

High-energy pulse compression by use of negative phase shifts produced by the cascade $\chi^{(2)}:\chi^{(2)}$ nonlinearity

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We report a simple optical pulse-compression technique based on quadratic nonlinear media. Negative nonlinear phase shifts are generated by phase-mismatched second-harmonic generation, and the phase-modulated pulses are then compressed by propagation through materials with normal dispersion. Millijoule-energy pulses from a Ti:sapphire amplifier are compressed from 120 to 30 fs, and calculations indicate that compression ratios of >10 are realistically achievable by use of this approach with optimal materials. The insertion loss of the compressor can be less than 10% of the pulse energy, and scaling to higher pulse energies will be straightforward. © 1999 Optical Society of America

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Pulse compression is an established technique for generating optical pulses shorter than those produced by lasers or amplifiers. Most commonly, additional bandwidth is generated by self-phase modulation in an optical fiber. The group-velocity dispersion (GVD) required for compression of the pulse is typically provided by gratings or prisms. Compressors based on single-mode fibers are limited to nanojoule pulse energies by higher-order nonlinear effects and ultimately by damage to the fiber. Thus, new approaches are needed for compression of the high-energy pulses that are now available from chirped-pulse amplifiers,¹ for example.

Bulk materials can be used for pulse compression based on third-order nonlinearity.² Continuum generation arising from self-focusing can severely distort the beam and impede effective compression. These effects limit the utility of compression in bulk third-order media.

One solution to this problem was reported by Nisoli *et al.*³ They achieved large spectral broadening by propagating pulses through a high-pressure noble gas confined in a hollow-core waveguide of fused silica. Excellent results were obtained, including compression from 140 to 10 fs. Pulse energies as high as 240 μJ were produced with 660- μJ input pulses. Although the compressed pulse energy is a substantial improvement on that which is achievable with ordinary fibers, these results do point out a limitation of this approach: Because the pulse does not propagate as a guided mode, the waveguide is lossy. Additional drawbacks include the susceptibility of the waveguide to optical damage, the complexity associated with handling the high-pressure gas, and a lack of commercially available components.

Recent research has shown that second-order nonlinearities can be exploited for pulse compression. Following research by Wang and Luther-Davies,⁴ Dubietis *et al.* demonstrated that pulses can be compressed in phase-matched type II second-harmonic generation.⁵ This approach relies on group-velocity mismatch (GVM) among the three interacting waves and requires division of the input pulse into *o* and *e* waves as well as on an appropriate predelay of one of the input pulses. Compression from 1.3 ps to 280 fs

was demonstrated, with energy efficiency of close to 50%. Dubietis and co-workers have also demonstrated the phase-matched generation of second-harmonic pulses shorter than the input fundamental pulse through pulse tilting.⁶

Here we show that negative phase shifts produced in phase-mismatched type I second-harmonic generation can be exploited for effective pulse compression. Our research is conceptually similar to that employed in traditional compressors: In a first stage the pulse accumulates a nonlinear phase shift, and the pulse is then compressed by dispersive propagation in a second stage [Fig. 1(a), inset]. The use of a self-defocusing nonlinearity reduces or eliminates the problems that can arise from Kerr self-focusing. Positive GVD is needed for compression, and this can be provided by a suitably chosen piece of transparent material. 120-fs pulses are compressed by a factor of ~ 4 , and higher compression ratios should be possible. The compressor is efficient, with the compressed pulse amounting to at least 85% of the input-pulse energy.

It is well known that the cascading of $\chi^{(2)}(\omega; 2\omega, -\omega)$ and $\chi^{(2)}(2\omega; \omega, \omega)$ processes leads to a nonlinear phase shift $\Delta\Phi^{\text{NL}}$ in a pulse that traverses a quadratic medium under phase-mismatched conditions for second-harmonic generation (SHG). The phase shifts can be either positive or negative, depending on the sign of the phase mismatch ΔkL ($\Delta k = k_{2\omega} - 2k_{\omega}$). Bakker and co-workers performed a theoretical study of the phase shifts generated by three-wave interactions,⁷ and cascade phase shifts were later measured

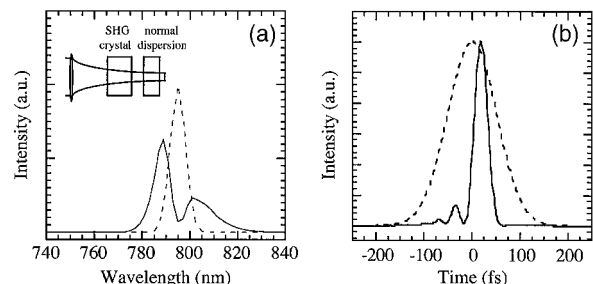


Fig. 1. (a) Calculated spectrum and (b) intensity profile of the compressed pulse. Dashed curves, the input pulse.

in KTP (Ref. 8) and periodically poled LiNbO₃.⁹ We recently employed cascade nonlinearities for Kerr-lens mode locking with $\Delta\Phi^{\text{NL}} < 0$ (Ref. 10) as well as for the production of pulses that are simultaneously solitons in time and space.¹¹

