

Combing time with an optical comb

An Optical Atomic Clock



Piotr Fita, a doctorate student at the Warsaw University Faculty of Physics, focuses on applications of ultrashort light pulses in studying the dynamics of chemical processes

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Technological advancement makes the world an increasingly fast-paced place, requiring timekeeping methods with ever greater precision



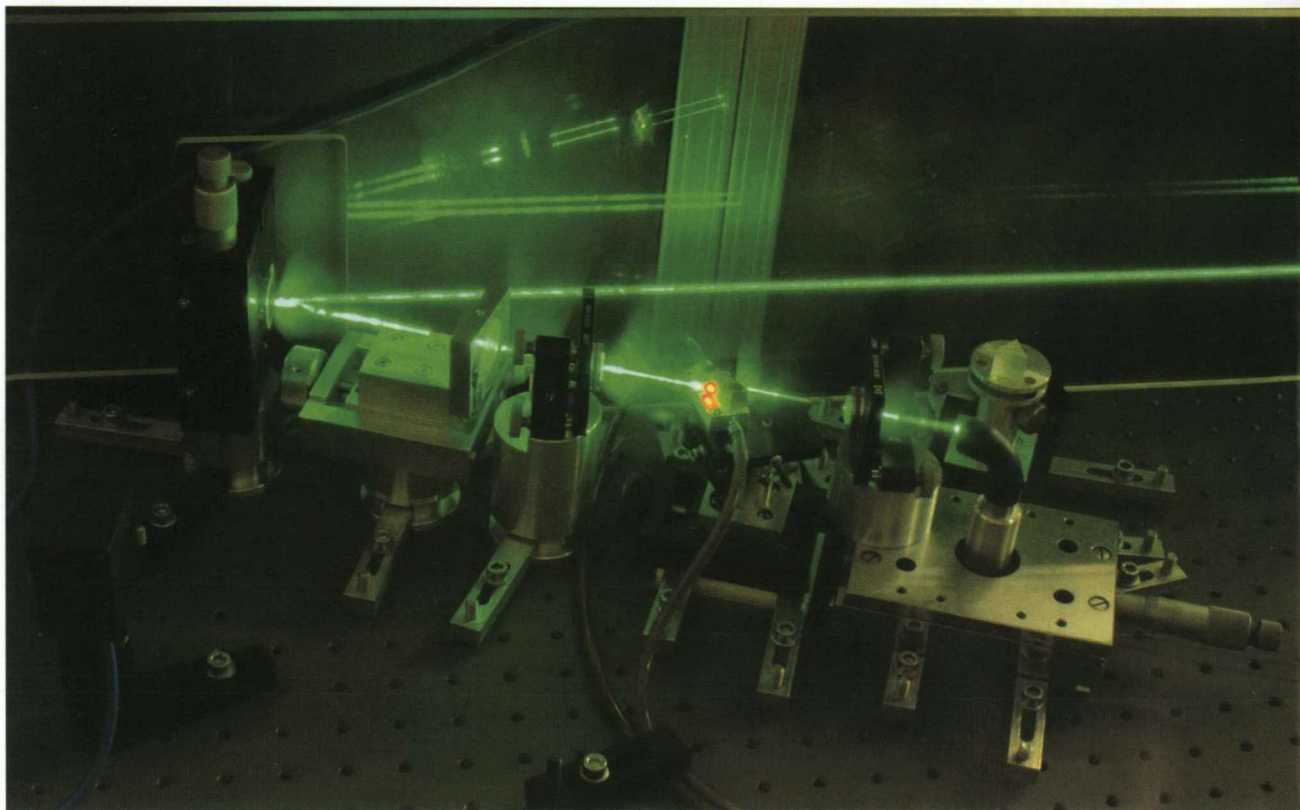
Prof. Czesław Radzewicz works with optics and spectroscopy, especially ultrashort light pulses

It is hard to imagine that nowadays we could be satisfied with knowing the time as loosely as people did several hundred years ago – with an accuracy down to the hour. Now even our alarm clocks can be synchro-

nized with a radio-broadcast time signal, making sure we will not be woken up a second later than desired. If timekeeping technology is progressing, it should go in two directions. Firstly, clocks need to grow increasingly smaller, cheaper, and simpler to operate, so as to better serve the needs of consumer electronics (the best example now being GPS devices). Secondly, they have to offer constantly improving accuracy to satisfy the increasing demands of diverse scientific fields – from precise spectroscopy of individual atoms all the way up to radio astronomy.

