Cascaded Phase Matching and Nonlinear Symmetry Breaking in Fiber Frequency Combs

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We report theoretical, numerical, and experimental studies of cascaded phase matching in fiber frequency combs and show how this mechanism is directly connected to the dynamics of supercontinuum generation. In particular, linking cascaded four-wave mixing with direct higher-order nonlinear processes allows us to derive a simple phase matching condition that governs nonlinear symmetry breaking in the presence of higher-order dispersion. We discuss how this mechanism provides a physical interpretation of soliton-induced Cherenkov radiation and associated spectral recoil in terms of phase-matched frequency mixing pumped by bichromatic pump pairs in the soliton spectrum. Theoretical and numerical predictions are confirmed via experiments using both quasicontinuous wave and picosecond pulse excitation.

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The generation of optical frequency combs exploiting the Kerr nonlinearity has impacted multiple fields of physics, and there is continued interest in understanding the underlying frequency conversion mechanisms [1,2]. Surprisingly, however, there is no unified approach to describing comb generation in all third-order media. While frequency comb generation in microresonators has been described in terms of discrete four-wave mixing (FWM) between a monochromatic pump and resonator modes sequentially excited in a cascade [2,3], supercontinuum comb generation in waveguides is described exclusively in terms of the propagation dynamics of a single input pulse with a continuous spectral envelope [4]. But the spectrum of a pulse train from a mode-locked laser consists of discrete frequency components [5,6], and so the natural question that arises is, in the context of frequency comb generation, how can we relate the two physical pictures of discrete mode interactions of FWM and the propagation dynamics of a spectrally continuous pulse? In this Letter, we present a theory of cascaded FWM that resolves this question.

We first note that a unique feature of cascaded wave mixing is that sequential frequency generation mimics the effect of a higher-order nonlinear susceptibility [7–9]. A particular component in a cascade can thus be coherently amplified if the medium's chromatic dispersion allows the corresponding higher-order process to be directly phase-matched, even if all the elementary steps in the cascade are completely mismatched [8]. Although this feature has been known since the early days of nonlinear optics, applications have been mainly restricted to generating a small number of harmonic components in bulk crystals or in precisely controlled gas-phase systems [10–12].

Optical fibers of course provide a flexible platform for controlling chromatic dispersion via waveguide engineering. In what follows, we present a general treatment of cascaded FWM in fiber systems for a bichromatic pump, analyzing how the frequency dependence of the fiber group-velocity dispersion can yield phase-matched amplification of a high-order component on one side of the pump. The resulting spectral asymmetry is related to the intrinsic convective nature of fiber systems and the fact that higher-order dispersion breaks the system time-reversal symmetry [13,14]. This description provides important new insights into the origin of soliton Cherenkov radiation, a phenomenon that is fundamental to the generation of supercontinuum frequency combs [15–17]. We also clarify the energy flow mechanism in this process that has to date remained unspecified, in both the description of Cherenkov radiation and the associated soliton spectral recoil [18].

Our theoretical predictions are confirmed by numerical simulations and experiments considering both continuous wave and picosecond pulse excitation. The presented theory of cascaded phase matching constitutes the first physical description of soliton Cherenkov radiation in terms of the discrete wave mixing formalism of nonlinear optics, and establishes a direct link between multiple FWM and the dynamics of supercontinuum generation. A major consequence of our work is therefore that it provides a unified description of the two existing approaches used to describe broadband frequency conversion. We anticipate that this description will provide improved understanding to many nonlinear phenomena such as the emergence of self-organization [19], soliton frequency conversion [20,21], or symmetry breaking in modulation instability [14]. In addition to optics, the physics of cascaded phase matching will universally apply to the countless systems modeled by envelope equations exhibiting frequency-dependent group-velocity dispersion, such as the extended Lugiato-Lefever, Ginzburg-Landau or nonlinear Schrödinger equations (NLSE).

