Laser Frequency and Time

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The following two contributions in this volume are highlighting some remarkable recent developments at the interface between precision laser spectroscopy and ultrafast laser physics. After decades of struggle, we have finally found a practical method for measuring the frequency of light with extreme precision [1]. Femtosecond laser optical frequency comb synthesizers are opening exciting new perspectives for atomic spectroscopy and they can provide the clockwork for optical atomic clocks that will eventually far surpass the accuracy of the best microwave cesium clocks. J. C. Bergquist et al. [2] are reporting on a first optical atomic clock at NIST based on a single trapped Hg^+ ion. The contribution by Jun Ye et al. [3] is illustrating the wealth of new opportunities for femtosecond laser frequency combs in the frequency and time domain.

1 Introduction

Counting the cycles per second of a laser beam has finally become a relatively simple task. The essential steps leading to this breakthrough have been completed at the Max-Planck-Institute for Quantum Optics in Garching in 1998 [4]. Many journals and newspapers have reported on these developments since the summer of 1999. Today, a universal optical frequency comb synthesizer can be realized with just one single mode-locked femtosecond laser such as a Kerr-lens moded-locked Ti:sapphire laser. The near infrared pulse train from such a laser oscillator is coupled into a special micro-structured quartz fiber which broadens the spectrum to span a spectacular rainbow of visible colors. The light emerging from the fiber is not ordinary white light; it consists of several hundred thousand sharp spectral lines, forming an evenly spaced optical frequency comb. The spacing between two neighboring comb lines is precisely equal to the pulse repetition frequency of the laser, typically in the range between 50 MHz to 1 GHz. For a freely running laser, the comb lines are shifted relative to the integer harmonics of the repetition frequency by some unknown offset frequency that is caused by the slippage of the carrier wave relative to the envelope from pulse to pulse. With a comb spanning more than an optical octave, it has become straightforward to measure or control this offset frequency. If the offset frequency is set to zero, all the comb lines oscillate at precise integer harmonics of the laser repetition frequency.

Such a femtosecond laser frequency comb provides a direct link between the optical and the microwave region. This link can be used in either direction. If the laser repetition frequency is controlled with a cesium atomic clock, the comb

generator synthesizes a vast grid of optical reference frequencies that can be traced directly to the primary microwave cesium frequency standard on which the definition of the second has been based since 1967. Most experts had serious doubts that such a feat would ever be possible because even the best radiofrequency oscillators have so much phase noise that the comb structure should be completely washed out at optical harmonic frequencies. Fortunately, the cavity of the femtosecond laser provides the necessary stabilizing flywheel action, so that exotic solutions such as cryogenic sapphire microwave resonators are not needed.

To use the comb synthesizers in the opposite direction, one of the comb lines may be locked to a sharp optical resonance of a cold trapped ion, of cold atoms, or suitable molecules. All the other comb frequencies and also the pulse repetition frequency become then known rational multiples of this optical reference frequency. Such a device can act as a clockwork that divides the high frequency of a „pendulum“ oscillating with the frequency of light into the microwave region, where it can be processed and counted with existing electronic circuitry.

By slicing time into smaller intervals, optical atomic clocks may eventually provide a 1000-fold improvement in accuracy over today's best cesium clocks. One can think of numerous applications for better clocks. They can play an important role in time and frequency metrology. By improving the synchronization of clocks over large distances, they will have an impact on very long base line interferometry in radio astronomy or even optical astronomy. Other applications include satellite navigation, precise tracking of remote space probes, telecommunications, observations of the variability of earth's rotation, geodesy with millimeter precision or studies of variations in pulsar periods. For the participants of ICAP, the most intriguing prospects are perhaps the new horizons in precision atomic spectroscopy, new tests of special and general relativity, or laboratory searches for conceivable slow variations of fundamental constants. Our own research in this field has long been motivated by precision spectroscopy of the simple hydrogen atom which permits unique confrontations between spectroscopic experiment and quantum electrodynamic theory [5].

