ThesisTitle

by

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Other Degrees

A thesis submitted to the

Faculty of the Graduate School of the

University of Colorado in partial fulfillment

of the requirements for the degree of

Doctor of Philosophy

Physics Physics

2018

This thesis entitled:
ThesisTitle
written by Daniel C. Cole
has been approved for the Physics Physics

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Cole, Daniel C. (Ph.D., Physics)

ThesisTitle

Thesis directed by Dr. Scott A. Diddams

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.

Acknowledgements

The work in this thesis would not have been possible...

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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 terahertz-scale optical frequencies of a mode-locked laser could be effectively measured by electronics operating much more slowly, with bandwidth limitations on the gigahertz scale. The new frequency comb technology 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 myriad contexts, including 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 enabled these applications and others, and combs are now versatile tools for measurement in many contexts 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 that go beyond the 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 and lower size, weight, and power (SWAP) requirements, making them particularly appropriate for applications like arbitrary microwave and optical waveform generation, telecommunications, and broadband, fast-acquisition-time spectroscopy. Moreover, low-SWAP combs bring the features that make mode-locked laser-based combs attractive to 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 parametric generation of these frequency combs based on 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 c.w. 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 basics of optical frequency comb technology.

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 c.w. laser, gives the frequencies of these modes as

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

where $n \sim f_{carrier}/f_r \gg 0$ for the optical modes that make up the comb and f_0 is the carrier-envelope offset frequency. 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 c.w. 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 c.w. 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 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 periodically-recurring optical pulses with baseband (centered at zero frequency) field envelope $A(t)$ multiplying the carrier wave of angular frequency ω_c:

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

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 a broadband spectrum for E. 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

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

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:

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

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

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

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

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

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 c.w. laser, but it hides 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 c.w. 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 two microwave frequencies, f_0 and f_0, 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.

have I made clear that these are microwave frequencies

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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.**??**. 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. This frequency-doubled light is heterodyned with the native light in the supercontinuum with mode number near $2N$. The frequency of the resulting beat is f_0:

make fig

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

$$= 2(Nf_r + f_0) - (2Nf_r + f_0) \tag{1.9}$$

$$= f_0. \tag{1.10}$$

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

cite Tara, others

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; this point is relevant to chapters **??** and **??**.

1.2 Emerging applications for frequency combs

The work presented in this thesis is motivated by a set of applications that can leverage high frequency-comb repetition rate; or low frequency-comb size, weight, and power; or both. In general these applications exist outside the laboratory, in fields such as

Here i suppose I'd like to present some example applications

Chapter 2

Microresonators

This chapter introduces the basic physics of Kerr-nonlinear optical ring resonators, and the two subsequent chapters describe results obtained in these systems. correct?

An optical ring resonator, shown schematically in Fig. **??**, guides light around a closed path in a dielectric medium by total internal reflection, similar to the mechanism that guides light in an optical fiber. The utility of these resonators lies in the fact that light can circulate for many roundtrips before it is coupled out or dissipated, which means that very high circulating intensities can be achieved.

A ring resonator supports propagating guided optical *modes* of electromagnetic radiation that occur at (vacuum) wavelengths that evenly divide the optical round-trip path length: $\lambda_m = n_{eff}(\lambda_m)L/m$, with associated resonance frequencies $\nu_m = c/\lambda_m = mc/n_{eff}(\nu_m)L$, leading to constructive interference from round-trip to round-trip. Here L is the physical round-trip length of the resonator, m is the azimuthal mode number, and $n_{eff}(\lambda_m)$ is an effective index of refraction that depends on the resonator geometry and the mode's transverse mode profile (see e.g. [**REFHERE**] for more information). The free-spectral range f_{FSR} of a resonator is the *local* frequency spacing between modes, calculated via:

