. Higher intensity light is focused more strongly by this effective lens. An aperture placed near the cavity then serves to pass only highly focused light, and the cavity again preferentially amplifies short, high peak-power pulses. Such an aperture may be hard (see Fig. 3), or soft, *e.g.* stronger pumping of the gain medium near the center of the cavity.

MODE-LOCKED LASERS IN THE TIME DOMAIN: ULTRASHORT PULSES

When a mode-locked laser is output coupled through a partially-transmitting cavity mirror and propagates outside the cavity, it manifests itself as a train of pulses described by the product of a periodic envelope function and an optical carrier frequency (see Fig.4). At a level simpler than a description through knowledge of the full spectral content of the pulse train, it has two important parameters that describe its behavior in the time domain. The first is the repetition rate f_r, the frequency with which the laser pulses arrive at the output coupler, or equivalently the frequency with which the pulses pass some point external to the cavity. This frequency is naturally given by $f_r = v_g/2L$. The second is the pulse-to-pulse

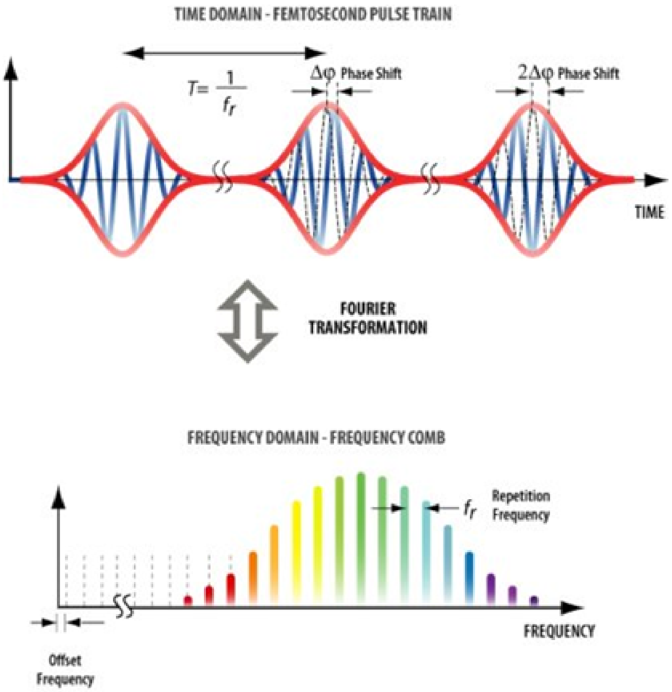

FIG. 4. Time and frequency domain representations of the pulse train of a mode-locked laser.

phase shift $\Delta\phi_{CE}$ of the carrier with respect to the envelope. This originates from the fact that phase and group velocities differ in the cavity so that the pulse envelope and the carrier wave propagate with different speeds through the cavity. Treating the carrier as a standing wave in the cavity reveals that at the output coupling mirror its phase varies as $\omega_c t$, so with a pulse roundtrip time in the cavity of $T = 1/f_r$, the pulse-to-pulse phase shift in the output-coupled pulse train is $\Delta\phi = 2L\omega_c/v_g$. In practice this can always be discussed modulo 2π.

MODE-LOCKED LASERS IN THE FREQUENCY DOMAIN: FREQUENCY COMBS

To understand the origin of the frequency comb, we must consider the Fourier transform of a periodic pulse train with a pulse-to-pulse phase shift, which overall does not necessarily describe a periodic function (i.e. if $\Delta\phi_{CE}/2\pi$ is some irrational number, the more likely scenario). In the simple case $\Delta\phi_{CE} = 0$, the function repeats with period $T = 1/f_r$ and naturally has spectral content at integer multiples of the repetition rate mf_r, the amplitudes of which are given by the specifics of the envelope function.

