Optical frequency metrology

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Extremely narrow optical resonances in cold atoms or single trapped ions can be measured with high resolution. A laser locked to such a narrow optical resonance could serve as a highly stable oscillator for an all-optical atomic clock. However, until recently there was no reliable clockwork mechanism that could count optical frequencies of hundreds of terahertz. Techniques using femtosecond-laser frequency combs, developed within the past few years, have solved this problem. The ability to count optical oscillations of more than 10^{15} cycles per second facilitates high-precision optical spectroscopy, and has led to the construction of an all-optical atomic clock that is expected eventually to outperform today's state-of-the-art caesium clocks.

For more than a century, precise spectroscopy of atoms and molecules has been crucial in the discovery of the laws of quantum physics, in the determination of fundamental constants, and in the realization of standards for time, frequency and length. The advent of highly monochromatic, tunable lasers and techniques for nonlinear Doppler-free spectroscopy in the early 1970s had a marked impact on the field of precision spectroscopy[1,2]. Today, we are able to observe extremely narrow optical resonances in cold atoms or single trapped ions, with resolutions ranging from 10^{-13} to 10^{-15}, so that it might ultimately become possible to measure the line centre of such a resonance to a few parts in 10^{18}. Laboratory experiments searching for slow changes of fundamental constants would then reach unprecedented sensitivity. An all-optical atomic clock[3] based on such resonances could satisfy the growing demands of optical frequency metrology, fibre-optical telecommunication or navigation. However, until recently, the lack of an optical frequency counter to act as the clockwork mechanism prevented the construction of such a device.

Most spectroscopic experiments still rely on a measurement of optical wavelengths rather than frequencies. Unavoidable distortions in the geometric wavefront have so far made it impossible to exceed an accuracy of a few parts in 10^{10} with a laboratory-sized wavelength interferometer. To obtain highly accurate results it is necessary to measure the frequency of light rather than its wavelength. This is because time can be measured much more precisely than any other physical quantity, and counting the number of cycles in a second is as accurate as the clock that is used to determine the duration of the second. Because the value of the speed of light was defined in 1983 to be precisely 299,792,458 m s^{-1} in vacuum, the conversion between frequency and wavelength can be done without loss in accuracy. But the potential for high accuracy of frequency measurement has been restricted mainly to radio frequencies (up to 100 GHz) for many years, and it has been extremely difficult to extend it to the domain of rapid optical oscillations.

The early approach to this problem made use of harmonic frequency chains. Such chains start with a caesium atomic clock that operates, by definition of the SI second, at 9,192,631,770 Hz (equivalent to the ground-state hyperfine splitting of the caesium atom). This clock defines the frequency at the lower end of the chain from which higher harmonics in nonlinear diode mixers, crystals and other nonlinear devices are generated[4–8]. Phase-locked transfer oscillators are needed after each step, so that a chain traversing a vast region of the electromagnetic spectrum becomes highly complex, large and delicate, and requires substantial resources and considerable efforts to build and operate. This is the reason why only a few harmonic chains have ever been constructed. Another significant drawback of this approach is that the chains are designed to measure just one single optical frequency.

None of the problems associated with harmonic frequency chains has been solved since the first chain was constructed about 30 years ago[4]. In 1998 our laboratory introduced a revolutionary new approach that vastly simplifies optical frequency measurements[9,10]. We showed that the modes of a mode-locked femtosecond laser can be used as a precise ruler in frequency space, as they form a series of frequency spikes called a frequency comb[11,12]. This work has now culminated in a compact and reliable all-solid-state optical frequency synthesizer that requires only a single mode-locked laser, but is nevertheless capable of measuring essentially any optical frequency[13–17]. As a universal optical frequency-comb synthesizer it provides the missing link between optical and microwave frequencies. For the first time, small-scale spectroscopy laboratories can measure or synthesize any optical frequency with extreme precision. Femtosecond frequency-comb techniques have since started to gain widespread use, with precision measurements in Rb (ref. 13), Ca (refs 18,19), CH_4 (ref. 20), H (refs 15,16), Hg^+ (ref. 18), I_2 (refs 14,21–24), Sr^+ (refs 24,25), Yb^+ (refs 24,26) and In^+ (ref. 27).

