Mid-IR femtosecond frequency conversion by soliton-probe collision in phase-mismatched quadratic nonlinear crystals

XING LIU,1 BINBIN ZHOU,1 HAIRUN GUO,1,2 AND MORTEN BACHE1,*

1DTU FotoniX, Dept. of Photonics Engineering, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark
2Present address: Ecole Polytechnique Federale de Lausanne, CH-1015 Lausanne, Switzerland
*Corresponding author: moba@fotonik.dtu.dk

Received 25 May 2015; revised 29 June 2015; accepted 29 June 2015; posted 30 June 2015 (Doc. ID 241572); published 7 August 2015

We show numerically that ultrashort self-defocusing temporal solitons colliding with a weak pulsed probe in the near-IR can convert the probe to the mid-IR. A near-perfect conversion efficiency is possible for a high effective soliton order. The near-IR self-defocusing soliton can form in a quadratic nonlinear crystal (beta-barium borate) in the normal dispersion regime due to cascaded (phase-mismatched) second-harmonic generation, and the mid-IR converted wave is formed in the anomalous dispersion regime between $\lambda = 2.2-2.4 \, \mu m$ as a resonant dispersive wave. This process relies on nondegenerate four-wave mixing mediated by an effective negative cross-phase modulation term caused by cascaded soliton-probe sum-frequency generation. © 2015 Optical Society of America

OCIS codes: (190.7110) Ultrafast nonlinear optics; (190.5530) Pulse propagation and temporal solitons; (320.2250) Femtosecond phenomena; (190.4380) Nonlinear optics, four-wave mixing.

http://dx.doi.org/10.1364/OL.40.003798

The optical soliton is remarkably robust as it can survive dispersive or dissipative effects and even collisions. Yet it is quite susceptible to perturbations as it can shed phase-matched resonant radiation to a so-called soliton-induced optical Cherenkov wave (a.k.a. dispersive wave) when perturbed by higher order dispersion [1–6]. On the other hand, the soliton can also act as an impenetrable potential barrier when colliding with a linear (i.e., dispersive) wave, suggesting the analogy to an optical equivalent of an “event horizon” [7–9]. Such a collision has been explained using Doppler shifts [7] or by potential scattering theories [10,11]. A more direct explanation is to generalize the above-mentioned Cherenkov phase-matching condition from degenerate four-wave mixing (FWM) to nondegenerate FWM (a soliton interacting with two dispersive waves) [9,12–15]. The collision between the soliton ($\omega_s$) and a linear “probe” wave ($\omega_p$) can become resonantly phase-matched to a new frequency (the “resonant” wave $\omega_r$) according to the FWM phase-matching condition, mediated by cross-phase modulation (XPM). This frequency converts the probe to the resonant wave, which when completely depleting the probe gives rise to an apparent reflection of the probe on the soliton. However, this much debated “reflection” occurs only in the soliton reference frame and not in the stationary laboratory frame, so all waves are forward moving. Using “FWM language,” the probe is frequency-converted to the resonant wave. When the probe group velocity is higher than the soliton, the resonant wave group velocity will be lower than the soliton, so it will travel away from the soliton after formation.

Soliton-probe collisions have been studied in fibers and bulk media with a positive Kerr nonlinearity. However, through cascaded (strongly phase-mismatched) quadratic nonlinear interactions, an effective negative Kerr-like nonlinearity may be generated (in bulk this corresponds to a self-defocusing effect) [16]. When this happens, temporal soliton formation may occur in the normal group-velocity-dispersion (GVD) regime [17–20], and Cherenkov phase-matching naturally occurs in the anomalous GVD regime at longer wavelengths [21]. This was suggested for efficient near- to mid-IR femtosecond pulse conversion [22], and demonstrated experimentally [23,24].

Here we present numerical simulations of the collision of near-IR probes and self-defocusing temporal solitons to generate mid-IR resonant waves. The soliton self-defocusing nonlinearity is generated by cascaded second-harmonic generation (SHG). The probe is launched in the anomalous GVD regime, so the soliton acts as a scattering potential barrier only if the probe XPM nonlinearity is negative, and this turns out to require self-defocusing cascaded sum-frequency generation (SFG) between the soliton and probe. Thus, the whole process requires simultaneously cascaded self-defocusing pump SHG and cascaded self-defocusing pump-probe SFG. We show that colliding the probe with a higher order soliton, it can be completely converted to the mid-IR resonant wave, whose wavelength is widely tunable by varying the probe wavelength.

