

Chapter 1

Introduction

1.1 Overview

The recent introduction [1,2] of phase-stabilized, wide-bandwidth frequency combs based on mode-locked femtosecond (fs) lasers has provided optical frequency markers that may be directly linked to optical or microwave standards. Many laboratories have since constructed frequency combs for a variety of exciting applications. These include: measurements of absolute optical frequencies and precision laser spectroscopy [3], development of optical atomic clocks [4, 5], optical frequency synthesis [6] and broadband, phase-coherent spectral generation [7, 8], along with coherent synthesis of optical pulses [9], phase-sensitive extreme nonlinear optics [10] and pulse timing stabilization.

Ultrashort pulses may also be applied to study coherent evolution in atomic and molecular systems. In particular, coherent wave packet motion has been observed and even actively controlled [11]. In addition, current work involves the use of fs lasers to achieve quantum coherent control in semiconductors [12] and in studying the temporal dynamics of biological physics [13]. These studies may require pulses which are tailored to probe or indeed selectively drive the dynamical process. This necessitates precise control over the amplitude- and phase of the ultrashort pulses, otherwise known as optical pulse synthesis [14].

On the road to tailoring optical pulses, one powerful demonstration was the phase-coherence established between two independent femtosecond lasers. Coherent optical

pulse synthesis from two fs lasers has been reported in 2001 [9], where light pulses were generated with durations shorter than those obtainable from the individual lasers. This leads to an extended coherent bandwidth to be used in ultrafast experiments.

Before the advent of phase-stabilization, research showed [15] that the longitudinal modes of a 73-fs Kerr-lens mode-locked Ti:S laser [16] are uniformly distributed in frequency space within an experimental resolution of better than 10^{-16} and that the mode spacing differs from the pulse repetition frequency by less than 10^{-15} . Thus, passively mode-locked Ti:S lasers can possess an inherent stability and that is one of their most powerful features.

On an apparently independent path, precise spectroscopic studies of atoms and molecules have always been performed with continuous wave (cw) lasers, refined over the years to be very narrow spectrally, in order to enable probing of very narrow features [17]. Precision atomic and molecular spectroscopy, enabled by the progress in cw laser stabilization, has been one of the most important fields of modern scientific research, providing the experimental underpinning of quantum mechanics and quantum electrodynamics (e.g. detailed investigations of atomic and molecular structure, determination of fundamental constants, realization of time, frequency and length standards, tests of special relativity, progress in optical communications etc.).

In the first experiments using mode-locked femtosecond lasers, they served only as rulers for studying atomic systems and did not directly interrogate the atoms. Hänsch and coworkers used the comb spectrum of a fs laser to span an optical interval of 50 nm, and improved the measurement accuracy of the D_1 resonance line in cesium by almost three orders of magnitude [18], providing a new value for the fine structure constant. To measure the D_1 line, its frequency was compared against the fourth harmonic of a CH_4 -stabilized laser employed as the absolute frequency reference. The fs comb was used as a frequency ruler to measure the resulting frequency difference of 18.39 THz (this frequency interval contained roughly 244 000 comb lines). To uniquely determine the

exact number of comb lines involved and therefore the unknown frequency, an optical cavity was employed to select every 20th mode, thus increasing the pulse repetition rate by a factor of 20.

Diddams and colleagues at JILA later bridged a frequency gap of 104 THz [19], yielding an improved frequency measurement of the 1064 nm Nd:YAG laser by using a 778-nm two-photon transition in ^{85}Rb as an absolute reference. In a following experiment, the output of the laser was spectrally broadened to an optical octave in a microstructure fiber, permitting a direct measurement of an iodine-stabilized Nd:YAG, and then measurements of ‘known’ frequencies within the laser spectrum, such as the well established transitions at 633 nm and 778 nm [20].

