# Spectral shift of femtosecond pulses in nonlinear quadratic PPSLT Crystals

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**Abstract:** Spectral blue- and red-shifts in a range of 100 nm are achieved by propagating 40 fs pulses with a 70 nm spectrum centered at 1450 nm in a 25-mm-long periodically poled stoichiometric lithium tantalate crystal. We show experimentally that these shifts, originating from a phase-mismatched second harmonic generation process under conditions of strong group-velocity mismatch, can be efficiently controlled by acting on pulse intensity and phase-mismatch.



**OCIS codes:** (190.5940) Self-action effects; (190.5530) Pulse propagation and solitons; (190.4410) Nonlinear optics, parametric processes; (190.7110) Ultrafast nonlinear optics.

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## 1. Introduction

Self-induced effects occurring during propagation of ultra-short light pulses in nonlinear media can be exploited to perform a number of manipulations of their properties in the spatial, temporal and spectral domains [1]. In particular, self-induced spectral-shifts could be exploited for the realization of devices enabling to tune the carrier frequency of femtosecond pulses with continuity and over a broad range. These devices, that are potentially much simpler than optical parametric amplifiers [2], could enhance the performances of sources such as fibre lasers [3], which are very compact and efficient but with a fixed frequency.

In the last decades, many theoretical and experimental studies, performed on ultrashort-pulse propagation in cubic media (i.e., liquids, optical fibers, atomic vapors and semiconductors), showed that the cubic nonlinear self-steepening effect, due to propagation in a medium with an intensity-dependent index of refraction, causes asymmetric self-phase modulation and asymmetric frequency spectra of the propagating pulses [4-9]. In quadratic media, cascaded processes allow large equivalent cubic nonlinearities to be synthesized [10]. Effective self-phase modulation can be induced on a fundamental frequency (FF) wave by cascading interaction with the generated second harmonic (SH) wave under suitable phase-mismatch conditions. In the presence of strong group-velocity-mismatch (GVM) self-phase modulation is accompanied by an effective self-steepening effect, that can lead to shifts of the FF pulse spectrum of controllable sign and magnitude [11,12]. A spectral red-shift of 1 nm was first observed in PPLN waveguides by investigating spatial trapping of 4 ps pulses with a 1.5 nm spectrum [13,14], while spectral blue-shifts of 10 nm were achieved in BBO by propagating 120 fs pulses with a 10 nm spectrum centered at 790 nm [15]. In the latter case a deep theoretical investigation of the phenomenon was reported, ascribing spectral shifts to an effective quadratic Raman effect.

In this paper we demonstrate experimentally that, by acting on phase-mismatch and pulse intensity of 40 fs FF pulses, spectral blue- and red-shifts can be efficiently controlled in a range of 100 nm after propagation in a 25-mm-long periodically poled stoichiometric lithium tantalate crystal (PPSLT). The physical origin of spectral shifts is ascribed to an effective self-steepening effect and discussed with the help of a simple analytical model underlining the parameters that govern asymmetric self-phase modulation and spectral shift of the propagating pulses. This model has the advantage of highlighting in an intuitive way the influence of phase-mismatch, intensity, pulse duration and also temporal chirp on the frequency shifts.

## 2. Theoretical analysis

Let us consider a nonlinear type I interaction of a FF wave and a SH wave travelling in a medium with a large quadratic nonlinearity, in a quasi-phase-matched (QPM) geometry. By assuming first-order QPM, plane waves propagating in z direction and a time frame moving with the linear group velocity of the FF pulse, the equations that govern the interaction are [16]:

$$\begin{aligned} j\frac{\partial w}{\partial z} - \frac{\beta''_{FF}}{2}\frac{\partial^2 w}{\partial t^2} + \chi_{FF} v w^* e^{-j\Delta kz} &= 0 \\ j\frac{\partial v}{\partial z} - j\delta\frac{\partial v}{\partial t} - \frac{\beta''_{SH}}{2}\frac{\partial^2 v}{\partial t^2} + \chi_{SH} w^2 e^{j\Delta kz} &= 0 \end{aligned} \qquad (1)$$