The cascaded quadratic ($\chi^{(2)};\chi^{(3)}$) nonlinear effects effectively act as cubic nonlinearities. On a microscopic level, up- and down-conversion steps occur repeatedly, as in the case SHG from $\omega + \omega \rightarrow 2\omega$ and back again $2\omega \rightarrow \omega + \omega$, due to the nonzero phase mismatch. On a macroscopic level, the pump pulse experiences this cascaded three-wave mixing as
an FWM Kerr nonlinearity, which may become self-defocusing. This competes directly with the material cubic nonlinearity; the effective nonlinearity is thus the sum of the cascading and the material Kerr nonlinear coefficients. Thus, the soliton-dispersive wave interaction can readily be described by the nonlinear Schrödinger equation with its cubic nonlinearity replaced by this effective nonlinearity, see more in [21], and then invoking the nondegenerate FWM phase-matching condition [13] \( k_{\text{in}}(\omega) = k_{\text{sol}}(\omega_0) + f[k_{\text{sol}}(\omega_p) - k_{\text{sol}}(\omega_p)] \). Here, \( k_{\text{in}}(\omega) \) is the dispersion relation of the linear wave (in bulk media simply determined, e.g., by the Sellmeier equation). \( k_{\text{sol}} = k_{\text{in}}(\omega_0) + (\omega - \omega_0)/v'_{\text{sol}} + q_{\text{sol}} \) is the soliton dispersion relation; due to its nondispersive nature, it is simply a wave packet with a constant group velocity \( v'_{\text{sol}} \) and its accumulated nonlinear phase \( q_{\text{sol}} \) will cancel out for the \( J = +1 \) case that we will focus on here. The parameter \( J \) switches between the degenerate case \( J = 0 \) (Cherenkov radiation) and the nondegenerate case \( J = \pm 1 \), due to the presence of the weak, i.e., linear, probe at frequency \( \omega_p \).

In a BBO quadratic nonlinear crystal (\( \beta \)-barium borate, \( \text{BaB}_2\text{O}_4 \)), the resonant waves are phase-matched in the mid-IR beyond \( \lambda = 2.0 \, \mu\text{m} \), as seen from the dispersion relations in Fig. 1 for the main case considered here, namely a 1.65 \( \mu\text{m} \) probe colliding with a 1.1 \( \mu\text{m} \) soliton.

The BBO crystal is cut for type-I SHG, where two \( \epsilon \)-polarized photons at the fundamental wave (FW) frequency \( \omega_1 \) generate a second-harmonic (SH) \( \epsilon \)-polarized photon at \( \omega_2 = 2\omega_1 \). The numerical simulations use the coupled electrical fields with minimal approximations [25]; see [26] for the BBO \( \chi^{(2)} \) and \( \chi^{(3)} \) tensor components and the absence of Raman effects. The model automatically includes any possible \( \chi^{(2)} \) interaction, such as SFG and difference frequency generation (DFG), and both inter- and intra-polarization (i.e., type 0, I, and II) interactions. As mentioned above, including the SFG process is crucial for the self-defocusing soliton to act as a potential barrier on the probe. Our plane-wave model neglects diffraction, assuming that the defocusing nonlinearities prevent self-focusing collapse of the beam (see more in [27]). Such simulations have shown good agreement with experiments using well-collimated beams and large spot sizes [17–21,23,24]. The BBO mid-IR material loss is initially neglected as to better focus on the collision physics.

Pumping with a strong \( \omega \)-wave at \( \omega_1 \), phase-mismatched SHG occurs to the \( \epsilon \)-wave at \( 2\omega_1 \). For a self-defocusing \( \epsilon \)-polarized temporal soliton to form at \( \omega_1 = \omega_0 \) we require [17,28]:

(a) the effective Kerr nonlinearity \( n_{\text{eff}}^{\text{SPM}}(\omega_0) = n_{\text{Kerr}}^{\text{SHG}}(\omega_0) + n_{\text{Kerr}}^{\text{SHG}} \) must be negative, and this is controlled by \( n_{\text{Kerr}}^{\text{SHG}}(\omega_0) \propto -\varepsilon_{\text{eff}}/\Delta k_{\text{in}}^{-1} \) by making the SHG phase mismatch \( \Delta k_{\text{in}} = k_{\text{in}}(2\omega_0, \theta) - 2k_{\text{in}}(\omega_1, \theta) \) small enough; (b) the GVD must be normal \( [k(2)(\omega_0) > 0] \), which in BBO means \( \lambda_0 < 1.488 \, \mu\text{m} \);

(c) the effective soliton order [27] \( N_{\text{eff}} \geq 1 \), where \( N_{\text{eff}} = L_D n_1 \varepsilon_0 n_1(\omega_1) c |I_{1,\text{in}}|^2/2 \) and \( L_D = T_0^2/|k(2)(\omega_1)| \).