We take the simplest approximate approach to obtain guidelines for compressor design and performance and then refine these through numerical solutions of the appropriate wave equations; a systematic analysis of the compressor will be presented elsewhere. In general the maximum compression ratio will be proportional to $\Delta\Phi^{\text{NL}}$ impressed upon the initial pulse. The phase shift produced by the cascade process can be approximated as⁸

$$\Delta\Phi^{\text{NL}} \approx -\frac{\Gamma^2 L^2}{\Delta k L},$$

with $\Gamma \approx \omega d_{\text{eff}} |E_0| / c \sqrt{n_{2\omega} n_\omega}$. The cascade process produces a nonlinear frequency chirp in the presence of GVM. However, with large enough phase mismatch, the cycles of conversion and backconversion that generate $\Delta\Phi^{\text{NL}}$ occur before the pulses move away from each other because of GVM. Then $\Delta\Phi^{\text{NL}}$ follows the pulse intensity, so the frequency chirp is linear near the pulse peak. (Calculations that illustrate this issue are presented in Ref. 10.) To quantify this point, we introduce the characteristic group-mismatch length $L_{\text{GVM}} = c\tau_0 / (n_{1g} - n_{2g})$, where n_{1g} (n_{2g}) is the group index of the fundamental (harmonic) frequency and τ_0 is the initial pulse duration. For a crystal of length $L = NL_{\text{GVM}}$ we desire at least $2N$ conversion-backconversion cycles, which we arranged by setting $\Delta k L = 4N\pi$. Crystals with large values of d_{eff} and L_{GVM} are naturally best for this application. For the compression of 120-fs pulse at 800 nm we chose to use barium metaborate (BBO), which has $d_{\text{eff}} \approx 2$ pm/V and $L_{\text{GVM}} = 0.6$ mm. With a 17-mm BBO crystal (provided by Casix, Inc.), this implies that $\Delta k L \approx 120\pi$, and we chose $\Delta k L = 200\pi$. With intensities of ~ 50 GW/cm², $\Delta k L \approx 200\pi$ yields a net $\Delta\Phi^{\text{NL}} \approx -\pi$, which is necessary for a compression ratio of roughly 4. Materials with small Kerr nonlinearities are desired for the generation of net negative phase shifts. The nonlinear index of BBO is small, $n_2 \approx 5 \times 10^{-16}$ cm²/W. A final advantage of BBO is its lack of two-photon absorption at 400 nm.

In the reference frame of the fundamental pulse, the equations that govern the interaction between fundamental and harmonic fields E_1 and E_2 propagating in the z direction, neglecting GVD, are

$$\begin{aligned} \frac{\partial}{\partial z} E_1 &= iE_1^* E_2 \exp(i\Delta k z) + i2\pi(n_2 I_0) \\ &\times \frac{L_{\text{NL}}}{\lambda} (|E_1|^2 + 2|E_2|^2) E_1, \end{aligned}$$

$$\begin{aligned} \left(\frac{\partial}{\partial z} + \frac{L_{\text{NL}}}{L_{\text{GVM}}} \frac{\partial}{\partial t} \right) E_2 &= iE_1 E_1 \exp(-i\Delta k z) + i4\pi(n_2 I_0) \\ &\times \frac{L_{\text{NL}}}{\lambda} (2|E_1|^2 + |E_2|^2) E_2, \end{aligned}$$

where E_1 and E_2 are in units of the initial value of the peak fundamental field E_0 . The length that characterizes the nonlinear interaction is $L_{\text{NL}} = n\lambda / [\pi\chi^{(2)} E_0]$. Time is measured in units of the input-pulse duration τ_0 , and position is measured in units of L_{NL} . We solved the wave equations numerically, and the results are compared below with experimental results.

Input pulses of duration 120 fs and energy 600 μ J at 795 nm were produced by a Ti:sapphire regenerative amplifier. The 7-mm-diameter beam from the amplifier is compressed by a factor of 2 with a telescope to produce intensities of 20–60 GW/cm² on the BBO crystal. In the second stage of the compressor, a prism pair or a piece of bulk material provides the GVD to compensate for the phase shift accumulated in the quadratic medium.

With $\Delta k L = 200\pi$, the spectrum is expected to broaden by a factor of ~ 3 and to develop a multipy-peaked structure [Fig. 1(a)]. The GVD required for optimum pulse compression is ~ 1500 fs², and this should produce an ~ 30 -fs compressed pulse [Fig. 1(b)]. Since we neglect GVD in the SHG crystal, the GVD that must be supplied is smaller than the calculated value. However, the compressed pulse duration is not extremely sensitive to the GVD, so we provided the approximate calculated value. The measured spectrum [Fig. 2(a)] and pulse autocorrelation [Fig. 2(b)] produced with a prism pair controlling GVD agree reasonably with calculations. The zero-phase Fourier transform of the experimental spectrum [Fig. 2(c)] produces an autocorrelation [shown in Fig. 2(b)] that