The case of nonzero $\Delta\phi_{CE}$ is more nuanced. The function $A(t)$ describing the time evolution of a single pulse at a single point in space is an aperiodic function, and has some continuous Fourier spectrum. For a second pulse $A_\tau(t) = A(t-\tau)$ centered about $t = \tau$ we have

$$\begin{aligned}
\tilde{A}(\omega) &= \int \exp(-i\omega t)\, A(t)\, dt \\
\tilde{A}_\tau(\omega) &= \int \exp(-i\omega t)\, A(t-\tau)\, dt \\
&= \int \exp\left(-i\omega(t+\tau)\right) A(t)\, dt \\
&= \exp(-i\omega\tau)\tilde{A}(\omega)
\end{aligned}$$

and we see that each frequency component of the second pulse is phase shifted by an amount $\omega\tau$ from the spectrum of the first. An infinite pulse train $\sum_n A(t-n\tau)$ will thus only have spectral content where all the pulses constructively interfere, i.e. at frequencies $2\pi m/\tau = 2\pi m f_r$. If we include an additional pulse-to-pulse phase shift $\Delta\phi_{CE}$ in our analysis, the frequencies that constructively interfere are given by $\omega\tau - \Delta\phi_{CE} = 2\pi m$, and we see that the spectrum of the infinite pulse train with a pulse-to-pulse phase shift has spectral content at frequencies $2\pi m f_r + \delta$, where δ is a constant frequency offset equal to $f_r\Delta\phi_{CE}$. This discussion follows that of Jones *et al.*, [12].

For a more rigorous frequency domain analysis and the outright Fourier transformation of a periodic pulse train multiplied by a carrier having a pulse-to-pulse phase shift, the reader is referred to Cundiff, 2002 [9]. The outcome of either approach is the same: we see that the spectrum of our mode-locked laser pulse train consists of frequencies spaced uniformly by the repetition rate f_r and having an offset frequency $f_o = \delta/2\pi = f_r\Delta\phi_{CE}/2\pi$. For optical frequency metrology, it is crucial that the repetition rate of the pulse train be a frequency accessible to electronic frequency counters and that the absolute frequencies themselves be optical. The former is achieved by choosing cavities with microwave FSR, and the latter by choosing an appropriate gain medium. Typical KLM Ti:sapphire laser cavities have sufficient gain from roughly 300 to 450 THz and have FSR on the order of fifty to several hundred MHz, and octave spanning pulse trains with repetition rates above 1 GHz directly out of a cavity have been demonstrated [13, 14].

SPECTROSCOPY WITH A MODE-LOCKED LASER

A mode-locked laser cavity may of course have characteristics that drift in time, so for a frequency comb to be useful it is essential to stabilize its frequency characteristics. An early application of a frequency-stabilizied mode-locked laser to spectroscopy was performed by Udem *et al.* in the late 1990's [15]. A KLM Ti:sapphire laser with a repetition rate of 75 MHz was used to bridge the gap between the cesium D_1 line at 335 THz to the fourth harmonic of a stable He-Ne laser, a reference frequency at 354 THz. A schematic of the experimental set-up is provided in Fig. 5. A comb mode is locked via an 848 nm diode laser in series to the fourth harmonic of the He-Ne laser, calibrated by comparison to a cesium microwave standard.