Consider two distinct pumps at frequencies ω_{-p} and ω_{+p} separated by Δ with a mean frequency ω_0 such that

$\omega_{\pm p} = \omega_0 \pm \Delta/2$. Cascaded FWM induced by these waves drives the sequential generation of a comb of discrete sidebands separated by Δ [22–24]. The frequency of the nth component ($n = 1, 2, 3, \ldots$) is $\omega_{\pm n} = \omega_{\pm p} \pm n\Delta$, where the subscripts + and − indicate frequency up- or down-shifting, respectively.

The sequential FWM cascade driving the polarization of the nth component at $\omega_{\pm n}$ mimics the direct $2n + 2$ photon process $\omega_{\pm n} = (n+1)\omega_{\pm p} - n\omega_{\mp p}$ induced by a $\chi^{(2n+1)}$ susceptibility [9]. As a consequence, the nth component of the cascade will undergo coherent amplification when the direct higher-order process is phase matched, i.e., when

$$\phi_{\pm n}(z) = (n+1)\phi_{\pm p}(z) - n\phi_{\mp p}(z), \tag{1}$$

where $\phi_{\pm p}$ and $\phi_{\pm n}$ denote the pump and signal phases, respectively. From Eq. (1) we can determine the corresponding phase matching condition in terms of the pump waves and fiber properties. Assuming that the pumps experience only self-phase- and mutual cross-phase-modulation, their phases evolve as $\phi_{\pm p}(z) = [\beta(\omega_{\pm p}) + \gamma P_{\pm p} + 2\gamma P_{\mp p}]z$, where $\beta(\omega)$ is the frequency-dependent propagation constant, γ the nonlinear coefficient, and $P_{\pm p}$ the pump powers. The nonlinear phase shift experienced by the sidebands arises dominantly from pump cross-phase-modulation such that $\phi_{\pm n}(z) = [\beta(\omega_{\pm n}) + 2\gamma P_{+p} + 2\gamma P_{-p}]z$. Combining these conditions with Eq. (1) yields a general phase matching condition for the amplification of a particular higher-order component at $\omega_{\pm n}$:

$$\begin{aligned}\beta(\omega_{\pm n}) = \pm n[\beta(\omega_{\pm p}) - \beta(\omega_{\mp p})] + \beta(\omega_{\pm p})\\ + n\gamma[P_{\mp p} - P_{\pm p}] - \gamma P_{\pm p}.\end{aligned} \tag{2}$$

Under typical experimental conditions, the nonlinear terms in Eq. (2) can be neglected, and phase matching at $\omega_{\pm n}$ is determined only by the linear mismatch $\Delta\beta_{\pm} = (n+1)\beta(\omega_{\pm p}) - n\beta(\omega_{\mp p}) - \beta(\omega_{\pm n})$. Expanding $\beta(\omega)$ about ω_0 to third order yields that $\Delta\beta_{\pm} = 0$ is fulfilled for frequency separations $\Delta_{\pm} > 0$:

$$\Delta_{\pm} = \mp \frac{3\beta_2}{\beta_3(n + 1/2)}, \tag{3}$$

where β_2 and β_3 denote, respectively, the second- and third-order dispersion coefficients at ω_0. For conventional fibers $\beta_3 > 0$, and the phase-matched frequency is given by

$$\omega_{\pm n} = \omega_0 \pm 3\frac{|\beta_2|}{\beta_3}. \tag{4}$$

At this stage we note that (i) phase-matched amplification requires both pump waves to experience the same sign of β_2, (ii) the phase-matched frequency $\omega_{\pm n}$ always lies in the opposite dispersion regime to that of the pumps, (iii) amplification at ω_{+n} (up-shifted from the pump) or ω_{-n} (down-shifted from the pump) corresponds to pumps lying in the anomalous or normal dispersion regime, respectively.