Phase-stabilization of mode-locked lasers was enabled by the implementation of self-referencing techniques [21] by means of highly nonlinear microstructure fibers [22]. A self-referencing technique, that will be discussed in Chapter 3, was first demonstrated at JILA by Jones and coworkers [1]. They also made absolute frequency measurements using a phase-stabilized frequency comb referenced to the primary Cs standard. The stabilized frequency combs, providing a dense grid of reference frequencies that span substantial parts of the visible to near-infrared, have rapidly become extremely powerful tools for optical frequency metrology, opening the door to a new exciting era of remarkable progress. This is because atomic and molecular structural information can now be probed over a broad spectral range, with vastly improved measurement precision and accuracy enabled by this absolute frequency-based approach.

To date, however, these applications of fs lasers in frequency metrology used the frequency comb only as a reference ruler, while still employing the traditional cw laser-based spectroscopic approaches [23].

1.2 What about using the femtosecond comb directly for spectroscopy?

The main problem with using a pulsed laser for spectroscopy is the broad frequency bandwidth associated with a short pulse. The broad spectrum prevents high-precision measurements of state energies. This problem can be avoided by using a train of phase-coherent pulses, which permits frequency resolution orders of magnitude better than that associated with a single pulse.

T. Hänsch first introduced the idea of using coherent pulse trains from mode-locked lasers for the measurement of optical frequency intervals and high-resolution spectroscopy in 1976 [24]. A stable train of pulses forms a regular and discrete set of modes separated by the pulse repetition rate in the frequency domain. Hänsch showed that a sequence of pulses results in signal enhancement over single-pulse excitation for the atomic transition amplitude. In particular, he pointed out the resonant excitation probability is proportional to the squared number of pulses arriving within the atomic lifetime (for small incident light intensities).

The spectroscopic applications of this multi-pulse interference were experimentally demonstrated within the following two years. First, a train of phase-coherent pulses produced externally to the laser, by multiple reflections of a single pulse injected into an optical resonator, was utilized for Doppler-free two-photon excitation in Na [25]. Then, in 1978, the comb of a mode-locked picosecond dye laser [26] was used to measure the Na 4d fine-structure splitting, i.e. frequency intervals of ~ 1 GHz, via Doppler-free two-photon spectroscopy [27]. In this experiment, the axial mode separation was actively controlled in the frequency domain (equivalent to stabilizing the repetition frequency of a train of pulses). As the laser spectrum is scanned, resonant excitation occurs whenever the sum of two frequencies is equal to the two-photon transition frequency. The comb of lines was used as a ruler in frequency space to measure optical frequency differences.

The limitation of this technique was the small bandwidth of the available mode-locked dye lasers, preventing measurements of large optical frequency differences.

The powerful combination of cw stabilization, passively mode-locked lasers and microstructure fibers, culminating with the phase-stabilization of femtosecond lasers, has made it possible to meet some of the conditions necessary to probe atomic structure directly with a frequency comb for precise frequency measurements: (i) short pulses, i.e. wide bandwidth (ii) pulse-to-pulse coherence and (iii) absolute referencing of the frequency comb.

Many new technologies have been enabled by the phase-stabilization of femtosecond lasers. In order to be able to phase-stabilize a femtosecond laser, one needs to understand its pulse characteristics. The connection between the short pulses emitted from the laser and the powerful frequency comb generated is described in the following section.

1.3 Time- and frequency-domain description of mode-locked lasers

The output of a mode-locked laser is a very regular train of ultrashort pulses, with the interval between pulses τ usually ~ 6 orders of magnitude higher than the pulse duration. These short pulses are produced when a fixed phase relationship is established among all the lasing longitudinal modes of the laser cavity (i.e. mode-locking), resulting in their coherent addition and the formation of a light pulse. An isolated light pulse can be represented as a sinusoidal optical carrier of angular frequency ω_c , amplitude-modulated by an envelope function $\mathcal{E}(t)$, with the electric field thus given by $E(t) = \mathcal{E}(t) \exp[i(\omega_c t + \phi_0)]$. When traveling through a medium, the carrier propagates at the phase velocity, while the envelope advances at the group velocity. In general, the two velocities are different for a dispersive medium: $v_p = c/n$ and $v_g = c / \left(n + \omega \frac{\partial n}{\partial \omega} \right)$. For instance, traversing a distance l in a dispersive material gives a phase shift of $\omega_c \left(\frac{l}{v_g} - \frac{l}{v_p} \right) = \frac{l\omega_c^2}{c} \frac{\partial n}{\partial \omega} = -2\pi l \frac{\partial n}{\partial \lambda}$. This difference in the phase and group velocities