So great is the progress in this field that currently we can measure time better than any other physical quantity, with a relative measurement error of only 10^{-15} , corresponding to one second every 30 million years. Similarly, we can measure the frequency of a periodic signal (such as

The heart of a femtosecond laser: a sapphire crystal between concave mirrors



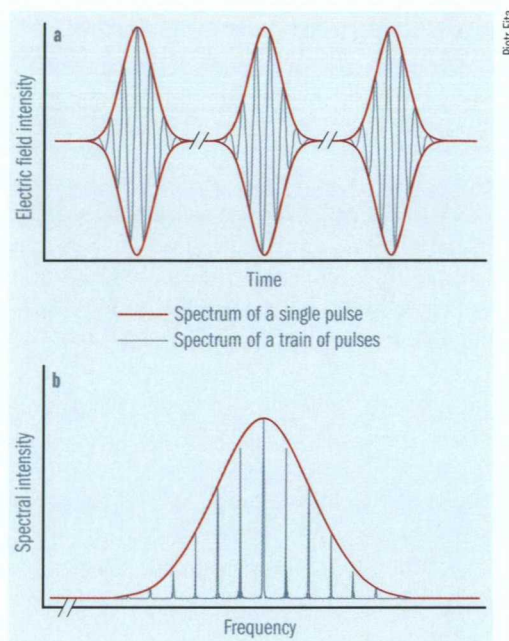
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electromagnetic waves) just as accurately since measurements of time and frequency employ the very same standard: a source of a signal oscillating in time with a well-defined period of oscillation. To measure a specific time-span one has to count the required number of cycles, and to measure a signal with a given frequency one has to count how many of its periods “fit inside” one second. The ability to gauge frequency is therefore the key to time measurements, but the existing electronics technology imposes serious limitations – the highest frequencies that can be directly measured using electronic devices do not exceed 10^{11} Hz, i.e. 100 GHz. Neither now nor in the near future should we expect electronic devices to become fast enough to count the cycles of electromagnetic waves in the visible spectral range, whose frequencies are of the order of hundreds of terahertz (1 THz = 1000 GHz). This makes the most precise frequency standards now known (optical transitions between various energy levels in atoms) extremely complex and difficult to operate. Although transitions between energy states in atoms are used in current atomic clocks their frequencies are in the microwave range (a few GHz) because they need to interface with electronics. Therefore they do not equal the potential capabilities of optical standards – but the latter have, until recently, required exceptionally complex frequency transformation systems which only the world’s largest metrological laboratories could afford.

Optical comb

A breakthrough came with the construction of the so-called optical frequency comb. It made its triumphant entry into the time-keeping world as a “clock-work” mechanism – it can relatively simply transfer signals between optical and radio frequencies (hundreds of megahertz) while retaining the accuracy of the optical frequency standard. This achievement earned John Hall and Theodore Hänsch a Nobel Prize in 2005.

The optical frequency comb emerged as a product of two previously independent fields of research in laser technology. Research into continuous wave lasers focused on achieving their greatest operational stability – that is, time-invariability of the light’s intensity and spectrum. At the



a: The oscillating electric field in ultrashort laser pulses (green curve) is enveloped by the red curve
b: The spectrum of a single pulse is shaped like the envelope of the spectrum of a stable train of pulses

same time, efforts were made to obtain the narrowest possible spectrum – in state-of-the-art systems its width is below 1 Hz, that is 15 orders of magnitude smaller than the frequency of the emitted waves. Physicists and engineers working with lasers producing ultrashort light pulses were moving in exactly the opposite direction. Pulses as short as a few femtoseconds ($1 \text{ fs} = 10^{-15} \text{ s}$), useful for studying the dynamics of very rapid processes, have a spectrum broader than 100 THz, against a frequency of approximately 400 THz (a wavelength of around 800 nm, on the border of the visible light and the near infrared). Such a broad spectrum is a direct consequence of the Fourier transform which imposes a lower bound on the product of the duration of a pulse and the width of its spectrum. In applications such as recording the dynamics of ultrafast processes one can treat each pulse separately, and therefore does not expect lasers to be exceptionally stable. Yet when used as a “clock-work” mechanism, a train of pulses generated by a femtosecond laser has to be considered as a single electric field waveform.

Let us compare the two approaches. The electric field waveform in a pulse (the green curve in figure a, p. 17) can be described as an oscillating function (cosine) multiplied by the envelope (for example, a Gaussian function – the red curve). The spectrum of

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such a pulse is also a Gaussian function (the red curve in figure b). However, for an appropriately long and regular train of pulses the spectrum is not smooth, but consists of a series of equidistant, very narrow lines (the green curve in figure b), whose envelope takes the shape of the spectrum of a single pulse. This is the optical frequency comb (in reality both the teeth and the spaces between them are very narrow; for a typical femtosecond oscillator several hundred thousand of them fit under the envelope, rather than several as shown in the figure). The distance between the teeth of the comb (f_r) is the pulse repetition frequency - the inverse of the time between two successive laser pulses. To observe an optical comb experimentally, a femtosecond laser has to meet very high requirements in terms of its stability since fluctuations in pulse-to-pulse intervals or the electric field phase result in chaotic movements of the individual teeth and blur the entire structure. However, in exchange for the effort put into stabilizing the laser, we obtain a spectrum that consists of a series of lines that correspond to optical frequencies, whose distance can range, depending on the construction of the laser, from several tens of megahertz up to several gigahertz. This makes it possible to lock oscillation in the radio or microwave range to optical oscillations.

