output, the physical origins of the electric field dynamics, and an overview of applications of femtosecond comb technology. Chapter 2 by Ippen, Kärtner and Cundiff discusses the development of ultrashort lasers, particularly focusing on how to achieve an octave-spanning spectrum and pulse dynamics that are relevant to the stability and control of the comb. Chapter 3 by Bartels describes in detail high-repetition-rate ring oscillators for precision frequency metrology. Chapter 4 by Gaeta and Windeler provides in-depth discussions relating to the physics of bandwidth generation and the underlying noise process during pulse propagation through microstructure fibers. Certain aspects of comb dynamics and stability are presented in Chapters 5 by Steinmeyer and Keller. An attractive approach presented in Chapter 6 by Kobayashi makes use of optical parametric generation to produce high-peak-power, femtosecond pulses in the IR spectral domain. A detailed review of the traditional harmonic-based frequency chain is provided in Chapter 8 by Schnatz, Stenger, Lipphardt, Haverkamp, and Weiss, while the new epoch of absolute optical frequency measurement using femtosecond comb technology is reviewed in Chapter 7 by Udem, Zimmermann, Holzwarth, Fischer, Kolachevsky, and Hänsch. Chapter 9 by Diddams, Ye, and Hollberg provides an account of the current state-of-the-art performance and characterization of femtosecond comb systems used for optical frequency measurement, synthesis, and optical atomic clocks. Chapter 10 by Baltuška, Paulus, Lindner, Kienberger, and Krausz provides a thorough discussion of the generation of the high-intensity pulses needed to access the regime of extreme nonlinear optics and a review of the results obtained for above-threshold ionization. Control of high-harmonic generation is addressed in Chapter 11 by Gibson, Christov, Murnane and Kapteyn. Stabilization of mode-locked lasers and their applications to ultrasensitive sensors are discussed in Chapter 12 by Diels, Jones, and Arissian.

The rapid progress during the last 5–6 years has been breathtaking and has made a tremendous impact on both science and technology. We foresee an undiminished potential for similar advances in the near future. We hope that the readers of this book will share our enthusiasm and benefit from the material presented in this book.

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Foreword

HISTORY OF OPTICAL COMB DEVELOPMENT

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In the past five years, progress in laser stabilization, optical frequency measurement, femtosecond laser development and stabilization, nonlinear optics, and related topics has been stunning and unexpected. The excitement surrounding the rapid evolution in these fields since 1999 gives us a hint of what it must have been like after 1927 when the first ideas of quantum mechanics were being introduced. With laser optics, however, the explosion of knowledge is based upon years of detailed, painstaking research in the independent fields of laser stabilization, ultrafast laser development, and highly nonlinear optics [1]. The coalescence of these fields has provided five years of almost unprecedented discovery while, at the same time, a new millennium in metrology has generated advances of fundamental value in the contributing fields and in their spin-offs. To give just two examples: (1) the precise synchronization of picosecond and femtosecond lasers [2] allows nonlinear surface probing at the single-molecule level using coherent anti-Stokes Raman microscopy and spectroscopy [3] and (2) the use of stabilized comb pulses allows time and frequency dissemination [4] over extended distances via fiber optics. The latter offers a new capability to metrologists and researchers interested in developing new accelerators and large-array radio telescopes.

Here we offer our perspective on how we got started on this incredible journey of discovery and why it is occurring at this time. The rest of this timely book highlights key technical advances from the perspective of the researchers who made them. Consequently, this book should be of interest to students, practitioners in this rapidly evolving field, and physicists, chemists, and biologists whose research will be enhanced by the insights and discoveries recounted here.

