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Few-cycle nonlinear mid-IR pulse generated with cascaded quadratic nonlinearities

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Generating few-cycle energetic and broadband mid-IR pulses is an urgent current challenge in nonlinear optics. Cascaded second-harmonic generation (SHG) gives access to an ultrafast and octave-spanning self-defocusing nonlinearity: when $\Delta k L \gg 2\pi$ the pump experiences a Kerr-like nonlinear index change $\Delta n = n_{\text{casc}} I$, where $n_{\text{casc}} \propto -d_{\text{eff}}^2 / \Delta k$, and d_{eff} is the effective quadratic nonlinearity. Due to competing material nonlinearities n_{Kerr} the total nonlinear refractive is $n_{\text{cubic}} = n_{\text{casc}} + n_{\text{Kerr}}$. Interestingly n_{cubic} can become negative (self-defocusing), elegantly avoiding self-focusing problems, and making it possible to excite solitons with normal dispersion [1].

Figure 1. (a) FOM $|n_{\text{casc}}|/n_{\text{Kerr}}$ for type 0 cascaded SHG using dielectric (red) and semiconductor (black) materials, with $n_{\text{casc}} = -2\omega_1 d_{\text{eff}}^2 / c^2 \epsilon_0 n^2(\omega_1) n(\omega_2) \Delta k$ and $n_{\text{Kerr}}(\omega) = K' G_2 (\hbar \omega / E_g) \sqrt{E_p / n^2(\omega) E_g^4}$, with the linear index $n(\omega)$, the band gap energy E_g , $K' = 7.3 \times 10^{-9} \text{ eV}^{3.5} \text{ m}^2 / \text{W}$ in dielectrics and $K' = 14.0 \times 10^{-9} \text{ eV}^{3.5} \text{ m}^2 / \text{W}$ in semiconductors, $E_p = 21 \text{ eV}$ is a constant. A FOM > 1 is required to excite solitons. (b) The DW phase-matching curves. (c) Numerical simulation of a 10-cycle $\lambda = 2.5 \mu\text{m}$ and $I_{\text{in}} = 50 \text{ GW/cm}^2$ pulse propagated 11 mm in a LiInS₂ crystal. 'A' and 'N' mark anomalous and normal dispersion.

Historically, *critical* (type I) cascaded SHG has been used. Recently we showed experimentally generation of strong and octave-spanning cascaded nonlinearities from a *noncritical* (type 0) interaction even without quasi-phase matching (QPM) [2]. This allows for excitation of few-cycle self-defocusing solitons at the pump wavelength, generation of octave-spanning supercontinua [2] and creation of long-wavelength Cherenkov radiation [3]. "Standard" type 0 mid-IR crystals have huge d_{eff} , but are often overlooked because of a large Δk value which cannot be reduced (as QPM methods are not developed or applicable). This limits the strength of n_{casc} so it is crucial to understand whether regimes with $n_{\text{cubic}} < 0$ can be found. Calculating the Kerr nonlinearity from the two-band model, Fig. 1(a) shows a figure-of-merit $\text{FOM} = |n_{\text{casc}}|/n_{\text{Kerr}}$; self-defocusing solitons require $\text{FOM} > 1$. LiNbO₃ and LiTaO₃ have an $\text{FOM} > 1$, but at around $\lambda = 2 \mu\text{m}$ the GVD changes sign and becomes anomalous (at this point the curves are terminated). Here the chalcogenide LiInS₂ and the semiconductors GaSe, CdSiP₂ and ZnGeP₂, which have large band gaps and large d_{eff} , come into play with an $\text{FOM} > 1$ for $\lambda > 2 \mu\text{m}$. Instead for e.g. CdGeAs₂, its large d_{eff} is counteracted by a very small band gap, giving a too large n_{Kerr} due to the E_g^{-4} scaling. None of the crystals with $\text{FOM} > 1$ support self-defocusing solitons beyond $\lambda = 5.5 \mu\text{m}$. However, once excited the soliton will shed radiation through optical Cherenkov radiation to a linear dispersive wave (DW) in the anomalous dispersion regime $\lambda > \lambda_{\text{ZD}}$. This can cover the long-wavelength range of the mid-IR. Fig. 1(b) shows the DW phase-matching curve $k_1(\omega) = k_1(\omega_{\text{sol}}) - (\omega - \omega_{\text{sol}}) / v_{g,\text{sol}}$. In Fig. 1(c) a numerical simulation of LiInS₂ shows that a 10-cycle input pulse at 2500 nm is soliton-compressed after 11 mm propagation to few-cycle duration. The soliton then generates a DW in the linear range (anomalous dispersion regime, $\lambda > 3.5 \mu\text{m}$), peaking at $\lambda = 8 \mu\text{m}$. This DW is very broadband and by isolating it with the equivalent of a long-pass filter we checked that it is a few-cycle pulse with excellent quality. The frequency conversion process is efficient, app. 10%. Much like what the near-IR for LN [2, 3], upon further propagation the soliton will fission into minor soliton pairs, each coupling to a dispersive wave in the anomalous dispersion regime. This will give a stronger peak around $\lambda = 8 \mu\text{m}$ but the coherence and quality drops. Eventually a very broadband supercontinuum will form across the spectrum.

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