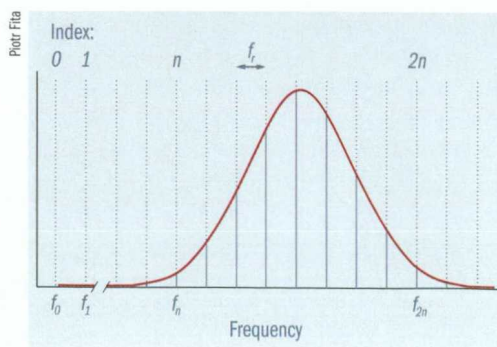
Where's the comb?

One more problem remains to be solved. If we extend the comb towards very low frequencies, it turns out that none of its teeth will precisely "hit" zero - the entire structure is shifted with respect to zero by a certain frequency that is unknown in advance, f_0 (figure on p. 18). As a result, the frequency of the tooth with a subscript n is given by $f_n = f_0 + n \times f_r$. The physical cause of this shift is the difference between the velocities of the maxima of the oscillating light wave and the velocity of the envelope. This can be observed if one carefully inspects Figure A on page 17 - in each pulse the oscillations are shifted differently with respect to the envelope. Applying a comb in metrology requires a precise identification of both characteristic frequencies f_r and f_0 , as otherwise the frequencies of the individual teeth in the comb cannot be identified. To measure f_r a

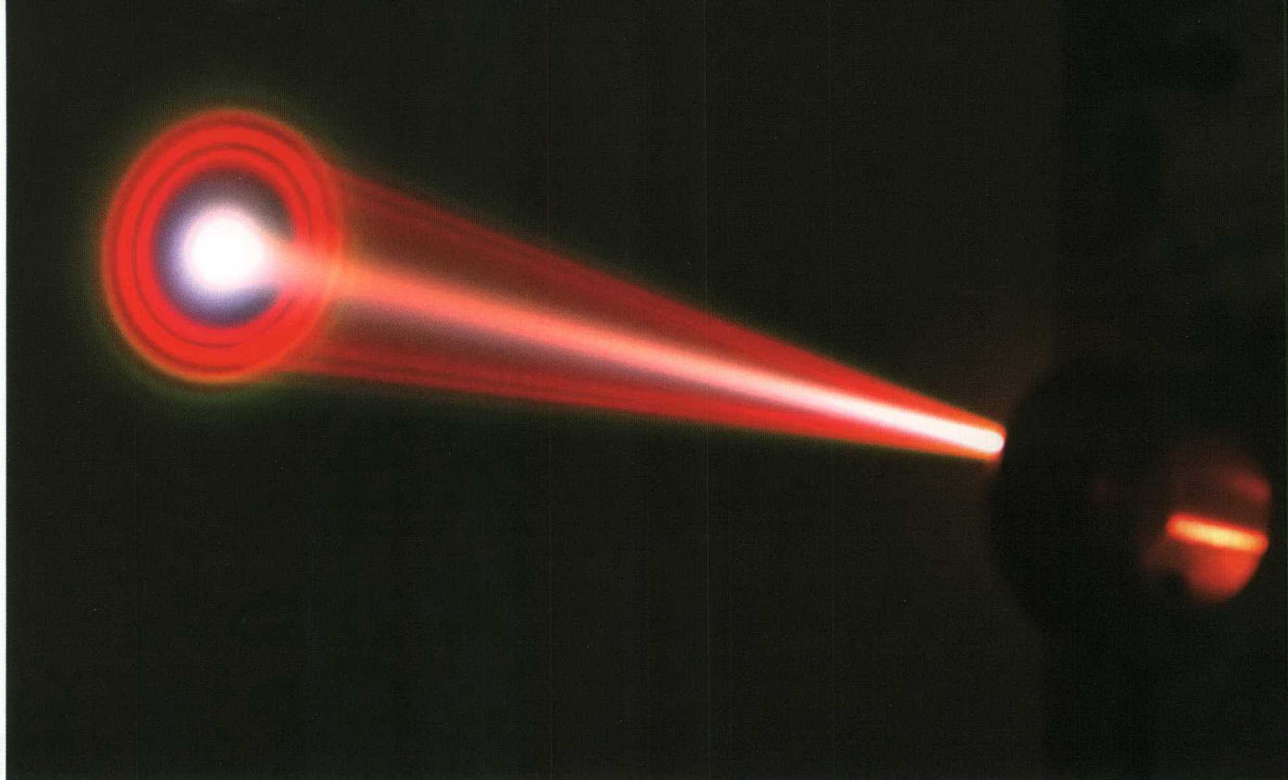
fast photodiode and an electronic frequency meter are sufficient. Measuring f_0 , on the other hand, is not an easy task, and relies on the use of extraordinary phenomena that occur when ultrashort laser pulses interact with matter.