The historical development of pulsed and continuous wave (cw) lasers diverged right from the beginning. The solid-state ruby laser of Ted Maiman involved kilojoule discharges into flash lamps and repetition rates from zero up to once per minute. (The laser's high ion density implied that a serious power level would be involved.) In contrast, the gas laser of Ali Javan had much lower gain margins and required temporal stability to allow its incremental approach to the laser threshold. From the early 1960s until about 1990, the pulse and cw laser communities continued to diverge. The cw laser folks liked to enhance the stability of their lasers, because stability was the main good feature they had. Certainly it wasn't the ability to burn holes through Gillette razor blades, a popular specification for pulsed lasers of the day. Over the years, the cw-laser teams learned to frequency stabilize their milliwatt-scale lasers, eventually reaching into the subhertz domain. Some members of the pulse-laser community escalated to kilojoule pulse energies delivered in nanoseconds for fusion target compression and even larger discharges to be delivered to potentially hostile moving targets.

On the other hand, the university research community wanted pulse lasers with high repetition rates that would enhance nonlinear responses, enable signal averaging over many pulses, and reduce the destructive impact of the probing radiation on the probed system. Thus began the search for laser media that were quiet and calm enough to lead to repeatable pulses, even at high repetition rate. First came the mode-locked He-Ne laser followed by actively mode-locked Argon lasers, which were developed commercially to give nanosecond pulses at 100 MHz rates. By the time synchronously pumped mode-locked picosecond dye lasers were introduced in the mid-1970s, the comb approach to frequency measurement was probably already inevitable, albeit more than 20 years in the future.

A driving force in the development of laser (and optical comb) technologies was the desire to learn more about the physical world. For instance, Professor Hänsch's group at Stanford (and later in Garching) focused on learning about the hydrogen spectrum, comparing hydrogen with deuterium to see the isotope shifts, accurately determining energies associated with various principal quantum numbers, and so forth. From the beginning, they realized that certain physical parameters, such as the Lamb shift or proton-size changes, could be isolated by using their quantum-number-scaling dependence and thus probed in the optical regime.

At Stanford, Hänsch's team demonstrated one of the first mode-locked "femtosecond" dye lasers (with a pulse length of less than one picosecond) in 1977 [5]. Their progress in laser research was closely connected to the invention of spectroscopic techniques in the sub-Doppler regime as part of their work on precision spectroscopy of the simplest atomic system, hydrogen. The group's new tunable dye-laser pulses were soon used in a landmark experiment to demonstrate Fourier's reality to us: stable pulse trains — evenly-spaced in time — represented as stable combs of frequency components. We note that in the late 1970s, Veniamin Chebotayev in Novosibirsk had also begun thinking about stable, repetitive laser pulse trains. Even though the early experiments could not yet verify the precisely equal spacing of the comb frequencies, Eckstein, Ferguson, and Hänsch [6] used a comb structure to measure some fine and hyperfine frequency intervals in atomic sodium in 1978. This classic paper would help define the future impact of ultrafast lasers on precision measurement. Then, as the work at Garching progressed over the years, one dream became clear and insistent: if *only* we could measure optical frequencies directly and accurately!

This vision led the Garching team to flirt with extended chains of synchronized frequency sources. In the late 1980s, their new idea [7] was to deal with a series of increasing frequency *differences* between tunable diode laser sources. In this way, they hoped to avoid one of the major headaches of traditional chains — the step-wise increase of *absolute* frequencies throughout the chain that obliges one to develop a different laser technology at many points to cover the 10^5 frequency ratio from the microwave regime to the visible. A number of laser-diode frequency-interval-divider stages were built. A four-stage system was used for one of the hydrogen frequency measurements.

The comb idea jumped ahead with the demonstration of intracavity modulator-based spectral comb generators by Kourogi et al. [8]. In these experiments, pulses were modulated onto stable cw laser beams. Researchers in both Garching [9] and Boulder [10] recognized the utility of these devices and launched frequency-measurement programs using their few terahertz width modulator-based optical combs to bridge the awkward frequency gaps.