The soliton-probe collision is modeled by launching a weak co-propagating \( \epsilon \)-polarized field: \( E_{\text{in}} = E_{\text{in}}(\cos(\omega_1 t) \ \text{sech}(t/T_1) + F_{\text{in}}(\cos(\omega_p t) \ \text{sech}(t - \tau)/T_p) \), where \( \tau \) is the delay time we take \( T_1 = T_p \). The soliton will efficiently scatter the probe if the soliton-induced XPM potential is a barrier. As the probe has anomalous GVD, this requires a negative probe XPM effective nonlinearity. This has the contributions [28]:

\[ n_{\text{eff}}^{\text{XPM}} = n_{\text{Kerr}}^{\text{SHG}} + n_{\text{casc}}^{\text{SFG}}(\omega_0 + \omega_p, \theta) + n_{\text{casc}}^{\text{DFG}}(\omega_0 - \omega_p, \theta), \] viz. the material Kerr XPM (which in BBO is identical to \( n_{\text{Kerr}}^{\text{SHG}} \), as both waves are \( \epsilon \)-polarized), as well as type I cascaded soliton-probe SFG and DFG. These cascading terms are similar in form to \( n_{\text{casc}}^{\text{SFG}} \) [28], and it turns out that \( n_{\text{casc}}^{\text{DFG}}(\omega_0 - \omega_p, \theta) \) is here negligible. Thus, the material Kerr XPM effect and the cascaded SFG effect are the main contributions to the XPM sign and magnitude. Only in certain regimes may \( n_{\text{eff}}^{\text{XPM}} < 0 \) ([28], Fig. 6), and the potential may even flip sign to become a well. We will investigate this phenomenon in another paper.

Figure 2 shows the results from a typical simulation. The BBO crystal angle is suitably chosen to give nonresonant [29] negative SPM and XPM nonlinearities [which occur between 17.5° < \( \theta < 20.0° \) ([28], Fig. 6)]. The soliton input intensity is chosen so \( N_{\text{eff}} = 2.0 \), allowing a higher order self-defocusing soliton to form. A weak probe is launched in the anomalous dispersion regime, which from Fig. 1(b) implies that its group velocity is larger than the soliton. It is therefore suitably delayed at the input so the interaction occurs over realistic crystal lengths. The time plot in (a) shows the probe colliding with the trailing edge of the strong soliton at around 10 mm. After the collision, a new wave emerges on the trailing edge; this is the resonant wave phase-matched to the soliton through the negative XPM nonlinearity. According to Fig. 1(b), the resonant wave will have a lower group velocity than the soliton, explaining why it is traveling away from the soliton trailing edge. Note that the soliton seems to split up into a double pulse after collision, but this is simply the uncompressed pedestal traveling faster than the soliton, which instead blue shifts and thus slows down (see below). In wavelength domain (b), the normalized spectral density (SD, calculated as \( S(\lambda) = |\Delta T/\lambda^2|^{1/2}/c \)) shows that between 10- and 20-mm propagation, the probe is almost completely converted to the resonant wave. It is exactly in this propagation range that the collision takes place in time domain. There is a good agreement between the predicted phase-matching frequency of the resonant wave, which is evident from the wavelength-domain phase-matching curves plotted in (c). We stress that the
observed mid-IR $\sigma$-polarized waves are not due to soliton-probe DFG; we chose a BBO cut so that no $\omega\omega\rightarrow\sigma$ DFG can occur due to its $d^{eff}$ being zero [28].

The soliton blue-shifts to 1.075 $\mu$m during propagation due to cascading-induced self-steepening [30]; while hardly evident in the plot, it was confirmed in the pulse spectrogram. This gives a new set of soliton dispersion curves [thin lines in (c)]. As these represent solitons, they are straight curves in frequency domain, albeit tilted instead of flat as the group velocity is different at 1.075 $\mu$m; in wavelength domain this means that they are curved due to the $\lambda \propto 1/\omega$ relation. These curves explain how the resonant wave is found slightly more red-shifted than the $\omega_p=\omega_1$ case predicted. We also see the Cherenkov ($J=0$) case in the spectrum, accurately predicted by the blue-shifted soliton phase-matching condition.