To understand the genesis of such phenomena, we have to realize how short these ultrashort pulses are: if a second were the age of the universe, the duration of the pulses in the comb would be in the tens of minutes, while the separation between them would be longer than 100 years. This means that a relatively low power of the laser beam is in fact concentrated in portions of energy which are very short and separate from each other. Consequently, in femtosecond pulses the momentary light intensity and thus the electric field become high enough to force electrons in the medium, through which the pulse travels, to perform motion more complex than the one they perform when subjected to "ordinary" (low intensity) light. The motion of electrons now includes oscillations with frequencies that are multiples as well as sums and differences of the frequencies of the oscillations in the light wave. Vibrating electrons, in turn, emit electromagnetic waves, and as a result, after passing through a transparent medium new frequencies appear in the light. This could lead to the broadening of the spectrum or to the appearance of waves of double frequency (such as blue light, if the incoming wave is of red hue), i.e. the second harmonic.

These two phenomena make it possible to measure the value of f_0 as follows: first the spectrum of the comb is broadened so that it encompasses more than an octave, which means that the highest frequencies are at least twice as high as the lowest ones. To this aim, the train of pulses is



The optical comb with teeth separation f_r is shifted with respect to zero by an unknown frequency f_0 . Determination of f_0 is possible if both teeth of subscripts n and $2n$ are present



passed through a photonic fiber – yet another great discovery of optics in recent years – a very thin (1–2 μm) quartz fiber surrounded by a network of thin-walled quartz tubes. Unlike standard optical fibers, in which the core is surrounded by a uniform cladding of a slightly different material, photonic fibers do not lengthen the transmitted pulses, which would cause a drop in the light intensity. This means that a pulse can retain its very high intensity in a long stretch of the fiber, while at the same time broadening its spectrum. In the case of a comb, this leads to the appearance of additional teeth on both sides. By choosing the right length of the fiber and the intensity of the incoming light, one can achieve an output spectrum wider than an octave. Next, using a filter, two teeth are selected from the comb, one with the subscript n , the other $2n$. However, their frequencies do not differ by a factor of 2, due to the presence of the shift f_0 . This enables one to determine f_0 by doubling the frequency of the tooth with subscript n (in the process of generating the second harmonic) and mixing it with the tooth bearing subscript $2n$. The mixing of two frequencies produces, among other components, one with a frequency that equals the difference between the two input frequencies, which in our case equals f_0 . For technical reasons it may be easier to choose groups of teeth rather than individual ones, which does not change the ultimate outcome.

The frequency f_0 can be at most equal to f_n , in other words it also lies in the radio or microwave range. Thus, we can easily com-

pare it with the frequency of the microwave atomic frequency standard and stabilize the laser so as to transfer the stability of the standard into the stability of the comb teeth frequencies. This method enables one to measure optical frequencies by relating them to existing frequency standards, as a result of which a very high precision of spectroscopic measurements can be achieved. Even more promising is the same approach but “from the opposite side”: a chosen comb tooth can be stabilized against the frequency of an atomic standard operating in the optical range (therefore more stable than a microwave one), transferring its stability to a low-frequency range. The periods of the latter can be readily counted using electronic devices. As a result, a clock whose potential precision can be greater than 10^{-18} and whose error is equivalent to several seconds within the age of the universe can be constructed!

The optical comb has made it possible to combine techniques of cooling and trapping ions and atoms, ultraprecise spectroscopy, nonlinear optics and laser stabilization, which will lead to the construction of a simple, easy-to-operate atomic clock with an optical frequency standard. This will be a joint success of the previously independent fields of optics, spectroscopy and atomic physics. ■

Further reading:

- Udem Th., Holzwarth R., Hänsch T.W. (2002). Optical frequency metrology, *Nature*, 416, 233–237.
- Takamoto M., Hong F.-L., Higashi R., Katori H. (2005). An optical lattice clock, *Nature*, 435, 321–324.

The interaction of ultrashort laser pulses with matter leads to new frequencies