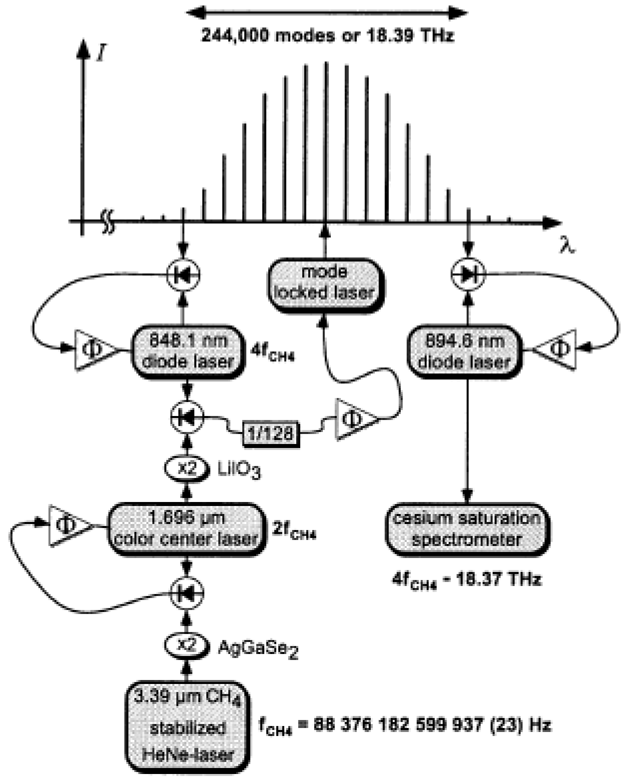

FIG. 5. A schematic depiction of the experimental set-up used to measure the frequency of the Cs D_1 line. Image [15].

This locking is performed by adjusting the position of a piezo-mounted cavity mirror. On passage through a filter cavity, the comb's repetition rate is increased by a factor of 20 to 1.5 GHz, at which point coarse wavelength measurements of the 895 nm diode laser (locked to the Doppler-free center of the cesium D_1 line) and the 848 nm diode laser reveal the relative mode-number separation between the two frequencies. Then simply monitoring the beat between the Cs D_1 frequency and the comb reveals the frequency difference between the transition and the reference. It is important to note that observation of the variation in the beat with the variation in the repetition rate is necessary to remove the sign ambiguity of the position of the transition line relative to its corresponding comb mode. This landmark experiment allowed a new computation of the fine structure constant α and was a convincing demonstration that mode-locked lasers had the potential to be useful tools in optical frequency metrology.

MICROSTRUCTURE OPTICAL FIBER: BROADENING TO SPAN AN OCTAVE

A frequency comb may be broadened in single mode fiber through self-phase modulation. A simple explanation of the mechanism begins by noting that fused silica, commonly used to fabricate optical fiber, exhibits the optical Kerr effect as dis-

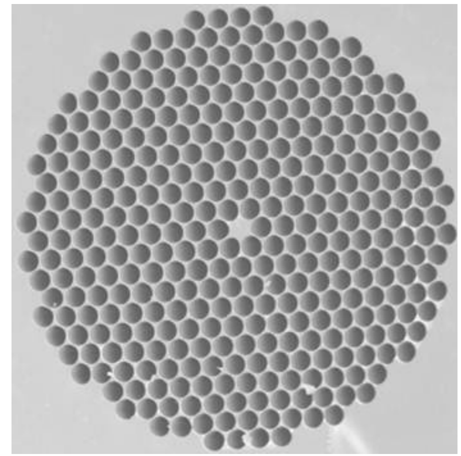

FIG. 6. Microstructure optical fiber. The large index change between the silica core and the honeycombed cladding very effectively confines light to the center of the fiber, providing for sustained intensities of hundreds of GW/cm^2, intensifying non-linear optical effects.

cussed above in the context of Kerr-lens mode-locking [16]. This Kerr effect manifests itself as an intensity dependent index of refraction $n(I) = n_o + n_2 I$. If we consider a pulse passing a fixed point inside an optical fiber with intensity profile $I(t)$, expressing the field as $E(t) = \exp(-i\omega_c t + kL)$ yields a time varying phase $\phi(t) = \omega_c t - \omega_c n(I)L/c$. With the instantaneous carrier frequency given by $d\phi/dt$, we find that there is a time-varying frequency chirp across the pulse given by $\omega(t) - \omega_c = -\omega_c/c \times n_2 dI/dt$, so that for a simple pulse envelope in fused silica fiber ($n_2 = 2.5 \times 10^{-16}cm^2/W>0$), the leading edge of the pulse is red-shifted, while the trailing edge is blue shifted. This broadens the spectrum of the pulse in the frequency domain.

While this spectral broadening is possible in any fiber exhibiting a positive Kerr coefficient, for efficient spectral broadening large phase shifts must be induced, requiring sustained high peak intensities for propagating pulses. So called "microstructure optical fiber" was introduced in 1999 that provided for these high intensities by efficiently confining the propagating modes to a silica core surrounded by a cladding honeycombed with air holes (Fig. 6) [17]. Using this fiber, sustained peak intensities of hundreds of $\mathrm{GW/cm^2}$ have been observed, and the spectra of frequency combs can be broadened to exceed optical octaves.