The same femtosecond frequency-comb techniques are also opening new frontiers in ultrafast physics. Control of the phase evolution of few-cycle light pulses[9,17,28–30] provides a powerful new tool for the study of highly nonlinear phenomena that depend on the phase of the carrier wave relative to the pulse envelope, such as above-threshold ionization[31], strong-field photoemission or the generation of soft-X-ray attosecond pulses by high-order harmonic generation[32].

Femtosecond light pulses

More than 20 years ago, the frequency comb of a mode-locked picosecond dye laser was first used as a ruler in frequency space to measure the sodium 4*d* fine-structure splitting[33]. This route was pursued further in the 1980s (refs 34,35), but the attainable bandwidths were never large enough to make it a widespread technique for optical frequency metrology. Broadband femtosecond Ti:sapphire lasers have existed since the beginning of the

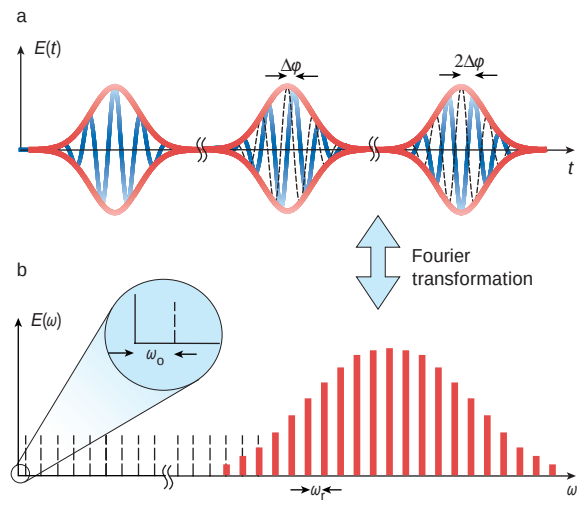

Figure 1 Consecutive pulses of the pulse train emitted by a mode-locked laser and the corresponding spectrum. **a**, As the carrier wave at ω_c moves with the phase velocity while the envelope moves with a different group velocity, the carrier wave (blue) shifts by $\Delta\varphi$ after each round trip with respect to the pulse envelope (red). **b**, This continuous shift results in a frequency offset $\omega_o = \Delta\varphi/T$, which prevents the comb from being comprised of exact harmonics of the pulse repetition frequency ω_r (refs 9,10,29,56,57).

1990s, and we have shown conclusively that such lasers can be crucial in this field[11,12].

To understand the mode structure of a femtosecond frequency comb and the techniques applied for its stabilization, consider the idealized case of a pulse circulating in a laser cavity with length L with a carrier frequency ω_c, as shown in Fig. 1. The output of this laser is a sequence of pulses that are essentially copies of the same pulse separated by the round-trip time $T = v_g/2L$, where v_g is the cavity's mean group velocity defined by the round-trip time and the cavity length. But the pulses are not quite identical. This is because the pulse envelope $A(t)$ propagates with v_g, whereas the carrier wave travels with its phase velocity. As a result, the carrier shifts with respect to the pulse envelope after each round trip by a phase angle $\Delta\varphi$ (Fig. 1). Unlike the envelope function, which provides us with a more rigorous definition of the pulse repetition time $T = \omega_r^{-1}$ by demanding $A(t) = A(t-T)$, the electric field is, in general, not expected to be periodic in time. If the periodicity of the envelope function is assumed, the electric field at a given place outside the laser resonator can be written as

$$E(t) = \mathrm{Re}(A(t)\exp(-i\omega_c t)) = \mathrm{Re}(\Sigma_n A_n \exp(-i(\omega_c + n\omega_r)t)) \quad (1)$$

where A_n are Fourier components of $A(t)$. This equation shows that, under the assumption of a periodic pulse envelope, the resulting spectrum consists of a comb of laser modes that are separated by the pulse repetition frequency ω_r.