$$f_{FSR} \approx \nu_{m+1} - \nu_m \approx \nu_m - \nu_{m-1}, \tag{2.1}$$

$$= \frac{\partial \nu_m}{\partial m}, \tag{2.2}$$

$$= \frac{c}{n_{eff}(\nu)L} - \frac{mc}{n_{eff}^2(\nu)L}\frac{\partial n_{eff}}{\partial \nu}\frac{\partial \nu}{\partial m}, \tag{2.3}$$

$$\Rightarrow f_{FSR} = \frac{c/L}{\left(n_{eff} + \nu\frac{\partial n_{eff}}{\partial \nu}\right)} = \frac{c}{n_g L} = 1/T_{RT}, \tag{2.4}$$

where $n_g = n_{eff} + \nu\frac{\partial n_{eff}}{\partial \nu}$ is the group velocity of the mode and T_{RT} is the mode's round-trip time. Importantly, both intrinsic material dispersion and geometric dispersion resulting from, e.g., different sampling of core versus cladding material properties for different transverse mode profiles, lead to a frequency dependence for each of the parameters n_{eff}, n_g, and f_{FSR}, and a resulting non-uniform spacing in the cavity modes in frequency despite the linearity of ν_m in m.

Unless special efforts are made, ring resonators are typically multi-mode, meaning that many different transverse mode profiles are supported. To calculate the frequency-dependent effective index $n_{eff}(\nu)$, thereby enabling calculation of the resonance frequencies and wavelengths, one must solve Maxwell's equations for the resonator geometry. Except in special cases of high symmetry [**microsphereresonators**], this is typically done numerically using finite-element modeling tools like COMSOL. The modes of an optical resonator, both within a mode family defined by a transverse mode profile and between mode families, must be orthogonal.

is there a good citation here?

The timescale over which circulating photons are dissipated in a resonator is fundamental to its fitness for applications. This is quantified by the basic relation for the number of circulating photons $N(t) = N_o e^{-t/\tau_\gamma}$ in the presence of solely linear loss, which defines the photon lifetime τ_γ. Two commonly used practical quantities are linked to the photon lifetime: the resonator finesse $\mathcal{F} = 2\pi\tau_\gamma/T_{RT}$, which for a ring resonator can be interpreted literally as the azimuthal resonator angle traced out by a typical photon over its lifetime; and the resonator quality factor $Q = \omega_c\tau_\gamma$, the phase over which the optical field evolves during the photon lifetime. The lifetime of a photon at a particular frequency is related to the cavity's full-width at half-maximum (FWHM) linewidth as we can calculate through a Fourier transform of the field $E(t) \propto \sqrt{N(t)}$ with angular carrier frequency ω_c:

$$\mathcal{F}\{E\}(\omega) \propto \int_0^\infty dt\, e^{-\left(\frac{1}{2\tau_\gamma}+i(\omega_c-\omega)\right)t}, \tag{2.5}$$

which immediately yields the Lorentzian lineshape

$$|\mathcal{F}\{E\}|^2 \propto \frac{1}{(\omega-\omega_c)^2+\frac{1}{4\tau_\gamma^2}}, \tag{2.6}$$

with FWHM linewidth $\Delta\omega = 1/\tau_\gamma$. With this relationship, the finesse and quality factor can be rewritten as simple ratios of the relevant frequencies: $\mathcal{F} = f_{FSR}/\Delta\nu$; $Q = \nu_c/\Delta\nu$, where $\Delta\nu = \Delta\omega/2\pi$.