In the meantime, the ultrafast laser community continued to work toward improved pulse train stability and shorter pulses. Researchers developed several solid-state lasers, such as the cw-pumped mode-locked Nd:YAG, that were attractive for their stability. However, pulse durations below 30 ps were difficult to obtain because of the gain-bandwidth limitation of the laser material itself. Eventually the "intelligent and beautiful princess" — the titanium-doped sapphire laser system — was introduced and developed, along with the important discovery of Kerr-lens mode locking by Wilson Sibbett in St. Andrews [11]. These inventions changed femtosecond lasers from delicate contraptions to simple and reliable devices. Soon afterwards, commercial Ti:sapphire femtosecond lasers became readily available and they offered sub-100 fs pulses by the early 1990s. By 1994, the Garching group had acquired a Coherent Mira* laser for frequency metrology experiments. This laser and other similar devices opened the door to solving an array of challenging problems.

The factors that limit the shortness of the generated laser pulses arise from two issues: (1) finite gain-bandwidth product (which is not a problem for Ti:sapphire) and (2) intracavity dispersion. A short pulse can be viewed as the superposition of many cw phase-locked modes, all of which oscillate at their own cavity-defined frequencies. For the pulse train to be stable in time, the modes must have a common frequency separation. Because of dispersion in the sapphire, intracavity air, and mirror coatings, these cavity frequencies are generally not exactly evenly spaced. This is particularly true as the spectral bandwidth dramatically increases. So even with the ~30% bandwidth of Ti:sapphire, further shortening of sub-100 fs pulses proved difficult until Asaki et al. [12] employed a sufficiently general analysis of the pulse laser cavity. This model included the index of refraction characteristics of the intracavity dispersion-compensating prisms, the resulting color influence on the refraction direction, associated cavity path lengths, and some modeling of the air and laser crystal dispersion. Space-time focusing of the light bullet in the laser crystal was another important consideration.

In the early 1990s, when most of researchers were working feverishly to shorten pulse widths, some dreamers began to think of pulses so short that their Fourier representation would span from radio frequencies up into the visible domain. This idea seemed like science fiction to one of the authors (JLH) and a likely possibility to the other (TWH). Of course, spectral self-broadening was well known. By focusing powerful amplified pulses into water or some solids, one could basically generate a white-light continuum. At elevated pulse energy levels, one expected serious disruption of the calmness of the intermolecular bonds; consequently, one would not expect to find a phase-stable repetition of the generated white light. Perhaps a glass sample *could* melt and recrystallize at a 100 MHz rate, emitting a similar thermal spectrum on every heating cycle. However, to form a coherent optical comb in the forward direction, the timing would need to be stable to ~1 radian — at the visible frequency! Few believed that this thermal process would be stable at the 0.3 fs level needed. Rather, it seemed clear that a more gentle process would be required, in which somewhat less-powerful laser pulses would strongly distort some atomic wave functions but not disrupt chemical bonds. Atomic frequencies are so high that when the pulse is gone, calmness can return; the next pulse would be able to generate just the same effect on the system. In this case, the phase-coherence of the source pulses could insure that the nonlinearly generated frequencies would be mutually coherent pulse-to-pulse.

Researchers in both Garching and Boulder set out to learn about this subject. In Europe, an amplified pulse was split into two parts and focused onto two separate spots in a CaF_2 crystal. The white light produced in each spot in the CaF_2 plate interfered with each other in a geometry that allowed the time delays to be equalized. Miraculously, stable white interference fringes could be seen by eye [13]. In this way, the Garching group realized that the phase of the nonlinearly generated light was stable enough to form an optical comb! This experiment led to Hänsch's detailed six-page proposal, dated March 30, 1997, for an octave-spanning self-referenced universal optical-frequency comb synthesizer. Following this proposal, developments in the art of femtosecond-laser frequency comb generation began to appear quickly.