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where w(z,t) and v(z,t) are the electric field envelopes (in V/m) at the FF and SH waves, respectively; β represents the propagation constant, β' the inverse group velocity, β'' the group velocity dispersion (GVD), $\delta = \beta'_{FF} - \beta'_{SH}$; $\Delta k = 2\beta_{FF} - \beta_{SH} + 2\pi/\Lambda$ is the effective wavevector-mismatch, where Λ is the QPM period; $\chi_{FF} = 2\chi^{(2)}/\lambda n_{FF}$ and $\chi_{SH} = 2\chi^{(2)}/\lambda n_{SH}$, where $\chi^{(2)}$ is the nonlinear coefficient, n is the refractive index, λ the FF wavelength. We consider the usual case where only the FF light is incident on the quadratic medium. In the limit of large phase-mismatch, an equation of motion for the FF field can be derived from (1) [11,12]:

$$j\frac{\partial w}{\partial z} - \frac{\beta''_{FF}}{2}\frac{\partial^2 w}{\partial t^2} + \frac{\chi_{FF}\chi_{SH}}{\Delta k}|w|^2 w - 2j\delta\frac{\chi_{FF}\chi_{SH}}{\Delta k^2}|w|^2\frac{\partial w}{\partial t} = 0 \qquad (2)$$

Equation (2) represents a generalized nonlinear Schrodinger equation [17]. An approximate analysis of Eq. (2) was performed in Ref. 15 in the absence of dispersion. Moreover, exact analytical solutions of Eq. (2) were found in Ref. [4-9], both in the presence and in the absence of dispersion. It was emphasized that the last term in Eq. (2) is responsible for self-steepening of the pulses, leading to temporal profile distortion and asymmetric frequency spectra. In the case of negligible GVD, the general solution $w(z,t) = \rho(z,t)\exp(j\phi(z,t))$ of Eq. (2) can be written as [8]:

$$\rho^2 = f(t + z\gamma\rho^2) \qquad (3a)$$

$$\phi = z\,\kappa\rho^2 + g(t + z\gamma\rho^2) \qquad (3b)$$

where $f(t)=\rho(t,0)^2$ is the initial pulse shape, g(t) is the initial phase distribution, $\gamma = 2\delta\chi_{FF}\chi_{SH}/\Delta k^2$ and $\kappa = \chi_{FF}\chi_{SH}/\Delta k$.

The nonlinearly induced asymmetry in pulse amplitude is manifested by the term proportional to z in Eq. (3a). As evident in Eq. (3a), for $\gamma<0$ (the usual case since the SH wave is slower than the FF one), both positive and negative mismatch conditions lead to a steepening of the trailing edge of the FF pulse, due to the dragging by the slower SH wave. This effect is directly proportional to the parameter δ because of increased dragging for larger GVM; it is inversely proportional to $\Delta k^2$, due the lower amount of SH and less efficient dragging for increasing phase-mismatch, and it increases with the FF pulse intensity. The group velocity reduction caused by self-steepening was recently demonstrated experimentally [18].

On the other hand, the nonlinearly induced asymmetry in frequency is manifested by the terms proportional to z in Eq. (3b). In the case of zero initial phase distribution (transform-limited input pulse), the variation of the instantaneous frequency is given by:

$$\delta\omega = -\frac{\partial\phi}{\partial t} = -z\kappa\frac{\partial\rho^2}{\partial t} \qquad (4)$$

In the case of negligible GVM ($\delta=0$), the pulse propagates undistorted and retains its symmetric shape. Therefore, Eq. (4) results in spectral modifications that are characteristic of self-phase modulation, with frequency shifts of opposite sign and equal magnitude on the leading and trailing edge of the pulse. On the other hand, in the presence of GVM, the asymmetric intensity profile of the pulse induced by self-steepening, unbalances the amount of frequency shift on the leading and trailing edges: in particular, for $\gamma<0$ the trailing edge of the pulse is steeper than the leading one and produces the strongest shift, towards the blue for $\Delta k<0$, and towards the red for $\Delta k>0$, the magnitude of such shift being larger for shorter input pulse-widths. It is worth noting that the relationship between group velocity modification and spectral shift of the FF pulse is quite different with respect to the linear case since the reduction of the FF group velocity occurring for $\gamma<0$ can be accompanied either by blue-shift [15,18] or by red-shift [13,14] depending on the sign of phase-mismatch. Finally Eq. (3b) shows that spectral shifts can be strongly affected by the initial pulse chirp.