inside the laser cavity causes the phase between the optical carrier and the peak of the envelope to evolve between successive pulses in the train. Let $\delta\phi$ denote the amount by which the carrier-envelope phase changes from pulse to pulse. After including this pulse-to-pulse phase-shift, the electric field of a train of pulses for a fixed spatial coordinate is

$$E(t) = \sum_n \mathcal{E}_n(t - n\tau) \exp[i(\omega_c t + \phi_0 - n\omega_c \tau + n\delta\phi)]. \quad (1.1)$$

By taking its Fourier Transform [28] we obtain the following expression for the angular frequencies

$$\omega_N = \frac{N 2\pi}{\tau} - \frac{\delta\phi}{\tau}. \quad (1.2)$$

Thus, the spectrum of the mode-locked femtosecond laser consists of a wide comb of discrete optical frequencies, offset from the exact harmonics of the repetition rate by a frequency proportional to the pulse-to-pulse carrier-envelope phase shift [Fig. 1.1]. The spectrum is described by the simple relation $\nu_N = Nf_r + f_0$, where N is a large integer on the order of 10^6 , $f_r = \frac{1}{\tau}$ is the pulse repetition rate and $f_0 = -\frac{1}{2\pi} \delta\phi f_r$ is the carrier-envelope offset frequency. Note that the relevant quantity here is $\delta\phi$ modulo 2π , i.e. f_0 is bounded by f_r .

The two laser parameters are dynamic quantities, sensitive to environmental perturbations affecting the laser cavity. Not surprisingly, there are no locking mechanisms between the pulse envelope and the optical carrier inside an unstabilized laser, so their relative phase $\delta\phi$ experiences fluctuations between successive pulses. Consequently, for spectroscopic studies with a femtosecond laser, both comb parameters f_r and f_0 need to be actively stabilized.

As a reminder, absolute optical frequency measurements are made with respect to the ground state hyperfine splitting in ^{133}Cs at 9.192631770 GHz, which provides the current definition for the second. This leap from the optical to the radio-frequency domain has been a daunting task in frequency metrology for the last 30 years, before

