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 $\Delta n = n_{\text{casc}}^\prime$, where $n_{\text{casc}}^\prime \propto -d_{\text{eff}}^{\text{casc}}/\Delta k$, and $d_{\text{eff}}$ is the effective quadratic nonlinearity. Due to competing material nonlinearities $\kappa_{\text{Kerr}}$ the total nonlinear refractive is $n_{\text{cubic}} = n_{\text{casc}} + \kappa_{\text{Kerr}}$. Interestingly $n_{\text{cubic}}$ can become negative (self-defocusing), elegantly avoiding self-focusing problems, and making it possible to excite solitons with normal dispersion [1].

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 $d_{\text{eff}}$, but are often overlooked because of a large $\Delta k$ value which cannot be reduced (as QPM methods are not developed or applicable). This limits the strength of $n_{\text{cubic}}$ 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 FOM $=|n_{\text{casc}}^\prime|/n_{\text{Kerr}}$; self-defocusing solitons require FOM > 1. LiNbO$_3$ and LiTaO$_3$ have an FOM > 1, but at around $\lambda = 2 \mu$m the GVD changes sign and becomes anomalous (at this point the curves are terminated). Here the chalcospinides LiInS$_2$ and the semiconductors GaSe, CdSiP$_2$, and ZnGeP$_2$, which have large band gaps and large $d_{\text{eff}}$, come into play with an FOM > 1 for $\lambda > 2 \mu$m. Instead for e.g. CdGeAs$_2$, its large $d_{\text{eff}}$ is counteracted by a very small band gap, giving a too large $n_{\text{Kerr}}$ due to the $E_g^{-4}$ scaling. None of the crystals with FOM > 1 support self-defocusing solitons beyond $\lambda = 5.5 \mu$m. However, once excited the soliton will shed radiation through optical Cherenkov radiation to a linear dispersive wave (DW) in the anomalous dispersion regime $\lambda > \lambda_{2D}$. 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 LiInS$_2$ shows that a 10-cycle input pulse at 2500 nm is soliton-compressed after 11 mm propagation to few-cycle pulses in quadratic media," J. Opt. Soc. Am. B 19, 2505–2510 (2002).

References