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## 3. Experimental results

For the experimental observation and efficient control of the spectral phenomena we chose a PPSLT crystal because of its strong GVM, large nonlinearity and high optical damage threshold. The 25-mm-long (L) sample presents poling periods ranging from $\Lambda$ = 17.7 μm to $\Lambda$ = 21μm separated by non-poled regions. At the temperature of 160°C used in the experiments these periods correspond to phase matching wavelengths from 1440 nm to 1560 nm. The availability of different poling periods gave us the possibility of tuning the phase mismatch without changing the pulse spectrum. The near-IR pulses are derived from a non-collinear optical parametric amplifiers (NOPA) [2] pumped by the second harmonic of an amplified Ti:sapphire laser system (500 μJ, 150 fs, 1 kHz). The NOPA generates 70 nm nearly transform-limited pulses tunable in the near IR (1300-1500 nm) with widths around 40 fs (FWHM in intensity) and energy up to 1 μJ. The crystal length corresponds to 2.5 times the FF dispersion length, and to more than 200 times the GVM length between the FF and the SH waves arising from the nonlinear interaction. The pulses are focused in the middle of the PPSLT crystal with a spot size of 80 μm, giving a 60-mm confocal parameter, much longer than the crystal. This mild focusing enables to neglect both linear and nonlinear spatial effects. At the output of the sample the frequency spectra of the pulses are detected by a spectrometer.

Experiments were carried out by measuring the intensity dependence of the spectral profiles of the FF output pulse as a function of input pulse intensity for different phase-mismatch conditions. At low-intensity (0.1 GW/cm$^2$), independently of phase-mismatch, the FF output pulse spectrum is indistinguishable from the input one. At negative phase-mismatch values, corresponding to a self-defocusing cascaded nonlinearity, at high enough intensity, spectral blue-shifts of the injected pulses are observed. Typical experimental results obtained injecting FF pulses at 1450 nm with $\Delta kL = -80\pi$ are shown in Fig. 1(a). The blue-shift was observed to increase with the injected power, with a maximum value of about 40 nm at an intensity of 20 GW/cm$^2$. Under these conditions a spectral narrowing of ~20% also occurs. The experimental spectra can not be quantitatively fit using the above described analytical model due to the non-negligible role of dispersion, but they are nicely reproduced by full numerical simulations solving Eqs. (1), as shown in Fig 1 (b). The inset shows the evolution of the FF spectrum during propagation in the nonlinear crystal, demonstrating that the effect saturates after short propagation. A method for circumventing this limitation, based on engineered aperiodically poled crystals, was recently proposed [19].

In the time domain, at low intensity, the FF pulse is broadened due to GVD up to ~170 fs. At higher intensity pulse broadening is partially balanced by the nonlinear phase-shift due to the cascaded second-order interaction, resulting in a ~70 fs pulse-width at 20 GW/cm$^2$, and a

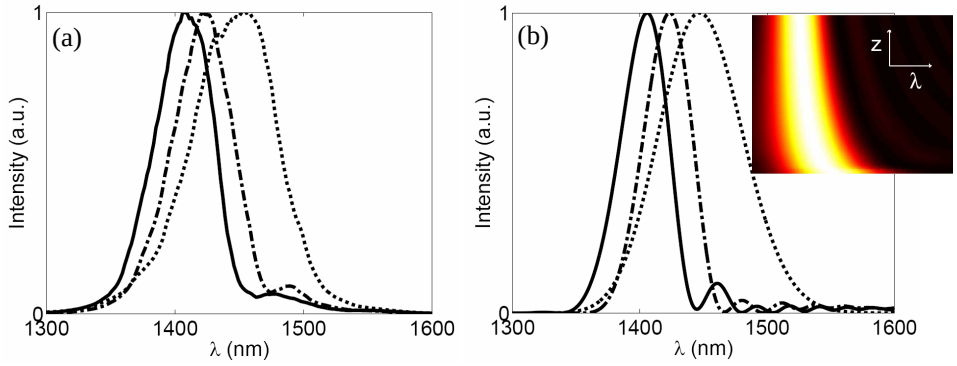


Fig. 1. Experimental (a) and numerical (b) FF spectra at the output of the crystal: I = 0.1 GW/cm$^2$ (dotted line); I = 7 GW/cm$^2$ (dash-dotted line); I = 20 GW/cm$^2$ (solid line). The phase mismatch is $\Delta kL = -80\pi$. The inset shows the FF spectral evolution in the λ-z plane at I = 20 GW/cm$^2$.

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group-delay shift induced by self-steepening takes place (see Ref. 18 for details). It is worth noting that in the non-poled region of the crystal, even at high intensity, no spectral modification was observed, thus indicating a negligible contribution by third order susceptibility. The spectral shift is thus a pure self-effect of quadratic interactions.

