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Optical frequency combs have revolutionized precision metrology by enabling measurements of optical frequencies, with implications both for fundamental scientific questions and for applications such as fast, broadband spectroscopy. In this thesis, I describe the development of comb generation platforms with smaller footprints and higher repetition rates, with the ultimate goal of bringing frequency combs to new applications in a chip-integrated package. I present two new types of frequency combs: electro-optic modulation (EOM) combs and Kerr-microresonator-based frequency combs (microcombs). First I describe the EOM comb scheme and, in particular, techniques for mitigating noise in the comb generation process, and I present the results of a proof-of-principle metrology experiment and some possible applications. Then I discuss developments in microcomb technology. I present novel soliton crystal states, which have highly structured fingerprint optical spectra that correspond to ordered pulse trains exhibiting crystallographic defects. These pulse trains arise through interaction of the solitons with avoided mode-crossings in the resonator spectrum. Next, I describe the direct and deterministic generation of single microresonator solitons using a phase-modulated pump laser. This technique removes the dependence on initial conditions that was formerly a universal feature of these experiments, presenting a solution to a significant technical barrier to the practical application of microcombs. I also discuss generation of Kerr combs in the Fabry-Perot (FP) geometry. I introduce a nonlinear partial differential equation describing dynamics in an FP cavity and discuss the differences between the FP geometry and the ring cavity, which is the geometry used in previous Kerr-comb experiments. Finally, I discuss a technique for reducing the repetition rate of a high-repetition-rate frequency comb, which will be a necessary post-processing step for some applications. I conclude with a discussion of avenues for future research, including the chip-integration of Fabry-Perot Kerr resonators and the use of band-engineered photonic crystal cavities to further simplify soliton generation.

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Chapter 1

Introduction

The invention of the optical frequency comb two decades ago provided a revolution in precision measurement by dramatically improving the resolution with which we can measure time. This revolution came about through the development of a simple scheme (that required markedly less simple advancements in capabilities in nonlinear optics) by which the hundreds-of-terahertz-scale optical frequencies of a mode-locked laser could be effectively measured by electronics with bandwidth limitations on the gigahertz scale. The first frequency combs immediately permitted measurement of fundamental properties of matter, for example the electronic transition frequency in hydrogen, with unprecedented levels of precision. Since those first demonstrations, optical frequency combs have played an integral part in experiments and applications in contexts ranging from record-setting optical clocks, systems for ultra-low-noise microwave synthesis, broadband spectroscopy applications, and stable long-term calibration of astronomical spectrographs for exoplanet detection. Further development of the technology beyond the first stabilization of the Ti:sapphire laser that heralded the frequency comb's arrival has enabled frequency combs to reach applications across many wavelength bands. The technology is reaching maturity, and frequency combs have been commercially available for some time.

In the last decade, methods for generating optical frequency combs without a mode-locked laser have suggested the possibility of bringing their capabilities to a wide set of applications outside the controlled environment of the research laboratory. These new frequency combs come with higher repetition rates, which makes them particularly attractive for applications where high power per comb mode, individual accessibility of comb modes, and fast acquisition times are desired; these applications include arbitrary microwave and optical waveform generation, telecommunications, and broadband, fast-acquisition-time spectroscopy. Moreover, these combs come with lower size, weight, and power (SWAP) requirements, which will enable them to bring the features that make mode-locked laser-based combs attractive into the field, enabling e.g. direct optical frequency synthesis on a chip [1].

This thesis focuses on this second generation of optical frequency combs. The bulk of the thesis covers microresonator-based frequency combs, and especially the nonlinear dynamics involved in the generation of these frequency combs via the Kerr nonlinearity. The penultimate chapter presents a second method for generating a high-repetition-rate frequency comb without modelocking that is based on active modulation of a seed CW laser and subsequent nonlinear spectral broadening. In the final chapter, I present experimental and theoretical investigations of a technique for repetition-rate reduction of frequency combs, which may prove useful for adapting low-SWAP combs and their intrinsically high repetition rates to some applications as the technology continues to develop.