The theory above is confirmed by using an extended NLSE to simulate the propagation of a bichromatic field in 80 m of dispersion-shifted fiber (DSF) with zero-dispersion wavelength (ZDW) $\lambda_{\mathrm{ZDW}} = 1553.8$ nm, nonlinearity $\gamma = 2.5\ \mathrm{W}^{-1}\,\mathrm{km}^{-1}$ and $\beta_3 = 1.6 \times 10^{-4}\ \mathrm{ps}^3/\mathrm{m}$ at the ZDW. For completeness, fourth-order dispersion $\beta_4 = -7.0 \times 10^{-7}\ \mathrm{ps}^4/\mathrm{m}$ is also included but has negligible influence. These parameters correspond to our experiments below, and we remark that, because we expand the propagation constant at the ZDW, we have $\beta_2 = 0$. Of course, for an arbitrary frequency $\beta_2(\omega) = \beta_3(\omega - \omega_{\mathrm{ZDW}}) + \beta_4/2(\omega - \omega_{\mathrm{ZDW}})^2$, with $\omega_{\mathrm{ZDW}} = 2\pi c/\lambda_{\mathrm{ZDW}}$ the zero-dispersion frequency.

Figures 1(a) and 1(b) illustrate the cascaded phase-matched amplification of the ($n = \pm 2$) sideband associated with an equivalent $\chi^{(5)}$ nonlinearity for pumps in the anomalous [Fig. 1(a)] and normal dispersion regime [Fig. 1(b)]. In 1(a) the central frequency is $\omega_0/2\pi = 188.5$ THz ($\lambda_0 = 1590.5$ nm) and $\Delta/2\pi = 5.4$ THz ($\Delta\lambda \sim 46$ nm); in 1(b) $\omega_0/2\pi = 196.6$ THz ($\lambda_0 = 1524.9$ nm) and $\Delta/2\pi = 4.3$ THz ($\Delta\lambda \sim 33$ nm). We see how the spectrum becomes asymmetric due to the amplification of the phase-matched component on the opposite side of the ZDW to the pumps, with 15 dB enhancement relative to other generated components. This is at first sight counterintuitive because the strength of cascaded processes is expected to diminish with order, yet it is precisely this result that highlights the significance of cascaded phase matching in yielding amplification of a higher-order component.

To gain more insight into the dynamics, Figs. 1(c) and 1(d) plot the evolution of the sidebands ω_1 and ω_2 and pump waves as indicated for anomalous dispersion pumping. Figure 1(c) shows how the amplitude of the nonphase-matched ω_1 sideband remains small and oscillates at

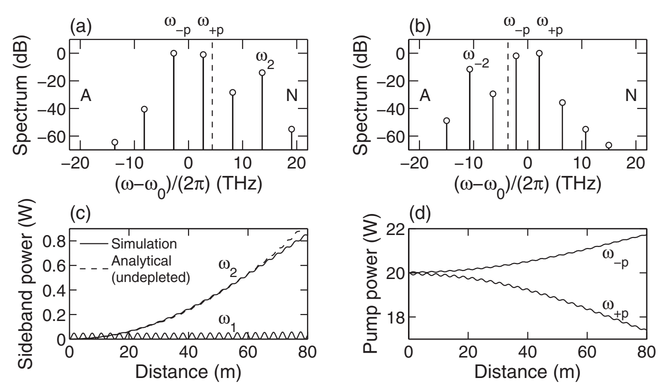

FIG. 1. Simulation results showing asymmetric output spectra for pumping in (a) anomalous and (b) normal dispersion regimes. (c), (d) Sideband and pump power evolution, respectively, for anomalous dispersion pumping. $P_{+p} = P_{-p} = 20$ W. Labels A and N indicate regions of anomalous and normal dispersion, respectively, and the dashed vertical line marks the zero-dispersion frequency.