The next year (1998) saw the first crucial test of a Kerr-lens mode-locked Ti:sapphire laser in Garching [14]. This experiment clearly proved the viability of femtosecond-laser frequency comb synthesizers. Before this experiment, some researchers had argued that the laser comb spectrum would be completely washed out by phase noise. Although some publications were held back due to the restrictions of German patent law, by 2000 the Garching team reported its first absolute frequency measurement with a comb (made in 1999) [15]. In a direct comparison with the transportable cesium fountain clock of the Bureau National de Métrologie – Systèmes de Référence Temps Espace, they measured the frequency of the hydrogen 1S–2S two-photon resonance with an uncertainty of 1.9×10^{-14}, which is more than an order of magnitude more accurate than any previous optical frequency measurement. This experiment firmly established the viability of optical frequency metrology with femtosecond laser frequency combs. It also electrified the frequency metrology community. The convenience and simplicity of octave-spanning frequency combs added to the attraction of this new approach.

In the meantime, researchers in Boulder were studying the spectral expansion associated with pulse propagation in an optical fiber as a possible replacement for their modulator-based comb generator. They showed that a cw test beam in the fiber developed a comb structure on it because of the co-propagating femtosecond pulses. However, the new comb only contained information about the repetition rate of the pulses, not about their optical frequency [16]. Of course, in retrospect, it is clear that this "cross-modulation" result (as opposed to "four-wave mixing") was preordained because of the huge difference in the phase velocities of the two colors. The interesting frequency-coupled wavelets never got a chance to coherently build up along the fiber's length. This experiment also showed that spectrally narrow features would be generated across a broad spectral range even when "pounding" on a fiber with powerful femtosecond pulses.

A classic 1999 paper [17] offered a complete description of issues and techniques for comb-based optical frequency measurement, including the famous carrier-envelope phase-slipping issue [18]. The Garching team made their first self-contained rf-to-optical frequency comparison [19] using a comb configuration that had a stabilized carrier-envelope-offset frequency. Since the comb spectrum did not yet span an octave, some interval-divider stages with auxiliary lasers had to be used for self-referencing.

Soon afterwards, the world was turned upside down by Jinendra Ranka, Robert Windeler, and Andrew Stentz. During the 1999 CLEO postdeadline session, they announced that they had demonstrated that *white light* could be produced in a revolutionary way by using an internally structured fiber incorporating a number of air holes [20]. They showed pictures in which the input dark red pulse gradually transformed itself into green and blue, with expansion into the IR direction occurring as well. Soon it was discovered that supercontinuum could also be obtained in tapered fibers [21].

The central rod in the fiber preform had been surrounded by a number of hollow tubes. When drawn down to fiber scale, the inner "core" was surrounded by air that presented a vastly larger index contrast than found in normal fiber (where the contrast may be ~0.01). Consequently, a single-mode microstructure fiber would have a much smaller diameter for a given wavelength, for example 1.7 instead of 5 micrometers for 800 nm light. This meant that a laser beam focused into this fiber would have a ~tenfold higher intensity and generate ~tenfold larger self-induced phase shifts during the pulse because of the Kerr effect, which is analogous to the quadratic Stark effect in atoms. Suddenly, researchers learned that white light could be generated by normal femtosecond oscillators without expensive and low-repetition-rate amplifiers!

Researchers also realized that the special fiber could help with the phase matching needed for a big coupling to accrue. In the "holey" fiber, a number of fiber parameters could be designer variables: the basic glass and its dispersion, the core size, the fractional angular coverage of supporting web, the size, and number of surrounding air holes. By design, it proved possible to make a strong cancellation between dispersion of the fiber core material and the dispersion associated with the geometric structure. In this way an input pulse could propagate vastly longer distances — millimeters rather than micrometers — before its peak intensity was diminished by the different propagation speeds of its spectral components.

Knowing that the pulses were so gentle the fiber was not damaged and that all the frequency components would be cross-coupled together by the nearly constant propagation speed, we predicted the output of the fiber would be a *spectral comb of coherent frequencies*. Of course, for precision metrology, this idea would have to be tested.