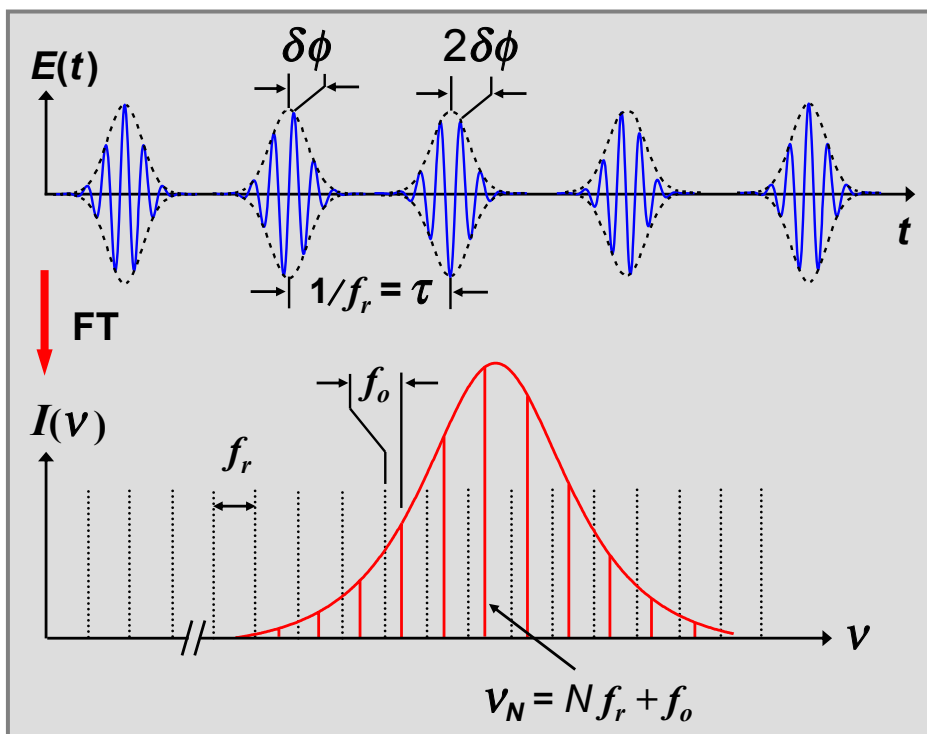


Figure 1.1: Correspondence between the time and frequency domains for a Ti:S mode-locked laser. In the time domain, the carrier-envelope phase changes at a defined rate between successive pulses in the train. In the frequency domain, the comb lines are spaced by the laser repetition rate f_r and the comb is shifted from integer multiples of f_r by an offset frequency f_0 . The Ti:S laser used in the experiment is centered at 778 nm and has a FWHM bandwidth of ~ 55 nm.

the introduction of the mode-locked approach. Because of the complexity involved, only a few harmonic frequency chains were ever implemented, for very specific optical frequency measurements. A few examples are: the 88 THz transition in CH₄ [29] (which led to the speed of light measurement [30]), the I₂-stabilized He-Ne transition at 473 THz [31], the 455 THz intercombination transition in ⁴⁰Ca [32], the $5s\ ^2S_{1/2} \rightarrow 4d\ ^2D_{5/2}$ transition at 445 THz in ⁸⁸Sr⁺ [33], the 2S-12D transitions in hydrogen and deuterium at 799 THz [34]. Stabilized frequency combs allow one to phase-coherently link any unknown optical frequency within the comb spectrum directly to a primary microwave standard.

1.4 Direct Frequency Comb Spectroscopy

Following our own theoretical studies [35], we take advantage of the phase-stable, wide-bandwidth femtosecond pulse train to bridge the fields of high-resolution spectroscopy and ultrafast science, in a spectroscopic study of laser-cooled ⁸⁷Rb atoms. This approach, which we call Direct Frequency Comb Spectroscopy (DFCS), involves using light from a comb of appropriate structure to directly interrogate atomic levels and to study time-dependent quantum coherence.

By utilizing a stabilized fs comb, multiple atomic states may be simultaneously and directly excited, by tuning the appropriate comb lines into resonance, and the subsequent dynamics may be probed. Furthermore, given that the comb has two independent degrees of freedom, it is always possible to simultaneously satisfy two-photon as well as one-photon resonance conditions.

In fact, DFCS may be applied to determine absolute frequencies for atomic transitions anywhere within the comb bandwidth. The entire transition spectrum can be efficiently retrieved by a quick scan of the laser repetition frequency. This obviates the need for a broadly tunable and absolutely referenced cw laser and is especially useful in the study of multi-photon processes where several laser sources may be required. Thus,

optical frequency combs are a highly efficient tool for precise studies of atomic structure.

In this work, we apply DFCS to determine absolute atomic transition frequencies for one- and two-photon processes, as illustrated in Fig. 1.2. DFCS enables studies of coherent pulse accumulation and multipulse interference, permitted by the relatively long-lived 5D and 7S states. The observed coherent pulse accumulation and interference effects are well modeled by our density matrix theory describing the interaction of the femtosecond comb with the atoms, as discussed in Chapter 2. We use the theory results to construct transition spectra to compare against the experimental spectra.