2.1 Basic experiments

In a typical microresonator frequency-comb experiment, a frequency-tunable pump laser is coupled evanescently into and out of the resonator using a tapered optical fiber (for e.g. free-standing silica disc resonators) or a bus waveguide (for chip-integrated resonators). The frequencies of the cavity modes can be identified by recording the transmission of the pump laser past the resonator as the pump frequency is varied, provided that sufficient overlap between the fiber field and the resonator field is achieved. In order to couple light into the microresonator so that substantial build-up of the intracavity field occurs, the pump laser must be held at a frequency near a cavity resonance. However, the thermal response of the microresonator introduces complexities into this process; in particular, in materials with positive thermo-optic coefficient $\partial n/\partial T$ the cavity resonance wavelength increases with increased circulating intensity, as intrinsic material absorption leads to heating of the resonator. This basic fact leads to so-called thermal 'bi-stability' behavior [**Carmon2004**] — it is possible to maintain 'blue' laser detuning $\nu_{laser} - \nu_{resonance} > 0$ indefinitely due to naturally occurring thermal self-locking, while 'red' laser detuning $\nu_{laser} - \nu_{resonance} < 0$ is thermally unstable. A second effect of the thermal shift of the resonance frequencies is that, when any substantial pump-laser power is coupled into the resonator, the observed cavity lineshape is no longer Lorentzian, and the symmetry between increasing- and decreasing-pump-frequency scans is broken, as shown in Fig. **??**.

2.2 Microring resonator Kerr frequency combs

The high circulating optical intensities accessible in resonators with long photon lifetimes find immediate application in the use of microresonators for nonlinear optics. The experiments described in this thesis are conducted in silica microresonators. Silica falls into a broader class of materials that exhibit both centro-symmetry, which dictates that the second-order nonlinear susceptibility $\chi^{(2)}$ must vanish, and a significant third-order susceptibility $\chi^{(3)}$. The n^{th}-order susceptibility is a term in the Taylor expansion describing the response of the medium's polarization to an external electric field: $P = P_0 + \epsilon_0\chi^{(1)}E + \epsilon_0\chi^{(2)}E^2 + \epsilon_0\chi^{(3)}E^3 + ...$ The effect of $\chi^{(3)}$ can be described in a straightforward way as a dependence of the refractive index on the local intensity, $n = n_0 + n_2 I$[**somethingelse**], where $n_2 = \frac{3\chi^{(3)}}{4n_0^2\epsilon^0 c}$[**CosoAndSolis**, **alsoLLbook?**]. The intensity-dependence of the refractive index is referred to as the optical Kerr effect.

The combination of the Kerr effect and the high circulating intensities that are accessible in high-finesse cavities provides a powerful platform for nonlinear optics. Specifically, the Kerr effect (or third-order susceptibility $\chi^{(3)}$) enables self-phase modulation, cross-phase modulation, and four-wave mixing, the last of which is depicted schematically in Fig. **??**.

In 2007, a remarkable result heralded the beginning of a new era for frequency comb research. Del'Haye et al reported *cascaded four-wave mixing* in toroidal silica microcavities on silicon chips, the result of which was a series of many co-circulating optical fields that were uniformly spaced by f_{rep} ranging from 375 GHz to ~750 GHz (depending on the platform)[**DelHaye2007**]. Measurements indicated that the frequency spacing was uniform to a precision of 7.3×10^{-18}, thereby establishing that the output of the system was a frequency comb. This result built on previous demonstrations of few-mode parametric oscillations in microresonators [**Kippenberg2004**, **Savchenkov2004**, **Agha2007**], and showed that the non-uniform distribution of cavity resonance frequencies could be overcome to generate an output with equidistant frequency modes. Demonstrations of frequency-

from initial kerr comb paper

comb generation in other platforms followed shortly, with realizations in ... A second important development occurred in 2012, when Herr et al reported the generation of frequency combs corresponding in the time domain to single circulating optical ‘soliton‘ pulses. In fact, it is now recognized that passive fiber ring resonators are formally equivalent to Kerr microrings, and solitons were generated in these systems in 2010.