The first question was: how can we get some "Magic Rainbow Fiber?" Our combined approaches to friends, colleagues, and administrators at Bell Labs all came to nothing, apparently because of the lawyers there. Luckily for the JILA team, its most recently recruited colleague, Steve Cundiff, had been in a nearby group in the same part of Bell Labs. By some unknown means, the JILA team came up with a sample of "Magic Fiber" to test by November 1999. The Garching group teamed up with the powerful fiber group of P. St. J. Russell at the University of Bath (UK), which had been working with both microstructure and tapered fibers. New results began immediately rolling in [21, 22], and the publication competition began in earnest! (Since then, many alternatives to the Magic Fiber approach have emerged, including supercontinuum generation in tapered fibers [21], octave-spanning laser oscillators without the use of external fibers, mode-locked fiber lasers with some highly nonlinear ordinary fiber for spectral broadening, and schemes incorporating difference-frequency generation to determine the carrier-envelope-offset frequency with combs spanning less than an octave.)

At this point, the Boulder team had a significant advantage. They had already worked on laser stabilization and optical frequency standards for many years because of JILA's affiliation with the Boulder campus of the National Institute of Standards and Technology (NIST). Indeed, the authors of this foreword first met in Novosibirsk in 1969 at a conference organized by the late Veniamin Chebotayev on the topic of stabilized lasers. There, JLH presented his progress with a methane-stabilized He-Ne laser.

While other researchers were busy improving cw and pulsed lasers, national metrology and standards laboratories around the world had been trying to verify the frequencies of their "as-maintained" national wavelength standards. The first such measurements occurred in Boulder and led, in 1972, to the measurement of the frequency and wavelength of the methane standard. This measurement, in turn, led to a new and definitive value for the speed of light. Other national laboratories joined in, and a long discussion ensued about the philosophical and practical issues associated with calculating meters from the frequency of light rather than simply adopting new wavelength standards as they became available. Within 10 years, national laboratories in Canada, the United Kingdom, Japan, Germany, and Gaithersburg, Maryland, had confirmed parts of the Boulder work and the frequency of the He-Ne iodine-stabilized red laser had been determined. In 1983, the meter was redefined based on the speed of light.

The reproducibility of most of the reference lasers developed during this era was so good that their imperfections had little practical consequence for length metrology. Still, the optical frequency standards business continued to develop as researchers sought better designs and new reference transitions. More importantly, each nation wanted to confirm its own standards at the highest level.

Both NIST and PTB built very good systems to measure the calcium intercombination transition at 657 nm, for example. In addition, the PTB team built up one of the best rf-to-optical harmonic frequency “chains,” which took advantage of several decades of work on system components. While PTB’s “traditional” frequency chain worked well, its sheer complexity was daunting. The German laboratory’s state-of-the-art measurement [23] of the Ca frequency was published in 1996 and had a frequency uncertainty of ~ 430 hertz, arising from both the Ca standard and the measurement scheme. (By 2003 using femtosecond comb techniques, both NIST and PTB were reporting uncertainties for this frequency in the ~10 hertz range, limited mostly by interesting spectroscopic issues with the optical Ca standard.)

Its experience with optical frequency standards allowed the Boulder team at JILA to take Ranka et al.’s groundbreaking discovery of the properties of Magic Fiber and run with it. The Boulder team was the first to measure (and control) the carrier-envelope-offset frequency with a ν-to-2ν self-referenced comb [24]. With this method, they determined the frequency offset of the comb lines from the positions of harmonics of the repetition rate. Some known optical frequencies were confirmed. Within a few months, the group was also attempting to generate femtosecond pulses of controlled shape, in which the pulse-to-pulse carrier-envelope-offset phase was under the experimenter’s control.