the coherence length $L_{\rm coh} = 2\pi/\kappa_1 \sim 3.1$ m. Here, $\kappa_1 = 2\phi_{+p} - \phi_{-p} - \phi_1$ is the elementary mismatch associated with the direct generation of the $n = 1$ sideband. On the other hand, despite the nonzero elementary phase mismatch, $\kappa_2 = \phi_{+p} + \phi_1 - \phi_{-p} - \phi_2 \sim -2\ \mathrm{m}^{-1}$, the cascaded process at ω_2 is phase-matched according to Eq. (2) and thus we see sideband amplification. This process still exhibits low amplitude modulation at $L_{\rm coh}$ since the nonlinear polarization driving the phase-matched sideband is proportional to the instantaneous amplitude of the intermediate field at ω_1. We also find that the oscillatory behavior is reproduced in the small-signal evolution obtained from the analytical integration of the corresponding FWM coupled-mode equations in the undepleted pump approximation [dashed line in Fig. 1(c)], in excellent agreement with the numerical simulations.

Figure 1(d) shows how the cascade also leads to asymmetry in the powers of the two pump waves, amplifying (attenuating) the pump at ω_{-p} (ω_{+p}) spectrally further (closer) from the phase-matched sideband. This change in pump powers highlights the equivalence with the direct fifth-order process $\omega_2 = 3\omega_{+p} - 2\omega_{-p}$, where annihilation of three ω_{+p} photons must be associated with the creation of a sideband photon at ω_2 and two pump photons at ω_{-p}.

These results identify phase-matched cascaded FWM as the mechanism behind symmetry breaking in the NLSE with higher-order dispersion. We further illustrate the generality of the physics in Fig. 2 which shows results using the same fiber parameters as above but considering a denser frequency comb with reduced pump detuning Δ so that phase matching occurs at higher-order [see Eq. (3)]. The pump waves are in the anomalous dispersion regime with Eq. (2) satisfied for the $n = 15$ sideband, and indeed we see amplification at ω_{15}. We also observe here how the developed pump spectral asymmetry induces a temporal drift of the modulated field as shown in Fig. 2(b), which establishes an important link between the cascade-induced symmetry breaking and the convective nature of perturbed NLSE systems [13].

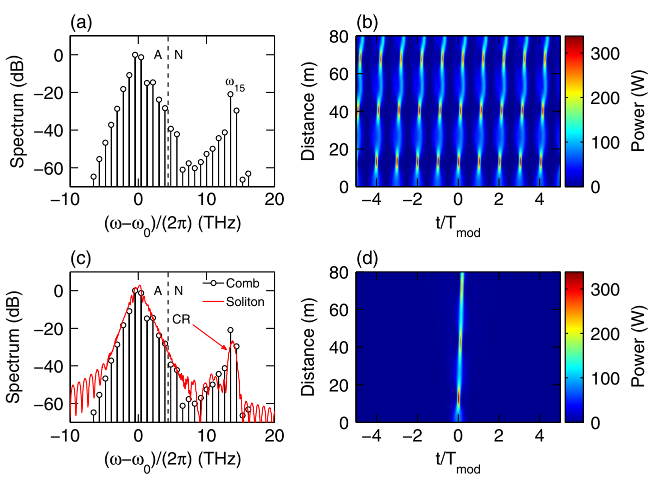

FIG. 2 (color online). Simulation showing amplification at ω_{15} in (a) frequency and (b) time domains. To establish the link with soliton-induced Cherenkov radiation, (c) compares the discrete comb spectrum with that of a soliton pulse corresponding to one cycle of the initial modulation. (d) Time-domain evolution. The mean pump frequency is $\omega_0/2\pi = 188.5$ THz and the pump detuning is $\Delta/2\pi = 0.87$ THz.