Because ω_c is not necessarily an integer multiple of ω_r, the modes are shifted from being exact harmonics of the pulse repetition frequency by an offset that can be chosen to obey $\omega_o < \omega_r$ simply by renumbering the modes

$$\omega_n = n\omega_r + \omega_o \quad (2)$$

with a large ($\approx 10^5$–10^6) integer n. This equation maps two radio frequencies ω_r and ω_o onto the optical frequencies ω_n. Although ω_r is readily measurable, and usually lies between a few tens of megahertz and a few gigahertz depending on the length of the laser resonator, ω_o is not easy to access unless the frequency comb contains more than an optical octave. The intuitive picture given here can even cope with a frequency chirp, that is, a carrier frequency that varies across the pulse. In this case the envelope function becomes complex in value and the comb structure derived above remains valid provided the chirp is the same for all the pulses. Under this assumption, which is reasonable for a stationary pulse train, $A(t)$ remains a periodic function.

The main presumption of equation (2) is that the mode spacing is the same across the frequency comb and that it equals the pulse repetition rate. To be able to use this property for high-precision measurements, one has to make sure that the simple, intuitive picture given above is correct with a high degree of accuracy. Initially, the crucial question was whether pulse-to-pulse phase fluctuations or the large multiplication factor n might destroy the coherence[36] or prevent it from being a comb all together. We first verified that there is indeed a comb of continuous-wave laser frequencies by overlapping the beam from the femtosecond laser with a single-mode diode laser on a photo-detector to observe beat signals between the modes and the continuous-wave laser. To test the mode spacing we have used two laser diodes that were phase locked to distant modes of the comb and used simultaneously as an input for a so-called optical frequency interval divider (OFID)[37,38]. An OFID locks three laser frequencies f_1, f_2 and f_3 to obey the relation $2f_3 = f_1 + f_2$ in a phase-coherent manner. This could be used, for example, to produce an output (f_3) at the precise midpoint of the frequency interval given by two input laser diodes (f_1 and f_2). If the modes are distributed equally on the frequency axis, and if there is an odd number of modes between the two OFID inputs, then one would expect the OFID output to coincide with another mode of the comb. After averaging all our data we concluded that there was no deviation from the perfect regular grid larger than 3 parts in 10^{17} (ref. 11). This experiment was key to the development that followed.

Improving the versatility of frequency measurements

At this point, the frequency comb generated by our mode-locked Ti:sapphire laser could bridge roughly 45 THz. The fourth harmonic of a methane-stabilized He–Ne laser that operated with high accuracy was used as an absolute frequency reference for the femtosecond comb, which allowed us to access new targets. These included the transition frequency of the caesium D_1 line[12], which is needed for a new determination of the fine-structure constant α (refs 39,40). And a sharp resonance in a single trapped indium ion[27] may eventually serve as a precise reference for an optical clock[41]. With the previous measurement schemes it would have been very difficult to measure

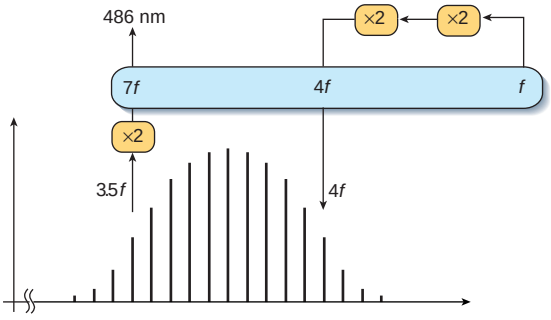

Figure 2 The first direct radio frequency–optical frequency conversion using a femtosecond laser. As explained in the text, the optical frequency interval divider (blue box) fixes the frequency ratios to precisely $7f{:}4f{:}f$. With the frequency quadrupling stage, which was already used to measure other optical transition frequencies relative to a He–Ne reference, the frequency comb fixes the interval $4f - 3.5f = 0.5f$ and therefore f and any other frequency in the set-up.

the frequency gap from the fourth harmonic of the He–Ne reference to these target frequencies, but with the frequency comb this was a straightforward task.