In the remainder of this chapter, I discuss the basic properties of frequency combs and explain how the optical frequencies making up a comb can be fully determined by electronics operating with gigahertz-scale bandwidths.

1.1 Optical frequency combs

An optical frequency comb is obtained by fully stabilizing the spectrum of an optical pulse train. The first frequency combs came about through full frequency-stabilization of modelocked lasers; this thesis focuses on frequency combs with pulse trains generated through other means.

1.1.1 Optical pulse trains and their spectra

In the time domain, a frequency comb consists of a train of uniformly spaced optical pulses arriving at the pulse train's repetition rate f_r . These pulses are typically very short compared to their repetition period $T = 1/f_r$. In the frequency domain, the comb consists of a set of modes that

are spaced by f_r in frequency and that have amplitudes determined by an overall spectral envelope centered at the optical carrier frequency, with bandwidth inversely related to the temporal duration of the pulses. The usual description of a frequency comb, which is natural for modelocked-laser-based combs that are not derived from a CW laser, gives the frequencies of these modes as

$$\nu_n = nf_r + f_0, \tag{1.1}$$

where $n \sim f_{carrier}/f_r$ for the optical modes that make up the comb and f_0 is the carrier-envelope offset frequency, which may be defined to be between 0 and f_r . The offset frequency results from the pulse-to-pulse evolution of the carrier wave underneath the temporal intensity envelope of the pulses due to a difference in group and phase velocities. An equivalent representation of the frequencies of the comb that is more natural for frequency combs directly derived from a CW laser, as described in this thesis, is

$$\nu_{\mu} = \nu_c + \mu f_r,\tag{1.2}$$

where ν_c is the frequency of the CW laser, the 'pump' or 'seed' laser, from which the frequency comb is derived and μ is a pump-referenced mode number, in contrast with the zero-referenced mode number n of Eq. 1.1. Fig. ?? depicts the properties of a frequency comb in the time domain and the frequency domain.

It is useful to consider a mathematical treatment of an optical pulse train to understand the relationships presented above. In the time domain, the electric field E(t) of the pulse train consists of optical pulses that arrive periodically and have baseband (centered at zero frequency) field envelope A(t) multiplying the carrier wave of angular frequency ω_c :

$$E(t) = \sum_{k=-\infty}^{\infty} A(t - kT)e^{i\omega_c t}.$$
 (1.3)

Here, T is the repetition period of the pulse train. Eq. 1.3 can be viewed as describing a laser of angular frequency ω_c with a time-varying amplitude. This temporal modulation leads to the distribution of the power across a spectrum whose width scales inversely with the temporal duration of A. Intuitively, the spectrum of the comb is the spectrum of the periodic baseband field envelope

 $\Sigma_k A(t-kT)$, shifted by the multiplication with $e^{i\omega_c t}$ so that it is centered around the optical carrier. More formally, we can calculate the spectrum $|\mathcal{F}\{E\}|^2$ by calculating

$$\mathcal{F}\left\{E\right\}(\omega) \sim \left(\sum_{k=-\infty}^{\infty} \mathcal{F}\left\{A(t-kT)\right\}\right) * \delta(\omega - \omega_c),$$
 (1.4)

which results from the convolution (denoted by *) theorem for Fourier transforms. We use the Fourier transform's property that a temporal translation results in a linear spectral phase shift to obtain:

$$\mathcal{F}\left\{E\right\} \sim \left(\mathcal{F}\left\{A\right\} \times \sum_{k=-\infty}^{\infty} e^{-i\omega kT}\right) * \delta(\omega - \omega_c).$$
 (1.5)

The quantity $\Sigma_k e^{-i\omega kT}$ is the Fourier-series representation of the series of δ -functions $\Sigma_\mu \delta(\omega - 2\pi\mu/T)$, so we get

$$\mathcal{F}\left\{E\right\}\left(\omega\right) \sim \left(\mathcal{F}\left\{A\right\} \times \sum_{\mu=-\infty}^{\infty} \delta\left(\omega - 2\pi\mu/T\right)\right) * \delta(\omega - \omega_c),$$
 (1.6)

and performing the convolution leads to the replacement of ω with $\omega - \omega_c$, leading to:

$$\mathcal{F}\left\{E\right\} \sim \sum_{\mu=-\infty}^{\infty} \delta\left(\omega - \omega_c - \mu\omega_r\right) \mathcal{F}\left\{A\right\} (\omega - \omega_c). \tag{1.7}$$

