Multistep cascading and fourth-harmonic generation

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We apply the concept of multistep cascading to the problem of fourth-harmonic generation in a single quadratic crystal. We analyze a new model of parametric wave mixing and describe its stationary solutions for two- and three-color plane waves and spatial solitons. Some applications to the optical frequency division as well as the realization of the double-phase-matching processes in engineered QPM structures with phase reversal sequences are also discussed.

Cascading effects in optical materials with quadratic (second-order or \(\chi^{(2)}\)) nonlinear response provide an efficient way to lower the critical power of all-optical switching devices [4]. The concept of multistep cascading [3] brings new ideas into this field, leading to the possibility of an enhanced nonlinearity-induced phase shift and generation of multicolor parametric spatial solitons. In particular, multistep cascading can be achieved by two nearly phase matched second-order nonlinear processes, second-harmonic generation (SHG) and sum-frequency mixing (SFM), involving the third-harmonic wave [3, 4]. In this Letter, we extend the concept of multistep cascading to nonlinear effects of the fourth order and the fourth-harmonic generation (FHG) in a single noncentrosymmetric crystal. In particular, we analyze a new model of multistep cascading that involves the FHG process, and describe its stationary solutions for normal modes — plane waves and spatial solitons. Our study provides the first systematic analysis of the problem of FHG via a pure cascade process, observed experimentally more than 25 years ago [3] and later studied in a cascading limit only [4].

We consider the FHG via two second-order parametric processes: \(\omega + \omega = 2\omega\) and \(2\omega + 2\omega = 4\omega\), where \(\omega\) is the frequency of the fundamental wave. In the approximation of slowly varying envelopes with the assumption of zero absorption of all interacting waves, we obtain

\[
\frac{\partial A}{\partial z} = \frac{i}{2k_1} \frac{\partial^2 A}{\partial x^2} + i\gamma_1 A^* S e^{-i\Delta k_1 z},
\]

\[
\frac{\partial S}{\partial z} = \frac{i}{2k_2} \frac{\partial^2 S}{\partial x^2} + i\gamma_1 A^2 T e^{i\Delta k_1 z} + i\gamma_2 S^* T e^{-i\Delta k_2 z},
\]

\[
\frac{\partial T}{\partial z} = \frac{i}{2k_4} \frac{\partial^2 T}{\partial x^2} + i\gamma_2 S^2 T e^{i\Delta k_2 z},
\]

where \(A, S\), and \(T\) are the envelopes of the fundamental-frequency (FF), second- (SH) and fourth-harmonic (FH) waves respectively, \(\gamma_{1,2}\) are proportional to the elements of the second-order susceptibility tensor, and \(\Delta k_1 = 2k_1 - k_2\) and \(\Delta k_2 = 2k_2 - k_4\) are the corresponding wave-vector mismatch parameters. We introduce the normalized envelopes \((u, v, w)\) according to the following relations:

\[
A(x, z) = (16z_0^2\gamma_2)^{-1} u(x/a, z/2z_0) e^{-i\Delta k_1 z/2},
\]

\[
S(x, z) = (8z_0^2\gamma_2)^{-1} v(x/a, z/2z_0), \quad T(x, z) = (4z_0^2\gamma_2)^{-1} w(x/a, z/2z_0) e^{i\Delta k_2 z},
\]

where \(a\) is the characteristic beam width, and \(z_0 = k_0 a^2\) is the diffraction length of the FF component. In order to describe a family of nonlinear modes characterized by the propagation constant \(\lambda\), we look for solutions in the form \(u(x, z) \rightarrow \lambda U(x\sqrt{|I|}, z|I|) e^{i\lambda z/4}\), \(v(x, z) \rightarrow \lambda V(x\sqrt{|I|}, z|I|) e^{i\lambda z/2}\), and \(w(x, z) \rightarrow \lambda W(x\sqrt{|I|}, z|I|) e^{i\lambda z}\), and obtain the normalized equations:

\[
\begin{align*}
4i\lambda W &+ s \frac{\partial^2 W}{\partial z^2} - \alpha_1 W - \chi U^* V &= 0, \\
2i\lambda V &+ s \frac{\partial^2 V}{\partial z^2} - \alpha_2 W + \chi U^* V &= 0, \\
4i\lambda U &+ s \frac{\alpha_2 W - \alpha_1 V}{\partial z^2} - \chi V^2 &= 0,
\end{align*}
\]