Direct radio frequency–optical frequency link

Combs used as rulers in frequency space can measure large frequency gaps between a precisely known optical reference and an unknown frequency. But to reference the comb directly to a precisely known radio frequency an absolute optical frequency could be mapped onto a frequency difference between harmonics or sub-harmonics of the same laser. In the simplest case this would be the interval between an optical frequency f and its own second harmonic $2f$ (ref. 14). The measurement of the large frequency gap $f = 2f - f$ by a chain of OFIDs or a femtosecond laser has been proposed in refs 37 and 9, respectively. But other intervals can be used as well, as illustrated in Fig. 2, which describes the first experiment of this kind[15,42]. The experimental set-up still uses the somewhat awkward quadrupling stage to create the fourth harmonic $4f$ of the methane-stabilized He–Ne laser. The frequency comb is then used to cross over from $4f$ to ~$3.5f$, and after doubling, a frequency in the vicinity of $7f$ is obtained. The loop is then closed by using an OFID stage that fixes f such that frequency ratios $7f:4f:f$ are precisely fulfilled ($2 \times 4f = 7f + f$). The easiest way to contemplate this is to note that the frequency comb locks $4f - 3.5f = 0.5f$ (44 THz) to an integer multiple of the caesium clock that controlled the mode spacing. By controlling $0.5f$ with a caesium atomic clock, we also know the frequency f and every other frequency in the set-up, including every mode of the comb, with the precision of the caesium clock. Thus the stabilization on methane is replaced by locking all frequencies directly to the caesium clock.

We have used the 486-nm output of this set-up (with some modifications not shown) to measure the absolute frequency of the hydrogen 1*S*-2*S* two-photon resonance, which occurs at the fourth harmonic of this wavelength. The mode spacing was controlled by a caesium atomic fountain clock constructed by André Clairon and co-workers at the Laboratoire Primaire du Temps et des Fréquences (LPTF, France). The outcome was one of the most precise measurements of an optical frequency so far recorded[16], providing a transition frequency value as accurate as 1.9 parts in 10^{14}, limited by the reproducibility of the hydrogen spectrometer. Together with other transition frequencies in hydrogen, our value for the 1*S*-2*S* resonance has resulted in much improved values for the Rydberg constant, which is now one of the most accurately known fundamental constants, and the Lamb shift of the 1*S* state, which now provides one of the most stringent tests of quantum electrodynamics[7].

Increasing the bandwidth of frequency combs

The first absolute measurement of an optical frequency with a femtosecond frequency comb has inspired further rapid advances in the art of frequency metrology. Spectral broadening resulting from self-phase modulation via the intensity-dependent index of refraction in an optical fibre was used to increase the widths of the frequency combs. Even though the dispersion inside the fibre changes the shape of the pulses significantly, it does so in the same way for all pulses. As the above arguments that led to the regular-spaced frequency comb assumed only the periodicity of the envelope function, they should also apply to whatever comes out of the fibre. With newly invented micro-structured photonic crystal fibres (PCF)[43–45], this spectral broadening is so pronounced that the resulting frequency comb can span more than an optical octave, even with the moderate output power from the laser oscillator. In collaboration with Phillip Russell, Jonathan Knight and William Wadsworth from the University of Bath, we have used such fibres to construct a much more compact version of the set-up (Fig. 3).

Alternative approaches to produce large-bandwidth frequency combs have also emerged. Short fibre tapers are now used for this purpose[46]; these are manufactured from standard telecom fibres by drawing them in a flame until their diameter shrinks to about 2 μm.