2 Towards an Optical Frequency Counter

The quest for an optical frequency counter goes back to the early days of the laser. As the session chairman who has been involved in these developments for decades and has had some hand in the recent advances, I should perhaps provide a personal perspective of this long adventure.

In the early 1960s, A. Javan, the inventor of the He-Ne-laser, was probably the first to superimpose the light of two cw-lasers with a beam splitter and to observe a beat signal with a photodetector. This was a remarkable experiment. It showed that lasers can behave like radio-frequency oscillators. They can produce a classical light

wave of well defined amplitude and phase. It also gave evidence that laser light can be of exceptional spectral purity. The discovery of spectral hole burning [6] and the "Lamb dip" [7] led to novel nonlinear methods of laser spectroscopy which could eliminate the Doppler-broadening of spectral lines, and soon Doppler-free resonance lines became so narrow that no classical spectroscopic technique could do them justice.

In the mid 1960s, A. Javan initiated a research program at MIT aimed at extending microwave methods of frequency measurement into the optical region. He experimented with delicate nano-scale metal whiskers to form metal insulator metal point contact diodes which could serve as antennas, harmonic generators, frequency mixers, and detectors up to very high frequencies. In 1967, he succeeded in measuring the frequency of a far-infrared HCN laser at 871 GHz in this way [8]. Building on this work, the laboratory of Ken Evenson at NBS in Boulder, Colorado, developed the first harmonic laser frequency chain [9]. After measuring a frequency of 88 THz in 1973, they went on to push such frequency measurements into the visible regime.

As recently as 1992, the Guiness Book of Records had an entry describing the highest measured frequency. The record refers to a yellow-green line of molecular iodine measured in 1979 by the Boulder team[10]. The accuracy of one part in 10^9 was not particularly impressive by today's standards. But, for the first time, the frequency of this line had been measured rather than the wavelength. In 1983, the same NBS laboratory succeeded in measuring the frequency of a 474 THz He-Ne-laser, stabilized to I_2, to some parts in 10^{11} [11]. The realization that it was now possible to measure the frequency of laser light led the International Committee of Weights and Measures (CGPM) in 1983 to do away with a standard for the meter. Instead, the velocity of light in vacuum, c, has been defined as 299 792 458 m/s. From then on, in order to find the wavelength λ of a laser in meter, one would simply measure its frequency f and use the relationship $\lambda f = c$.

Soon after this rule was in the books, the NBS frequency chain was abandoned, because it proved too complex, unreliable and expensive to maintain. For the next fifteen years there was not a single laboratory in the U.S. that would have been able to follow the new prescription. Things looked a bit brighter in Europe, notably in France and in Germany. In 1996, researchers from the PTB in Braunschweig laid claim to the first phase-coherent frequency measurement of visible radiation [12]. Like the earlier harmonic frequency chains, their new chain was designed to measure just one particular optical frequency, in this case the red intercombination line of atomic calcium, to serve as a visible frequency standard. The PTB chain starts with a 9 GHz cesium oscillator and first goes down in frequency so that a hydrogen maser and a quartz-stabilized rf oscillator can provide the necessary short-term phase stability. To move from here up to visible frequencies one has to traverse a vast region of the electromagnetic spectrum. Nonlinear devices are used in successive steps to generate higher harmonic frequencies. After each such step, the the feeble harmonic power has to be boosted with a phase-locked transfer

oscillator that also acts as a flywheel. More complex strategies are required to reach the desired final frequency. Thus, the entire chain was spread out over three laboratories in two different buildings.

For obvious reasons, such a large harmonic frequency chain would have been ill-suited for our precision spectroscopy of atomic hydrogen in a university-like setting. Searching for conceivable alternatives, I came up with the idea of a frequency interval divider chain at the time of the 1988 ICAP conference [4]. The plan was to measure the interval between a laser frequency and the second harmonic, which is equal to the laser frequency itself. If this interval is repeatedly cut precisely in half, one will eventually reach a sufficiently small gap that can be measured as a beat signal. Each divider stage contains a laser whose second harmonic frequency is phase-locked to the sum frequency of its two inputs. All these stages can operate in a limited near-visible region of the electromagnetic spectrum so that they can be implemented with small diode lasers, using nearly identical technology.