The results in Fig. 2(a) raise the connection with frequency conversion induced by pulses from mode-locked lasers where a spectral envelope modulates discrete modes at the cavity repetition frequency. In fact, we recall that Fig. 2(a) shows frequency conversion about the fiber ZDW with anomalous dispersion pumps, and we remark that the phase-matched frequency ω_{+n} from Eq. (4) is (neglecting the nonlinear contribution) exactly that of the Cherenkov field radiated by a soliton centered at the mean frequency ω_0 in the presence of third-order dispersion [15,16]. These results lead to the natural interpretation that Cherenkov radiation emitted by solitonlike pulses perturbed by higher-order dispersion arises from the cascaded FWM interaction between all the high and low spectral components of the soliton which—in pairs about the central frequency ω_0—contribute to pump the phase-matched process. [Note that the phase-matched frequency from Eq. (4) is independent of Δ, depending only on ω_0.]

To confirm this interpretation, Fig. 2(c) shows the propagation of a single pulse whose parameters match a single cycle of the modulated input field in Figs. 2(a) and 2(b). This pulse corresponds to a low-order soliton ($N = 1.87$) whose continuous spectrum after propagation closely follows the envelope of the discrete comb case. Most significantly, the position and amplitude of the Cherenkov wave shed by the soliton yield an excellent fit to the phase-matched sideband of the discrete frequency comb.

The interpretation of soliton Cherenkov radiation in terms of phase-matched cascaded FWM also provides a clear interpretation of soliton *spectral recoil* [18]. Specifically, we see how the pump wave asymmetry remarked upon in Fig. 1(d) will shift the center of mass of the continuous soliton spectrum in the direction opposite to the frequency of the generated radiation. To our knowledge, this is the first interpretation where a physical explanation of both the generation of Cherenkov radiation and soliton spectral recoil has been provided in terms of discrete wave mixing involving the individual spectral components of a propagating soliton pulse.

We have confirmed our theoretical and numerical results experimentally for cases of discrete two-pump and pulsed excitation. In the first experiments, we use two frequency-tunable external cavity lasers modulated with a 2.5% duty cycle to form 5 ns flattop quasi-continuous-wave (cw) fields which are amplified and injected into a 54 m long DSF with parameters as above. Figures 3(a) and 3(b) plot spectra at the DSF output for phase-matched amplification

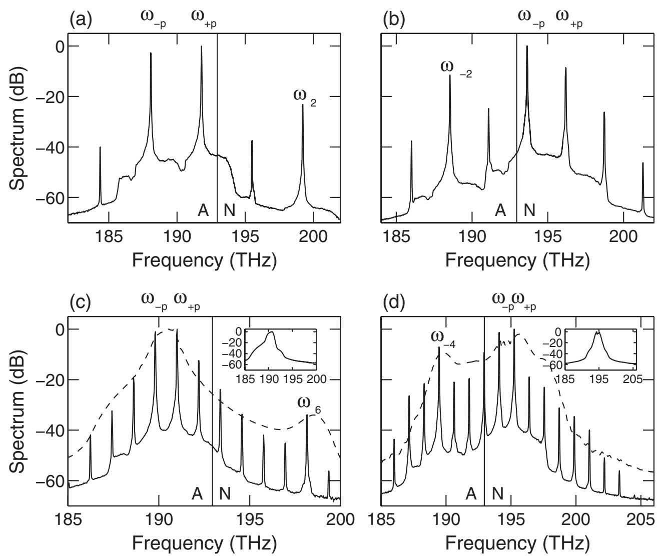

FIG. 3. Experimental results showing the phase-matched amplification of the (a) $n = 2$, (b) $n = -2$, (c) $n = 6$, and (d) $n = -4$ sideband. Solid vertical line indicates the zero-dispersion frequency, and the dashed curves in (c) and (d) show the output spectra for picosecond excitation. Insets in (c) and (d) show the input spectra of the picosecond source (axis labels are the same as in the main figure).

of the $n = 2$ and $n = -2$ sidebands which show enhancement of 15 dB compared to the intermediate first-order sidebands. Measurements demonstrating amplification of higher orders are shown for the $n = 6$ and $n = -4$ sidebands in Figs. 3(c) and 3(d), respectively. Parameters are given in Table I. Our experimental methodology was based on maximizing spectral asymmetry and the growth of a particular sideband order n by adjusting pump powers and external cavity laser frequency separation. In this way, we determine a particular experimental detuning Δ_e for a given n, and Table I also compares this experimental detuning with the expected value Δ_t obtained from Eq. (2). Agreement is excellent, with differences attributed to pump depletion and uncertainty in fiber dispersion.