The absolute frequency measurements in the pre-comb epoch (prior to 1999) were suddenly such a bother! The simplicity and efficiency of the new comb-based measurements attracted wide interest and provided a huge boost to the optical frequency metrology and standards field. Some parts of a total optical frequency metrology system even became available commercially (see, for example, http://www.menlosystems.com/*). Five years into the optical frequency comb epoch, the once-independent laser research subcommunities are now extremely interdependent, at least as viewed from the perspective of results. Of course, each advance has been primarily an independent step, attractive in its own context. Which market planner could have organized this beauty?

One question remains, however: how can we be sure of the frequency comb results? The accuracy of comb techniques has been the subject of a number of tests, but no problems have turned up so far. JILA has used the comb technique to measure “known” optical standards. Rather than discovering limitations of the comb technique, we have typically found that these measurements refine our knowledge of the “known” standard [25]. Thus the obvious method of testing the comb by measuring a physical standard by both methods *is not successful because of limitations of the standards themselves*.

Using another approach, the Garching team compared an octave-spanning frequency-comb synthesizer [26] with the more complex frequency synthesizer used in their 1999 hydrogen-frequency measurement. By starting with a common 10 MHz rf reference and comparing comb lines near 350 THz, they verified agreement within a few parts in 10^{16}; the precision of the experiment was probably limited by Doppler shifts due to air pressure changes or thermal expansion of the optical tables.

The Garching group also made additional accuracy tests on the comb-spacing uniformity using one stage of a frequency-interval divider [14]. The interval between comb lines near the edges of the “white” spectrum could be divided to find the “center” by two methods. Or, an edge line could be combined with one near the middle to seek a dispersive effect. No problems could be found. To really press to the testing limits, comb vs comb tests — using different comb frequencies, materials, or whatever we think could be important — are probably necessary. By late 2004, tests still had discovered no problems, and the accuracy in the context of frequency combs had reached 10^{-21} [27].

As we begin the next era in optical comb research, with each group measuring specific frequencies with femtosecond comb techniques, the realization of the cesium standard frequency is likely to be the first weak point. With a day’s averaging, the GPS system can help us know our local frequency standard’s average performance, but it takes about a day to deliver an accuracy $\sim 1 \times 10^{-14}$; achieving this accuracy requires us to know the timing comparisons separately with each of the satellites used in the test. To test optical frequency combs against the highest traditional standard, modern fountain cesium clocks are usually available either by fiber link [4] or by physical transport.

Femtosecond combs are now ready to accurately measure any desired frequency such as those of some isolated hydrogen atoms at rest in a field-free vacuum, single Hg^+ or Yb^+ ions in an ion trap, or a million cold Sr atoms trapped in an optical lattice/trap. In addition, frequency comb techniques are having an impact on ultrafast physics. By making it possible to produce few-cycle pulses with a stabilized carrier-envelope phase, these tools are leading to the discovery of novel phenomena in nonlinear light-matter interactions. For example, by 2004 amplified phase-stabilized pulses had been used to produce controlled bursts of soft x-rays with time durations in the attosecond range [28].

So where do we go from here?

In contrast to the digital security of frequency measurement, in the final accuracy-defining step, it usually comes out that spectroscopic line-shape issues are what limit our results. How well do we know the connection between the center of the observed resonance line and the desired physical quantity? Resonance frequencies can be shifted by fields, laser intensity, Doppler shifts, and ... Careful treatment of such issues is part of what makes our field so fun. And, of course, many interesting measurements can be designed so they measure *differences* or *temporal changes* and hence are better isolated from such limitations. But as our experiments improve, the line-center question will continue to reappear, ideally at an increasingly reduced sensitivity.

In summary, optical frequency combs have given us some comfortable metrological headroom for pushing ideas for new optical frequency standards and for measuring interesting physical constants. And, we feel confident that the new metrology based on integer arithmetic and femtosecond combs will be sufficient to reveal shortcomings in our spectroscopy ideas and implementations.

*Use of product name for technical information only and does not constitute an endorsement by NIST.

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