This expression indicates that the spectrum of the comb has frequency content at modes $\nu_{\mu} = \nu_c + \mu f_r$, and that their amplitudes are determined by the spectrum of the baseband field envelope, shifted up to the optical carrier frequency ν_c . This is the natural formulation in the case of a comb derived from a CW laser, but it obscures the carrier-envelope offset frequency in the difference between ν_c and the nearest multiple of the repetition rate, so that f_0 is the remainder of $\nu_c \div f_r$. In practice, if f_r is known, then a measurement of f_0 is equivalent to a measurement of the frequency of the input CW laser.

1.1.2 Frequency stabilization of optical pulse trains

The scientific need for a method to measure optical frequencies motivated the development of optical frequency combs. While the measurement bandwidth of electronic frequency counters has improved since 1999, it remains limited to frequencies roughly one *million* times lower than the frequency of, e.g., visible red light. Frequency combs present a method for measurement of the

unknown frequency f_{opt} of an optical signal through heterodyne with a frequency comb—if f_{opt} falls within the bandwidth of the frequency comb, then the frequency of the heterodyne between the comb and the signal is guaranteed to be less than $f_r/2$, which is typically a frequency that can be measured electronically, at least for modelocked-laser-based combs. Therefore, if the frequencies of the comb are known, measurement of the heterodyne of the comb with the signal reveals the frequency of the signal, provided that the comb mode number n, as defined by Eq. 1.1, can be determined. This can be done via a wavelength measurement if sufficient precision is available, or by measuring the change $\partial f_b/\partial f_r = \pm n$, where f_b is the measured frequency of the beat.

The unique utility of the optical frequency comb lies in the fact that measurement of the two frequencies f_r and f_0 , along with a measurement of the spectral envelope, is sufficient to determine the optical frequencies of all of the modes of the comb, thereby enabling frequency measurement of optical signals. Measurement of the repetition rates of optical pulse trains was possible for many years before the realization of optical frequency comb technology, as this can be done by simply impinging the pulse train on a photodetector. It was the confluence of several technological developments around the turn of the twenty-first century that allowed detection and measurement of the carrier-envelope offset frequency, thereby enabling creation of fully-stabilized modelocked-laser pulse trains: optical frequency combs.

The carrier-envelope offset frequency of a pulse train is challenging to measure because it describes evolution of the optical carrier wave underneath the intensity envelope, and therefore cannot be measured through straightforward detection of the intensity of the pulse train. Presently, the most straightforward way to measure f_0 is f-2f self-referencing, which is illustrated in Fig.??. make fig This can be performed only with a pulse train whose spectrum spans an octave—a factor of two in frequency. Given such an octave-spanning supercontinuum spectrum, a group of modes near mode number N is frequency-doubled in a medium with the $\chi^{(2)}$ nonlinearity[7]. This frequency-doubled light is heterodyned with the native light in the supercontinuum with mode number near 2N. The

frequency of the resulting beat f_b is:

$$f_b = f_{doubled} - f_{native} \tag{1.8}$$

$$= 2(Nf_r + f_0) - (2Nf_r + f_0)$$
(1.9)

$$= f_0. (1.10)$$

Generating the necessary octave-spanning supercontinuum spectrum typically requires nonlinear spectral broadening of the pulse train after its initial generation, except for in specific, carefully engineered cases. Achieving the required degree of spectral broadening while preserving the coherence properties of the pulse train is a significant challenge—typically this requires launching a train of high energy (\sim 1 nJ), temporally short (\leq 100 fs) pulses into the spectral-broadening stage, and meeting these requirements is one of the important engineering considerations in designing optical frequency comb systems, as discussed in Chapters ?? and ??.

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