The first such divider chain had only 4 - 5 stages and was not yet sufficient to realize an optical frequency counter, but it served to measure a 1 THz frequency gap that arose in a measurement of the hydrogen 1S-2S two-photon resonance frequency with a traditional harmonic frequency chain [13]. A CH_4-stabilized He–Ne-laser served as a transportable intermediate reference standard in that experiment. This laser, built in the laboratory of S. Bagaev in Novosibirsk, has been transported back and forth between Garching and Braunschweig perhaps a dozen times, in order to calibrate it against a cesium clock with the help of a PTB harmonic frequency chain. In the end, we could measure the 1S-2S frequency to within 3.4 parts in 10^{13}, a record at that time. Together with the frequencies of other resonances, measured by F. Biraben and coworkers in Paris, we were able to give a much improved value for the Rydberg constant, as well as for the 1S ground state Lamb shift. If one believes in QED, this measurement also provides an accurate value for the rms charge radius of the proton, far surpassing the accuracy of electron scattering experiments with large accelerators.

3 Femtosecond Laser Frequency Combs

After completion of these experiments, in March of 1997, I happened to be at the European Laboratory for Nonlinear Spectroscopy (LENS) in Florence, Italy, where M. Bellini was working with an amplified femtosecond laser system. By focusing the pulses into a crystal of CaF_2, he produced a white light continuum, as is common in many ultrafast laboratories. Out of serendipity I asked what would happen if one split the original laser beam in two and focused this light in two different spots. Would the two white light continua interfere? Of course, this would not be expected if the white light sources acted like sparks emitting incoherent light. However, when we tried the experiment, paying attention to proper timing, we

observed stable interference fringes of high contrast over the entire visible spectrum [14]. This implies that the two white light continua are phase-locked to each other and to the driving laser field.

I felt electrified by this insight, because it meant that a periodic train of such white light pulses could serve as a universal frequency comb synthesizer. Soon I had written down a 7-page detailed proposal, starting with the following abstract:

"CONFIDENTIAL - Proposal for a universal optical frequency comb synthesizer – T.W. Hänsch, Max-Planck-Institut für Quantenoptik - (March 30, 1997).

An optical frequency synthesizer is proposed which produces a wide comb of absolutely known reference frequencies throughout the infrared, visible, and ultraviolet range. To this end, a white light continuum with pulse repetition rate f_r is produced by focusing the output of a mode-locked femtosecond laser into an optical fiber or bulk medium with a third order nonlinear susceptibility. The rate of phase slippage of the laser carrier relative to the pulse envelope, f_0, is monitored by observing a beat signal between the wight light continuum and the second harmonic of the laser…."

Although we did not know yet how to produce phase-coherent white light pulses with a sufficiently high repetition rate, I considered this proposal compelling enough to ask two of my coworkers, Dr. Th. Udem and Dr. M. Weitz, to witness it with their signatures, and I discussed it confidentially with a number of good friends, including J.L. Hall from JILA who remained skeptical at that time, but later became a main driving force for a series of beautiful experiments in Boulder.

My confidence was based on more than just the Florence experiment. At Stanford University in the late 1970s, we had shown that a mode-locked picosecond dye laser emits a regular frequency comb that can be used as a ruler in frequency space to measure large line splittings [15]. Some people were surprised that you could do precise spectroscopy with short pulses, since such pulses have inevitably a broad spectrum. However, we are not working with an individual pulse but with a periodic pulse train emitted by a mode-locked laser. In the frequency domain, such a pulse train can be described as a coherent superposition of discrete modes. It is instructive to consider an arbitrarily shaped pulse circulating inside a laser cavity [16]. After each roundtrip, a weak copy of this pulse is escaping through a semitransparent mirror. A single pulse will have a broad and more or less complicated spectrum. Two identical copies separated by the roundtrip time T, will give rise to interference fringes in the spectrum, spaced by 1/T, similar to the spatial interference pattern in the famous Young's double-slit experiment. With a large number of pulses, the spectrum will resemble the diffraction pattern of a grating, with a regular comb of sharp lines which can be identified with the modes of the resonator. In reality, successive copies will not be identical but the phase of the carrier wave will change relative to the pulse envelope because of dispersion inside the cavity. Already in the 1970s, we realized that the comb lines of a laser are displaced from the precise integer harmonics of the repetition rate by an unknown offset frequency, which is equal to the rate at which the carrier phase changes from

pulse to pulse [17]. However, the laser spectrum was much too narrow to gain experimental access to this offset frequency.