The ability to broaden combs to more than an octave in bandwidth increases their utility beyond experiments similar to the Udem cesium spectroscopy discussed above, as it allows for more precise control of the frequency characteristics of these lasers. The frequency characteristics of an octave-spanning comb can be locked exactly, yielding an absolute optical ruler. This is done through direct measurement and control of both frequency parameters of the comb, f_r and f_o. Any comb with microwave repetition rate has f_r amenable to electronic measurement and control because the repetition rate may be observed and locked by impinging the comb on

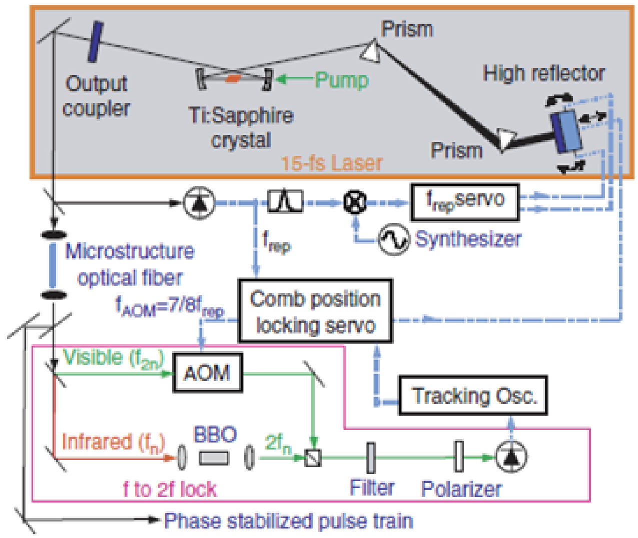

FIG. 7. Experimental set-up for locking the absolute positions of all comb modes through measurement and control of f_o and f_r. These frequencies are locked through cavity adjustments with two piezo-electric crystals. The microstructure fiber and β-barium borate crystal are necessary to generate an octave-spanning comb and frequency double it to observe f_o. Image [12].

a photodetector, measuring the comb's beat with itself, and locking this frequency by adjusting the cavity length as discussed above. However, measurement and control of the offset frequency f_o is only possible for octave spanning-combs.

By beating an octave-spanning comb against its second harmonic, the offset frequency is observed via $2(nf_r + f_o) - (2nf_r + f_o) = f_o$. The offset frequency can then be simply controlled by using two prisms to disperse the comb light onto one of the cavity mirrors and adjusting its angle, thereby imposing a group delay on the oscillating pulse.

An experimental set-up demonstrating application of this technique is given in Fig. 7 [12]. Here the group of Jones *et al.* demonstrates locking the offset frequency and repetition rate of an octave-spanning comb generated by broadening the spectrum of a KLM Ti:sapphire laser with repetition rate of 90 MHz and initial spectral width of 70 nm to span the range from 510 nm to 1125 at -20 dB (Fig. 8). The group demonstrated that locking f_r and f_o is equivalent to locking the pulse-to-pulse phase shift of the pulse train $\Delta\phi$ by measuring the second order temporal cross-correlation (i.e. cross-correlation between pulses i and $i+2$) and demonstrating that it was stable in time. This experimentally confirmed the derived connection between the time and frequency domains $2\pi f_o = f_r \Delta\phi_{CE}$, adding to the confirmation that the mode spacing is equal to the repetition rate, demonstrated in 1999 by Udem *et al.* [18].

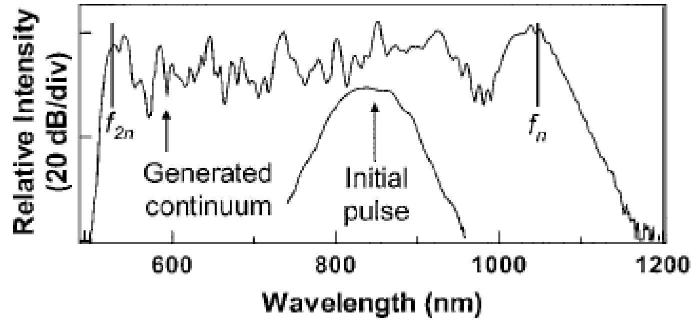

FIG. 8. Spectral broadening of a KLM Ti:sapphire laser for use in the experiment of Jones *et al*. The initial pulse is broadened in microstructure optical fiber to span more than an octave, allowing measurement and control of f_o. Image [12].