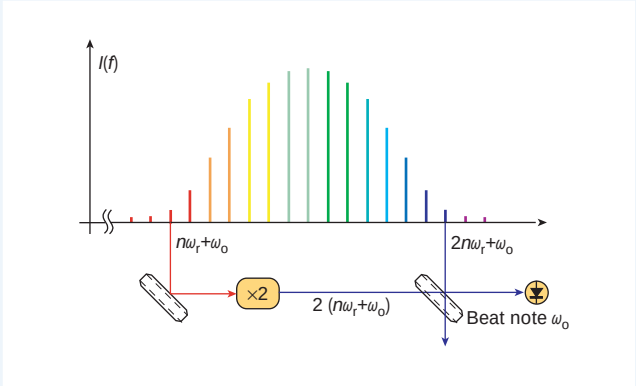

Figure 3 The principle of the single-laser optical synthesizer. A mode with the mode number n at the red wing of the comb and whose frequency is given according to equation (2) by $\omega_n = n\omega_r + \omega_o$ is frequency doubled in a nonlinear crystal. If the frequency comb covers a full optical octave, a mode with the number $2n$ should oscillate simultaneously at $\omega_{2n} = 2n\omega_r + \omega_o$. The beat note between the frequency-doubled mode and the mode at $2n$ yields the offset frequency $2(n\omega_r + \omega_o) - (2n\omega_r + \omega_o) = \omega_o$.

Just like in a PCF, the small core, which consists of the whole fibre, enhances the nonlinear interaction while the evanescent wave around the taper causes a smaller group-velocity dispersion. Another method uses the high intensity inside the laser cavity to efficiently broaden the spectrum[47].

The compact optical frequency synthesizer

With an octave-spanning frequency comb (Fig. 3), we can directly access the interval between an optical frequency f and its second harmonic $2f$, where f could be the 1064-nm line of a Nd:YAG laser. This greatly simplifies the experimental set-up shown in Fig. 2 and has first been reported by John Hall's group at the Joint Institute for Laboratory Astrophysics (JILA) in Boulder, Colorado, and by our group at the Max Planck Institute for Quantum Optics (MPQ) in Garching, near Munich[13–15,17]. The JILA group has also shown that

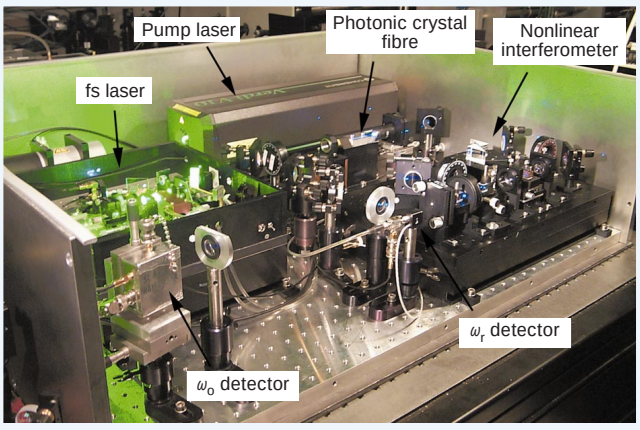

Figure 4 An optical synthesizer in a box. The device consists of one femtosecond laser powered by a green pump laser (Verdi model, Coherent), a photonic crystal fibre and a nonlinear interferometer (as illustrated in Fig. 3). It occupies only 1 m^2 on an optical bench with the potential for further miniaturization. The synthesizer is capable of linking a 10-MHz radio-frequency reference phase coherently in one step with the optical region, and provides a reference-frequency grid across much of the visible and infrared spectrum with comb lines that are separated by 625 MHz. This makes it an ideal laboratory tool for precision spectroscopy. The optical frequency synthesizer is now commercially available from Menlo Systems GmbH (www.menlosystems.com), co-founded by two of the authors (R.H. and T.W.H.).

the additional Nd:YAG laser was not necessary if the red part of the frequency comb itself is to be frequency doubled.

With this set-up, the two radio frequencies that enter equation (1), ω_r and ω_o, are determined. The pulse repetition rate ω_r is simply measured with a photo-detector anywhere in the beam and ω_o, which is smaller in value than ω_r, is derived as explained in Fig. 3. To obtain a stable comb for absolute optical frequency measurements, it is advantageous to phase lock both ω_r and ω_o to a precise radio-frequency reference such as a caesium atomic clock or Global Positioning System-controlled quartz oscillator. The mode number n may be determined by a coarse measurement of the mode in question, for example with a wavemeter. As soon as n, ω_r and ω_o are known, every frequency contained in the comb is known with roughly the same precision. Figure 4 shows the actual apparatus used in our laboratory.