At positive phase-mismatch values, corresponding to a self-focusing cascaded nonlinearity, at high enough intensity, spectral red-shifts of the injected pulses are observed. Fig. 2 shows experimental (a) and numerical (b) results for FF pulses injected at 1490 nm with $\Delta kL = +80\pi$. For an intensity of 7 GW/cm$^2$ a pronounced red-shift of 40 nm is observed,

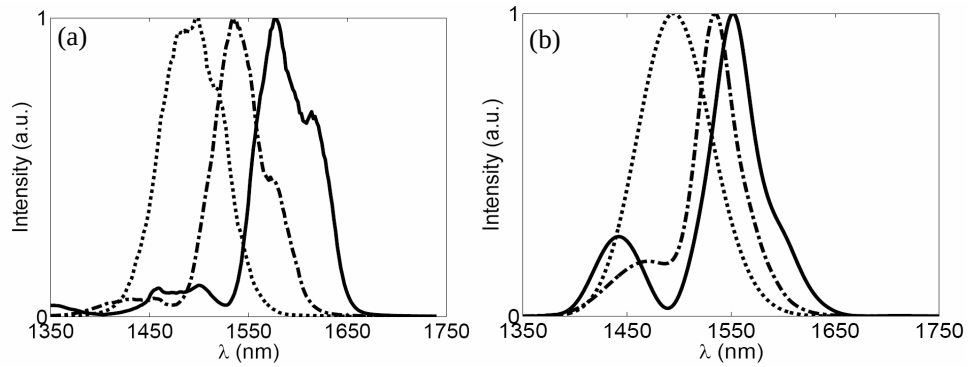


Fig. 2. Experimental (a) and numerical (b) FF spectra at the output of the crystal: I = 0.1GW/cm$^2$ (dotted line); I = 7 GW/cm$^2$ (dash-dotted line); I = 20 GW/cm$^2$ (solid line). The phase mismatch is $\Delta kL = 80\pi$.

which is well reproduced by numerical simulations. At higher intensity (20 GW/cm$^2$) an even stronger red-shift occurs (~90 nm), which can not be quantitatively reproduced by the numerical model due to the onset of self-focusing (spatial break-up instabilities) leading to an increase of the local intensity. Moreover, in the time domain the combined effect of positive dispersion and self-focusing cascaded nonlinearity gives rise to pulse broadening and also to pulse break-up at high intensities. These effects need to be considered in applications requiring good temporal quality of femtosecond pulses.

In Fig. 3 we summarize the observed dependence of the FF spectral shifts on the input intensity, together with the results of the numerical simulations, for different phase-mismatch values ($\Delta kL = -80\pi$, $\Delta kL = -400\pi$ and $\Delta kL = 80\pi$). The frequency shift of the FF wave

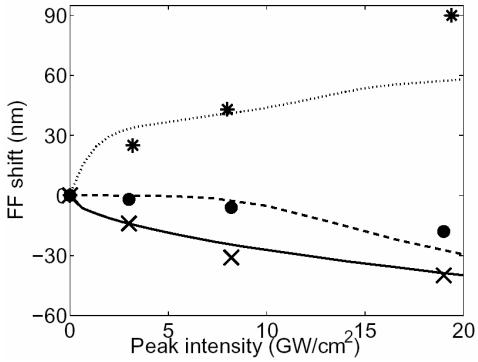


Fig. 3. Experimental (symbols) and calculated (lines) FF frequency shifts as a function of injected input intensity for different phase-mismatch conditions: $\Delta kL = -80\pi$ (crosses and solid line), $\Delta kL = -400\pi$ (circles and dashed line), $\Delta kL = 80\pi$ (stars and dotted line).

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increases with intensity for a given phase-mismatch value, and it decreases upon increasing the phase-mismatch for a fixed intensity value, in good analogy with the behavior discussed in. par. 2. It is worth pointing out that the theoretical model there reported, even if not suitable for a quantitative prediction of frequency shifts due to the strong dispersion, can however be of help in comparing the spectral shifts here reported with those obtained in previous works on this subject. According to Eq. (4) the magnitude of spectral shifts, being proportional to the time derivative of the intensity, increases with decreasing pulse duration for a given peak intensity and nonlinearity: thus, 40 fs pulses produce approximately 40 nm shifts, 120 fs input pulses result in ~ 10 nm shifts [15] while 4 ps pulses in about ~ 1 nm [13,14].

## 4. Conclusions

In this work we demonstrate experimentally that spectral shifts induced by quadratic processes in a regime of strong phase- and group-velocity-mismatch can be efficiently controlled in sign and magnitude through pulse intensity and phase-mismatch. Spectral red- and blue-shifts of more than 40 nm have been obtained by propagating 40 fs pulses in a 25 mm long PPSLT crystal. The role and the weight of the parameters affecting such shifts is highlighted with the help of a simple analytical model relating the observed nonlinear dynamics to the presence of effective quadratic self-phase modulation and self-steepening. These phenomenona can be exploited for the realization of a simple frequency shifter of femtosecond pulses emitted by non tunable laser sources.

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