OPTICAL FREQUENCY METROLOGY WITHOUT AN OPTICAL REFERENCE FREQUENCY

In addition to permitting the measurement and control of f_o, octave-spanning combs may be used to perform optical frequency measurements without an external optical reference. In 2000, the group of Diddams *et al.* demonstrated the application of a KLM Ti:sapphire laser to compare an unknown optical frequency directly to a microwave standard [19]. The group measured the 282 THz frequency of an Nd:YAG laser locked to a $^{127}I_2$ transition by employing a fiber-broadened, octave-spanning $f_r = 100$ MHz Ti:sapphire laser to measure the frequency difference between the laser and its second harmonic. This measurement technique is depicted schematically in Fig. 9. As in the experiment of the Udem group discussed above, course wavemeter measurements are used to determine the comb modes corresponding to the frequencies f_{1064} and $2f_{1064}$. The frequency of the Nd:YAG laser is then given by $f_{1064} = nf_r \pm (\delta_1 \pm \delta_2)$. As in the Udem experiment, there are initial ambiguities of the signs of the positions of the frequencies relative to their corresponding comb modes. The first sign ambiguity is removed by adding the two beats and determining if their sum remains constant as the offset frequency is allowed to vary, and second ambiguity may be removed by measuring only the beat of f_{1064} with the comb as the offset is allowed to vary.

To emphasize the significance of this result, we note that to measure the same optical frequency difference of 282 THz, an optical frequency interval divider would have required fourteen stages!

FURTHER APPLICATIONS AND FUTURE PROSPECTS

In the years following these developments, a host of other landmark experiments were performed demonstrating the utility of frequency-stabilized mode-locked lasers for optical frequency metrology. To name a few, the measurement accuracy of the In^+ clock transition was improved by more

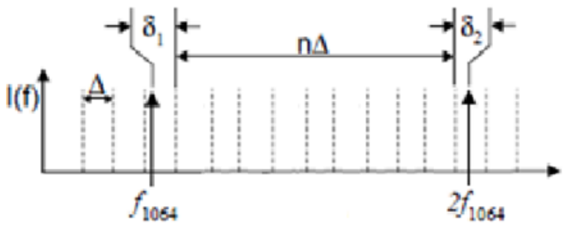

FIG. 9. A schematic depiction of the frequency measurement made by Diddams *et al.* by doubling an Nd:YAG laser. Observations of the changes in beat frequencies δ_1, δ_2 with variation in offset frequency removes the ambiguity in the sign of the relative position of the frequencies f and $2f$ with respect to their corresponding modes.

than two orders of magnitude, an all-optical clock based on a trapped $^{199}\mathrm{Hg}^+$ ion was demonstrated, and the ultraviolet $1S$-$2S$ atomic hydrogen transition was measured at a level which made it the most precisely measured optical frequency ever [20–22]. Another early application was the demonstration of optical frequency generation across 100 THz of bandwidth with a fractional uncertainty of 10^{-19} [23]. In addition to these landmark experiments with mode-locked laser combs, the field of frequency combs has grown in new, exciting directions. Fiber laser frequency combs have been developed, allowing for the commercialization of portable combs for signal generation and spectroscopy. Frequency combs have been extended to the extreme ultraviolet, permitting spectroscopy at even higher resolution, and hinting at the possibility of ultraviolet or even X-ray atomic clocks [24]. Two exciting areas of current research are those of frequency comb generation from monolithic microresonators, which could lead to the development of accurate, chip-based comb spectroscopy and molecular fingerprinting, and the application of combs to astronomical spectrograph calibration, which may allow direct observation of the universe's expansion history, an unprecedented result [25, 26]. These are just a handful of many potential scientific and technological applications as this exciting field continuous to develop.

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