In the 1990s, the technology of ultrafast lasers has advanced dramatically, in particular with the advent of Kerr-lens mode-locked Ti:sapphire lasers. In the simplest case, such a laser consists just of the laser crystal and two mirrors forming a cavity. The crystal acts as an optical Kerr medium with an intensity-dependent refractive index. A laser pulse with a Gaussian beam profile will thus create a lens that focuses the light at some location in the resonator. If an aperture is placed at this strategic location, the focused light can pass without losses. However, any mode that refuses to oscillate in lockstep cannot take advantage of the transient Kerr lens and will suffer high losses. Although such femtosecond lasers have been widely available, they have been used almost exclusively for studies of ultrafast phenomena. And until 1997, nobody had looked for any comb lines as far as we could tell.

Th. Udem, R. Holzwarth, and J. Reichert, experimenting in our laboratory at Garching with a commercial femtosecond laser (Coherent Mira), soon detected comb lines by superimposing the pulse train and the beam from a cw diode laser and observing a radio-frequency beat signal with a photodetector. We could then investigate how regular the spacing of the comb lines really is. As experimentalists, we were most worried about the lines far out in the wings of the spectrum, where the dispersion of the laser cavity is not well compensated. We would phase-lock two diode lasers to two arbitrarily chosen comb lines and use a frequency interval divider stage to lock another diode laser to the precise midpoint. The beat note with the nearby comb lines confirmed that the mode spacing is uniform across the comb within 3 parts in 10^{17} [18]. This uniformity is preserved if the laser spectrum is further broadened by self-phase-modulation in some length of standard optical communications fiber. We also verified that the mode spacing equals the repetition rate within 6 parts in 10^{16}.

In the very first measurent of an optical frequency with a femtosecond laser, we have determined the frequency of the cesium D_1 resonance line by bridging the gap to the fourth harmonic of a CH_4-stabilized 3.39 μm He-Ne laser with the comb spectrum serving as a ruler in frequency space [19]. The Cs line is needed for a new determination of the fine structure constant α by photon recoil experiments in the laboratory of S. Chu at Stanford.

In early 1999, we proceeded to measure the absolute frequency of the ultraviolet hydrogen 1S-2S two-photon resonance in a direct comparison with a microwave cesium clock in our own laboratory [20, 21]. Since we did not yet have an octave-spanning frequency comb, we employed some interval divider stages to produce two different rational multiples of the frequency f of a 486 nm dye laser in the hydrogen spectrometer. The interval between 4/7 f and 1/2 f could then be bridged with a fiber-broadened frequency comb. In the summer of 1999, a transportable cesium fountain clock, built by the team of A. Clairon and C. Salomon at LPTF in Paris, was brought to Garching to serve as a reference standard. In this way we have measured the hydrogen transition frequency within 1.8 parts in 10^{14},

surpassing the accuracy of any previous measurement of an optical frequency by more than an order of magnitude [21].

The realization of the originally envisioned octave-spanning frequency comb appeared within close reach in the spring of 1999, when researchers at Bell Labs reported on a holey "rainbow fiber". J. K. Ranka had focussed the output from a mode-locked femtosecond laser oscillator into a microstructured silica fiber with a small solid fiber core surrounded by air-filled canules, and observed dramatic spectral broadening to a rainbow of colors [22]. After the Florence white light experiments I felt certain that it must be possible to obtain sharp comb lines in such a train of white light pulses. Naturally, we tried immediately to obtain a sample of this magic fiber. J.L. Hall was at Munich at the time, and together we called many old friends at Bell Labs. However, the scientists had to capitulate to the company lawyers, and we never received the promised fiber. Fortunately we learned that the group of P.St.J. Russel at the University of Bath in the U.K. had long been experimenting with similar photonic crystal fibers [23], and they agreed to a scientific collaboration.