Our interpretation of Cherenkov radiation in terms of cascaded FWM has been confirmed by a second series of experiments using a commercial (Alnair Labs, MLLD-100) 10 GHz repetition rate hybridly mode-locked laser diode. We first tuned the laser central frequency to 190.4 THz (1574.6 nm, anomalous dispersion regime) and show the results in Fig. 3(c). At this central frequency, a discrete bichromatic pump pair would yield phase-matched amplification of the $n = 6$ sideband (see Table I). The spectrum of the picosecond pulses after propagation is shown as the dashed line in Fig. 3(c), superimposed on the results for the discrete cw experiments. There is a small difference between the pulse duration used in the experiments (680 fs) and the duration of a single cycle of the cw modulation (420 fs), but, nonetheless, there is remarkable agreement between the position and form of the Cherenkov radiation emitted by the subpicosecond pulses and the spectral envelope of the discrete FWM comb in the vicinity of the phase-matched higher-order sideband. (Note that the individual modes of the laser pulses are not resolved in the experimental spectrum.) This is a clear confirmation of our physical interpretation of Cherenkov radiation emitted by solitons in terms of phase-matched cascaded FWM.

TABLE I. Experimental parameters and theoretical detunings.

Sideband order n	P_{+p} W	P_{-p} W	$\omega_0/2\pi$ THz	$\Delta_e/2\pi$ THz	$\Delta_t/2\pi$ THz
2	18	4.2	189.9	3.6	3.6
−2	2	41	194.9	2.6	2.9
6	20	9	190.4	1.2	1.1
−4	11	40	194.6	1.2	1.5

Our experiments also allow us to study spectral deformation of pulses injected in the normal dispersion regime, and results are shown in Fig. 3(d). Relative to the extensive studies of anomalous dispersion regime soliton dynamics, this area of research has been given relatively little attention. Our results, however, show that the same process of phase-matched cascaded energy transfer across the ZDW also occurs when pumping in the normal dispersion regime. Here, the 10 GHz laser was tuned to a central frequency of 194.6 THz (1540.6 nm, normal dispersion regime, pulse duration 920 fs) corresponding to a mean frequency where a discrete bichromatic pump pair yields phase-matched amplification of the $n = -4$ sidebands as shown in Fig. 3(d). The figure superimposes the pulse spectrum after propagation (dashed line) on the cw experimental case, and we see clear asymmetric deformation and a spectral peak in the anomalous dispersion regime in excellent agreement with the phase-matched frequency ω_{-4} corresponding to discrete bichromatic pumping. Again, we can see how cascaded FWM provides a physical interpretation for the unexplained appearance of spectral peaks for normal dispersion pumping near the fiber ZDW [25,26].

In conclusion, we have reported the first theoretical, numerical, and experimental study of cascaded phase matching in optical fibers. We have demonstrated that (i) cascaded FWM can mimic a directly phase-matched higher-order process that results in spectral asymmetry through the amplification of one particular comb sideband and (ii) that this process provides a physical interpretation of soliton Cherenkov radiation in terms of phase-matched mixing processes pumped by bichromatic pump pairs in the soliton spectrum. Describing nonlinear pulse propagation and soliton perturbations in terms of discrete wave mixing links for the first time the physics of supercontinuum with the multimode frequency conversion mechanism of cascaded FWM. This description is expected to provide insights into a plethora of optical systems, including Kerr combs generated in ring resonators. Finally, because symmetry breaking is an inherent characteristic of many nonlinear phenomena, we anticipate that the cascaded mechanism reported here will also be manifested in a wide range of other systems such as plasma physics, Bose-Einstein condensates, and hydrodynamics [27,28].

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