The spectral and temporal cuts in Fig. 3(a) shows the SD at input, during (z = 15 mm) and after collision (z = 30 mm). Since the soliton order is above unity, the $\sigma$-polarized spectra (thick lines) show that the soliton at collision is considerably extended toward the probe spectrum, and the final spectrum shows that the probe is almost completely depleted leaving only the $J = +1$ and $J = 0$ resonant waves. The $\sigma$-polarized third harmonic is also evident, and in the $\epsilon$-polarized spectra (thin lines), the various SHG and SFG components are evident as well. The same cuts are shown in time domain focusing in (b) on the probe (using a band-pass filter) and in (c) on the resonant wave (using a long-pass filter). The probe at 15 mm is around half-depleted, giving most of its depleted energy to the resonant wave that is located at the same temporal position. After 30 mm, the weak probe does not show on a linear scale (in the plot it is amplified 10 times). The resonant wave is now delayed 400 fs, and it is reduced in intensity and increased in time due to dispersion. It

has a Gaussian profile since it is a linear and not a soliton wave. Finally, (d) and (e) show the energy, normalized to the total input energy, of the soliton, probe, and resonant waves. The soliton initially looses around 2% of its energy through SHG to the $\epsilon$-polarized SH [(e) also shows the total $\epsilon$-polarized energy], causing the initial ripples at $z < 1$ mm. The soliton-probe interaction occurs between $z = 5–20$ mm, and the resonant wave builds up in energy. After 20 mm, the probe is depleted. The energy ratio (conversion efficiency) of the resonant wave to the probe is around 0.72, close to the limit posed by the photon-to-photon ratio $\omega_p/\omega_s = 0.73$ as dictated by the Manley–Rowe relation. The energies from a simulation where the probe never collides with the soliton (dashed lines) show as expected no energy at the resonant wave, and the probe remains unaffected.
Fig. 5. Long-wavelength part of the $\sigma$-polarized SD for various probe wavelengths shown after $z = 20$ mm (except $\lambda_p = 1.75$ mm where $z = 30$ mm) with (thick) and without (thin) IR losses of BBO. The dashed lines show the input states. The soliton was the same as in Fig. 2 and $I_p = 10$ GW/cm$^2$.

Figure 4 shows the probe and resonant wave energies versus $N_{\text{eff}}$ (controlled by $I_{1,\text{in}}$) for (a) $\lambda_p = 1.65$ mm fixed and four different $I_{p,\text{in}}$ values, and (b) $I_{p,\text{in}} = 10$ GW/cm$^2$ fixed and three different $\lambda_p$ values. In (a), complete probe depletion happens for intensities up to 30 GW/cm$^2$, all ending up at the same plateau, whose level is dictated by the Manley–Rowe relation (as $\omega_p$ is fixed). For $I_{p,\text{in}} = 40$ GW/cm$^2$, the probe conversion is incomplete, and we found that before collision, nonlinear spectral broadening had occurred. Thus the probe should remain weak (linear) in order to ensure complete depletion of the soliton. Since different probe frequencies are used, the plateau levels vary, in accordance with the Manley–Rowe relation. Clearly, probe depletion requires $N_{\text{eff}} > 1$. Generally, the resonant wave growth follows a logistic sigmoid function, whose slope scales as $I_{p,\text{in}}^{-0.5}$, and the midpoint $N_{\text{eff,0}}$ increases linearly with $I_{p,\text{in}}$ and decreases linearly with $\lambda_p$. This latter scaling comes from the fact that as $\lambda_p$ increases, it approaches the zero group-velocity mismatch (GVM) wavelength, where the probe and soliton have identical group velocities, see Fig. 1(b). The detuning from GVM has traditionally been kept small to get strong scattering, with the dilemma that the resonant wavelength is almost fixed and $\omega_p$ is tuned. Thus the probe-resonant wave conversion is possible when the probe wavelength is tunable by varying the probe wavelength. Obtaining phase matching further into the mid-IR is possible in other crystals, and interestingly, the system allows to change the XPM term sign to study the barrier versus well potential effect.

Funding. Technology and Production, Danish Council for Independent Research (11-106702).

REFERENCES