To check the integrity of the broad frequency comb and evaluate the overall performance of the optical synthesizer we have compared it with the previous $7f$:$4f$:f variant. To compare the two we use its output at $4f$ (354 THz) and a mode at 354 THz of the f:$2f$ optical synthesizer. After averaging all data we obtained a mean deviation from the expected beat frequency of 71 ± 179 mHz at 353 THz. This corresponds to a relative uncertainty of 5.1×10^{-16} (ref. 17). Similar precise testings of octave-spanning combs have been performed recently by Scott Diddams and co-workers at the National Institute for Standards and Technology (NIST) in Boulder, Colorado[48].

The all-optical clock

An optical clock consists, like any other clock, of an oscillator that defines the ticks in time and a counter that keeps track of these cycles. In a caesium clock, for example, the oscillations are those of the electron precessing around the spin of the nucleus. An electronic counter advances the second hand every time the counter has completed 9,192,631,770 oscillations. This number was chosen when the SI second was redefined for the last time in 1967. If we consider the history of timekeeping and compare clocks as different as sun dials, pendulum clocks and quartz clocks it is obvious that they get more accurate as the oscillation frequency increases[3]. This is simply because a higher oscillation frequency divides time into smaller pieces.

The caesium 9.2-GHz oscillator has been used since the end of the 1950s. But operating at much higher frequencies has now become possible after tremendous advances in laser spectroscopy in the 1970s (refs 1,2) that resulted in trapped ions[49] and trapped atoms[50] standards in the 1980s. Systematic uncertainties can be reduced to 10^{-18} for some of these standards[41]. When it became possible to count these optical oscillations with harmonic frequency chains in the late 1960s (ref. 4), physicists started to think seriously of running an optical clock. However, working with these counters was so tedious that most of the chains never reached the stage where they could operate continuously, even for minutes. So these chains were used only to calibrate some chosen frequencies, such as iodine- or methane-stabilized He–Ne lasers, which could then be reproduced in other laboratories that could not afford the huge efforts and resources of setting up a harmonic frequency chain. The calibrated lasers were used mostly as wavelength references in interferometers for the realization of the metre, and in some scientific experiments as frequency references.

With the development of the femtosecond frequency synthesizer, a reliable, running optical clock has now come into reach. Owing to its simplicity it can already run for hours and may eventually become a genuine turn-key system.

Currently, the set-up that probably comes closest to an 'optical clock' is operated at NIST[3] and uses a transition at 1,064 THz in a trapped single mercury ion. The measurement of the stability of this clock is limited essentially by comparison with the NIST hydrogen maser ensemble, which is one of the most stable radio frequencies available. Other national standard institutions are making progress in the same direction, including work on Yb^+ by Physikalisch-Technische Bundesanstalt[26] and on Sr^+ by the National Research Council of Canada[25] and the National Physics Laboratory (Teddington, UK)[24]. At MPQ, Herbert Walther's research group are preparing a clock based on a trapped indium ion[41].

We believe that the development of accurate optical frequency synthesis marks only the beginning of an exciting new period of ultra-precise physics, and that optical clocks will open a new window to nature where we can expect new discoveries and phenomena. One example is the quest for natural constants that vary spatially or that would drift slowly in time as the Universe evolves, as discussed by some theoreticians[51,52]. Until now, experimental laboratory tests have not been able to detect such behaviour[53], but recent experiments are believed to provide evidence for a cosmological evolution of the fine-structure constant[54]. Additionally, these clocks may help to refine general relativity, which still poses one of the main problems in physics, as it refuses proper quantization. The precision of the best test is 'only' as good as 7 parts in 10^5 (ref. 55), which might be the reason why small corrections attributable to a quantized theory have not yet been discovered. Finally, industrial applications such as satellite navigation, communication and network synchronization could benefit greatly from this technology. □

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