where \(s = \text{sign}(\lambda) = \pm 1\), \(\chi = \gamma_1/(4\gamma_2)\) is a relative strength of two parametric processes, and the normalized mismatches are defined as \(\alpha = 4 + \beta/\lambda\) and \(\alpha_1 = 1/4 + \beta_1/\lambda\), where \(\beta = 8\Delta k_2 z_0\) and \(\beta_1 = -\Delta k_1 z_0\).

First, we analyze the plane-wave solutions of Eq. (1) which do not depend on \(z\). In this case, the total intensity \(I\) is conserved, and we present it in terms of the unscaled variables as \(I = I_u + I_v + I_w\), where \(I_u = |u|^2/4\), \(I_v = |v|^2/4\), and \(I_w = |w|^2/4\). Solutions \(\{U_0, V_0, W_0\}\) exist if \(\alpha > 0\). It has a fixed phase velocity \(\lambda = -\beta/4\) and an arbitrary amplitude, being unstable for \(I_w > \beta^2/4\) due to a parametric decay instability.

Two-mode solution \(\{0, \sqrt{2} \alpha, 1\}\) describes a parametric coupling between SH and FH waves, and it exists for \(\alpha > 0\), bifurcating at \(\alpha = 0\) from the FH mode. Coupling of this two-mode plane wave to a FF wave can lead to its decay instability, provided \(|I_1| < \alpha_1^{(cr)} = \chi\sqrt{2\alpha}\). To understand the physical meaning of this inequality, we note that the family of solutions characterized by the propagation constant \(\lambda\) corresponds to a straight line in the \((\alpha, \alpha_1)\)-parameter space, see Figs. 1(a,b). Moreover, all such lines include the point \((4, 1/4)\) as the
asymptotic limit for $|\lambda| \to +\infty$. This special point belongs to the instability region if the relative strength of the FF-SH interaction exceeds a critical value, i.e. for $\chi > \chi^{(cr)} = 1/(8\sqrt{2}) \approx 0.088$. However, for $\chi < \chi^{(cr)}$ this decay instability is suppressed for highly intense waves, owing to a strong coupling with the FH field.

Finally, a three-mode solution, $V_0 = \alpha_1/\chi$, $W_0 = V_0^2/(2\alpha_1)$, $U_0 = \sqrt{2V_0(1-W_0)/\chi}$, exists for (i) $\alpha > 0$ and $0 < \alpha_1 < \alpha_1^{(cr)}$, (ii) $\alpha > 0$ and $\alpha_1 < -\alpha_1^{(cr)}$, and (iii) $\alpha < 0$ and $\alpha_1 > 0$. In the limit $|\lambda| \to +\infty$, such three-wave modes are possible only for $\chi > \chi^{(cr)}$. In the region (i), stability properties of the three-wave solutions are determined by a simple criterion: the modes are stable if $\partial I/\partial |\lambda| > 0$, and unstable, otherwise. For the parameter regions (ii) and (iii), oscillatory instabilities are possible as well. Existence and stability of all types of stationary plane-wave solutions of the model (1) are summarized in Figs. (a-d).

In general, the system (1) is nonintegrable and its dynamics are irregular. However, we find that in some cases a quasi-periodic energy exchange between the harmonics is possible. Figure (a) shows one such case, when the intensities of unstable two-wave and stable three-wave stationary modes are close to each other, and an unstable two-wave mode periodically generates a FF component. Less regular dynamics are observed for other cases, such as for the generation of both SH and FH waves from an input FF wave [Fig. (b)]. This example also illustrates the possibility of effective energy transfer to higher harmonics close to the double phase matching point.
order of the mode guided by the two-component parametric soliton waveguide [2]. For a single-hump mode \((n = 0)\) the behavior of this cut-off is very similar to that of the plane waves. Indeed, in the cascading limit \((\alpha \gg 1)\), we have \(V_m \simeq 2 \sqrt{\alpha}\), and \(\alpha_{1}^{(cr)} \simeq 2 \chi \sqrt{\alpha}\), which differs by \(\sqrt{2}\) from the corresponding result for plane waves. The critical value of \(\chi\) for one-hump solitons can also be found from the approximate solution, \(\chi^{(cr)} \simeq 0.132\). We performed numerical simulations and found that the accuracy of our approximation is of the order of (and usually better than) \(1\%\) in a wide range of parameters \((\chi > 10^{-2} \text{ and } \alpha > 10^{-2})\), see Fig. 4(a).