The air-holes surrounding the fiber core cause a large step in the effective refractive index, so that the critical angle of total reflection permits tight focussing and high intensities in the core. Since some of the light travels as an evanescent wave in air, the region of zero group velocity dispersion can be shifted from a wavelength of 1.3 μm in bulk silica to below 800 nm, so that an injected laser pulse is not quickly spreading due to dispersion. In an elementary model, the phenomenon of white light continuum generation in such a fiber can be understood in terms of four-wave-mixing due to a third order nonlinear susceptibility. Alternatively, in the time-domain, we expect a self phase modulation due to the intensity-dependent refractive index. In the absence of dispersion, the intense central part of a pulse travels with a relatively slower velocity so that the pulse becomes distorted, and we expect a red-shift at the beginning of the pulse and blue-shift at the end. In reality, the process of white light generation in photonic crystal fibers is considerably more complex, with phenomena such as phase-matching, soliton splitting, shock-wave formation and stimulated Raman- and Brioullin scattering leading to a complicated spectral and temporal structure. However, for the purpose of frequency comb generation, it is only important that the process is sufficiently reproducible to maintain the phase coherence between successive pulses. We have since learned that suitable white light continua can also be generated with a tapered ordinary fiber or even directly with a femtosecond laser oscillator.

Once the comb is spanning more than an octave, it is very easy to determine the offset frequency f_0. For instance, one may take one of the comb lines near the red end of the spectrum, with a frequency $f_n = n f_r + f_0$, displaced by an offset f_0 from a precise integer multiple of the repetition frequency f_r, amplify it in some laser, such as a small diode-pumped Nd.YAG laser, and then double its frequency to $f_{2n} = 2n f_r + 2f_0$., which is now displaced by twice the offset frequency. A beat note with a nearby comb line $f_m = m f_r + f_0$ reveals directly the offset frequency f_0. The

absolute frequency of each comb line is then determined by the two radio-frequencies f_r and f_0 and an integer mode number. For a sufficiently large line spacing, as e.g. provided by a small Ti:sapphire ring laser [24] (GigaOptics GmbH, GigaJet), this mode number can be uniquely identified with a common wavemeter. As already envisioned in the original proposal, the auxiliary Nd:YAG laser is not necessary, if one works with several thousand comb lines at once and thus uses pulses of sufficient intensity to allow efficient frequency doubling in a nonlinear crystal. By superimposing the doubled and the original comb pulses on a photodetector, a collective beat note is observed which readily gives the offset frequency f_0. This latter scheme has first been experimentally realized by D. J. Jones et al. at JILA [25], who obtained a sample of the Bell Labs fiber a few weeks before our laboratory received a photonic crystal fiber from the University of Bath.

In a first stringent test, R. Holzwarth has compared a single laser optical frequency comb synthesizer with the more complex frequency chain used in the hydrogen 1S-2S measurements [26]. Starting with a common 10 MHz rf oscillator and comparing comb lines near 350 THz, he could dcemonstrate agreement to within 6 parts in 10^{16}. The performance has likely been limited by Doppler-shifts due to air pressure changes or thermal expansion of the optical tables. Very recently, J. Stenger et al. at the PTB [27] used a laser frequency comb to measure optical frequency ratios. As a test case, they measured the ratio between the second harmonic and the fundamental frequency of a Nd:YAG laser. They confirmed the expected result of 2 to within 7 parts in 10^{19}. Since then, M. Zimmermann, a graduate student at Garching, has pushed a similar measurement to 4 parts in 10^{21}. Therefore, we can be optimistic that the femtosecond laser clockwork will not limit the envisioned performance of future atomic clocks.

With ultraprecise femtosecond laser frequency comb synthesizers becoming available commercially [28] we can now think about new precision experiments to test fundamental physics laws in new and unexplored territory. If history is any guide, the biggest surprise would be if we found no surprise.

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