Kerr-comb generation can be motivated and partially understood through the cascaded four-wave mixing picture, but the phase and intensity degrees of freedom for each comb line mean that this picture gives rise to a very rich space of comb phenomena, as evidenced by the various behaviors reported in Kerr microresonators. A very useful model for understanding this rich space is the Lugiato-Lefever equation (LLE), which was shown to describe microcomb dynamics by Chembo and Menyuk [**Chembo2013**] through Fourier-transformation of a set of coupled-mode equations describing four-wave mixing and by Coen et al [**Coen2013**] through time-averaging of an Ikeda map for a low-loss resonator (as first performed by Haelterman, Trillo, and Wabnitz [**Haelterman1992**]). The LLE is a nonlinear partial-differential equation that describes evolution of the normalized cavity field ψ over a slow time $\tau = t/2\tau_\gamma$ in a frame parametrized by the ring's azimuthal angle θ co-moving at the group velocity at the frequency of the pump laser. The equation as it will be used throughout this thesis reads:

$$\frac{\partial\psi}{\partial\tau} = -(1+i\alpha)\psi + i|\psi|^2\psi - i\frac{\beta}{2}\frac{\partial^2\psi}{\partial\theta^2} + F. \tag{2.7}$$

Here F is the normalized strength of the pump laser; F and ψ are normalized so that both F^2 and $|\psi|^2$ take the value 1 at the absolute threshold for cascaded four-wave mixing: $F = \sqrt{\frac{8g_0\Delta\omega_{ext}}{\Delta\omega tot^3}\frac{P}{\hbar\Omega_0}}$, $|\psi|^2 = \frac{2g_0T_{RT}}{\hbar\omega\Delta\omega_{tot}}P = \frac{2g_0Ln_g}{c\hbar\omega\Delta\omega_{tot}}P$, where $|\psi(\theta,\tau)|^2$ is the instantaneous normalized power at the co-moving azimuthal angle θ. Here $g_0 = n_2c\hbar\Omega_0^2/(n_g^2V_0)$ is a parameter describing the four-wave mixing gain, $\Delta\omega_{ext}$ is the rate of coupling at the input/output port, $\Delta\omega_{tot} = 1/\tau_\gamma$ is the FWHM resonance linewidth, P is the pump-laser power, $\hbar$ is Planck's constant, and Ω_0 is the pump-laser frequency. The parameters n_2, n_g, and V_0 describe the nonlinear (Kerr) index (see Eqn. **??**), the group index of the mode, and the effective nonlinear mode volume at the pump frequency; L is the physical round-trip length of the ring cavity.

The LLE is formulated in terms of dimensionless normalized parameters; this facilitates comparison of results obtained in platforms with widely different experimental conditions. The parameters α and β describe the normalized frequency detuning of the pump laser and second-order dispersion of the resonator mode family into which the pump laser is coupled: $\alpha = -\frac{2(\Omega_0 - \omega_{res})}{\Delta\omega_{tot}}$, $\beta = -\frac{2D_2}{\Delta\omega_{tot}}$; here $D_2 = \frac{\partial^2\omega_\mu}{\partial\mu^2}\Big|_{\mu=0}$ is the second-order modal dispersion parameter, where μ is the pump-referenced mode number of Eq. 1.2. The parameters $D_1 = \frac{\partial\omega_\mu}{\partial\mu}\Big|_{\mu=0} = 2\pi f_{FSR}$ and D_2 are related to the derivatives of the propagation constant $\beta_{prop} = n(\omega)\omega/c$ via $D_1 = 2\pi/L\beta_1$ and $D_2 = -\frac{D_1^2}{\beta_{prop,1}}\beta_{prop,2}$, where $\beta_{prop,n} = \partial^n\beta_{prop}/\partial\omega^n$. The subscript *prop* is used here to distinguish the propagation constant from the LLE dispersion coefficients $\beta_n = -2D_n/\Delta\omega_{tot}$. Expressions for higher-order modal dispersion parameters D_n in terms of the expansion of the propagation constant can be obtained by evaluating the equation $D_n = (D_1\frac{\partial\omega}{\partial\mu})^n\omega_\mu$.

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