![FIG. 3.](image)

(a) Regions of existence and stability of three-mode parametric solitons [shading is the same as in Fig. 2(a)]. Open diamonds — an analytical approximation, dark circle — exact phase-matching point. The dash-dotted line corresponds to the solutions at \(\beta = 2\) and \(\beta_1 = -0.15\), for which the power vs. \(\lambda\) dependences are shown in (b): thick — two-wave (SH + FH), and thin — three-wave solitons; solid and dashed lines mark stable and unstable solutions, respectively. Open circle is the bifurcation point. (c,d) Development of a decay instability of a two-wave soliton corresponding to \(\lambda = 1\) in (b), and generation of a three-component soliton: (c) FF, SH, and FH peak intensities vs. distance shown by dotted, solid, and dashed curves, respectively; (d) evolution of the FF component. For all the plots \(\chi = \chi^{(cr)}/2\).

Quite remarkably, for both positive \(\alpha\) and \(\alpha_1\) the stability properties of solitons [see Figs. 3(a,b)] and plane waves [see Figs. 2(a,c)] look similar. Specifically, stability of two- and three-component solitons is defined by the Vakhitov-Kolokolov criterion \(\partial P/\partial \lambda > 0\), where \(P = \int_{-\infty}^{\infty} I dx\) is the soliton power, except for the region \(\alpha_1 < \alpha_{1}^{(cr)}\) where two-component solutions exhibit parametric decay instability. An example of such an instability is presented in Figs. 3(c,d), where an unstable two-wave soliton generates a stable three-wave state. Such instability-induced dynamics are very different from that of plane waves where, instead, quasi-periodic energy exchange is observed [see Fig. 2(a)]. In the case of localized beams, diffraction leads to an effective power loss and convergence to a new (stable) state.

Similar to other models of multistep cascading [4], Eqs. (1) possess various types of exact analytical solutions, which can be found at \(\alpha = \alpha_1 = 1\) and \(\chi > 1/\sqrt{2}\): \(\alpha_1 = \alpha/4\) \((0 < \alpha < 1)\) and \(\chi = 1/(3\sqrt{2})\) or \(\chi = [(3\alpha)/(4 + 2\alpha)]^{1/2}\). Details will be presented elsewhere.

In order to observe experimentally the multistep cascading and multi-frequency parametric effects described above, we should satisfy the double-phase-matching conditions. Using the conventional quasi-phase-matching (QPM) technique [2] for FHG via a pure cascade process in LiTaO\(_3\), we find that there exists only one wavelength (2.45\(\mu m\)), for which two parametric processes can be phase-matched simultaneously by the different orders \(m\) of the QPM structure with the period \(\Lambda_Q = 34\mu m\). However, for the so-called phase-reversal QPM structures [10] characterized by two periods, the QPM period \(\Lambda_Q\) and the modulation period \(\Lambda_{ph} (\Lambda_{ph} > \Lambda_Q)\), double-phase matching is possible in a broad spectral range, provided the periods are selected to satisfy the conditions: \(\Lambda_Q = 4\pi|\Delta k_1 + \Delta k_2|^{-1}\), \(\Lambda_{ph} = 4\pi m|\Delta k_1 - \Delta k_2|^{-1}\), where \(m\) is the grating order. Thus, the engineered QPM structures suggested in [10] are more efficient than, e.g., the Fibonacci superlattices, and they can be used to achieve double-phase matching and to support different types of multistep cascading processes.

In conclusion, we have introduced a new model of multistep cascading that describes the fourth-harmonic generation via parametric wave mixing. We have analyzed the existence and stability of the stationary solutions of this model for normal modes — plane waves and spatial solitons. We have also discussed the possibility of double-phase-matching in engineered QPM structures with phase-reversal sequences.

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