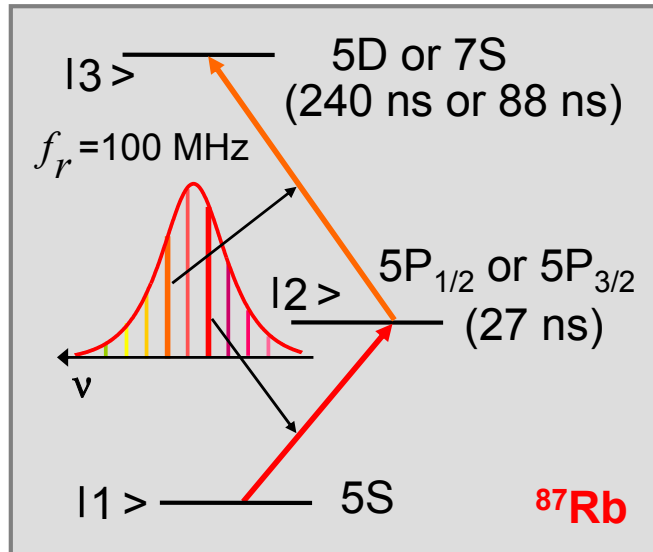


Figure 1.2: Schematic of the ^{87}Rb energy levels participating in the 5S-5D and 5S-7S two-photon transitions and 5S-5P one-photon transitions. These are the relevant levels probed using direct frequency comb spectroscopy (DFCS).

We present the details of the implementation of the femtosecond stabilization scheme and the MOT setup in Chapter 3. These are integrated together for spectroscopic studies in Chapter 4.

As in the case of precision spectroscopy performed with cw lasers, the use of the femtosecond comb as a precision probe requires a careful understanding of all systematic effects. We find that the dominant effects are the mechanical effect of the optical comb

on the atomic motion, the light shift by the probe laser, and the Zeeman frequency shifts. In Chapter 4, we address the mechanical action of the probe laser by using a counterpropagating beam configuration. We observe Stark shifts generated by detunings from the intermediate states in the two-photon processes and we reduce their effect by always probing on resonance with the intermediate state of interest.

We first study the 5S-5D two-photon transitions in ^{87}Rb , leading to high-resolution spectroscopy for some of the transitions, as presented in Chapter 5. The measurement results are comparable to the highest resolution measurements made with cw lasers. By determining the previously unmeasured absolute frequency of the 5S-7S two-photon transitions in ^{87}Rb , we show that prior knowledge of atomic transition frequencies is not essential for this technique to work, and indicate that it can be applied in a broad context. When resonant enhancement is enabled by a comb component tuned near an intermediate 5P state, we observe two-photon transitions occurring between initial and final states that differ by one unit of the total angular momentum ($\Delta F = \pm 1$), which are absent for far-detuned intermediate states. This capability of accessing adjacent excited hyperfine levels from the same ground state allows for direct measurements of hyperfine splittings.

We then use DFCS to measure one-photon transitions, and choose the 5S-5P transitions as an example, as detailed in Chapter 6. The measurement of 5P states is also carried out indirectly via the 5S-5D two-photon transitions, by studying their resonant enhancement when comb components are scanned through the intermediate 5P states. We compare the 5P measurements obtained via one-photon and two-photon DFCS and clearly demonstrate the importance of population transfer in working with multilevel systems probed by multiple comb components. Incoherent processes such as optical pumping govern the population dynamics beyond the atomic decoherence time.

For the case of the indirect 5P frequency measurements via the 5S-5D two-photon transitions, the experimental data do not yield directly the lineshape. In this case, we

use the theory prediction to adjust the raw data and retrieve a Lorentzian lineshape, as described in Chapter 6.

The optical coherence of a phase-stabilized pulse train provides a spectral resolving power approaching that of state-of-the-art cw lasers. At the same time, the narrow linewidth of individual comb lines permits a precise and efficient determination of the global energy-level structure, providing a direct connection among the optical, terahertz, and radio-frequency domains.