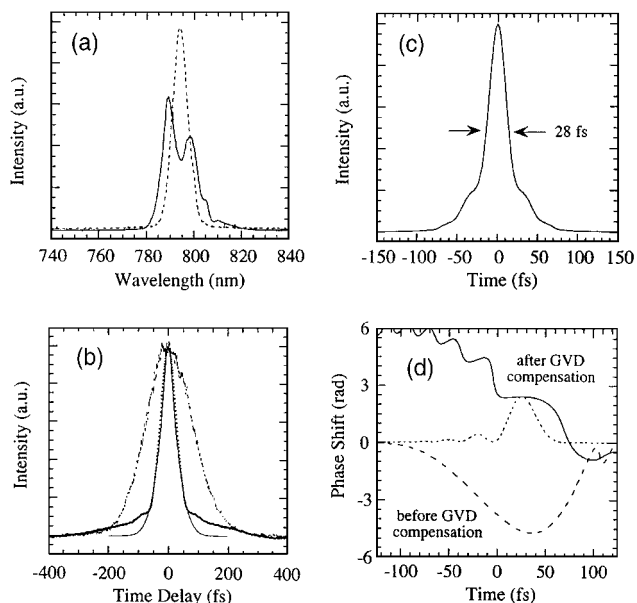


Fig. 2. (a) Measured spectrum and (b) autocorrelation (symbols) of the compressed pulse. The autocorrelation of the transform-limited pulse in (c) derived from the measured spectrum is also shown (solid curve) in (b) for comparison. (a), (b) Dashed curves, the input pulse. The temporal phases of the pulse before (dashed curve) and after (solid curve) the dispersive propagation stage are shown in (d), with the compressed pulse intensity shown (dotted curve) for reference. Note that the pulse duration is ~ 140 fs before GVD compensation.

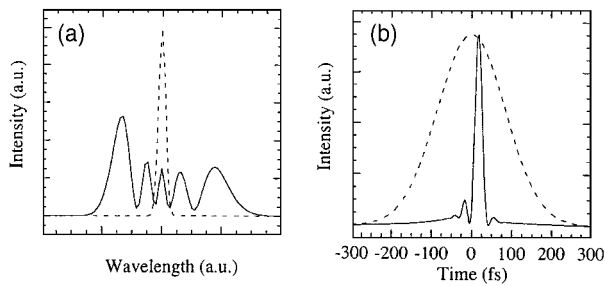


Fig. 3. (a) Calculated spectra and (b) intensity profiles for compression of a 200-fs pulse at $1.55 \mu\text{m}$. A 2-cm periodically poled LiNbO₃ crystal is assumed, with incident intensity $20 \text{ GW}/\text{cm}^2$ and $\Delta kL = +800\pi$. Dashed curves, the input pulse.

agrees with experiment near zero delay but deviates from the pedestal measured at >75 fs from the peak. The calculated phase variation [Fig. 2(d)] indeed implies good compression of the majority of the pulse but some residual phase variation in the wings. Effective compression was realized with ΔkL from 150π to 300π . Smaller values of ΔkL in this range produce the narrowest pulses but increase the energy in the wings, whereas larger values produce broader pulses of higher quality. SHG at such a large phase mismatch introduces a loss of 5% or less. The remainder of the loss could be eliminated by antireflection coatings on the BBO crystal.

The use of negative phase shifts permits the construction of a compressor consisting of only the SHG crystal and a piece of bulk material. It is important to avoid excessive nonlinear phase shift and two-photon absorption, so materials with small third-order nonlinearities (and large GVD) are naturally best for the dispersive stage. A 2-cm piece of calcite was available and presents $\sim 1500 \text{ fs}^2$ of dispersion. Although the dispersion was not optimal, ~ 30 -fs pulses were generated when the calcite replaced the prism pair. Similar compression ratios have been obtained with pieces of sapphire and LiIO₃ used to provide dispersion. Compressed pulse energies of at least $520 \mu\text{J}$ (85% of the input energy) were obtained with all three bulk materials.

The compressor based on negative nonlinear phase shifts offers substantial practical advantages. It is extremely simple, and there are no critical parameters or adjustments. The pulse duration is not highly sensitive to the pulse energy, perhaps owing to the saturable nature of the cascade nonlinearity. Scaling the compressor to energies of at least 10 mJ is simply a matter of increasing the aperture of the SHG crystal to maintain intensities of $\sim 50 \text{ GW}/\text{cm}^2$. With 1-mJ and 100-fs pulses the power that is incident upon the SHG crystal is already several orders of magnitude above the critical power for self-focusing by means of the Kerr nonlinearity, but the Kerr phase shift is compensated for by the negative cascade phase shift. In this connection it is worth mentioning that we investigated pulse compression with $\Delta\Phi^{\text{NL}} > 0$ (obtained with $\Delta kL < 0$). Severe beam distortions and spatially inhomogeneous continuum light accompanied

the desired spectral broadening and precluded effective compression. Compression with positive phase shifts may be effective with longer or less-energetic pulses. The pulses compressed with $\Delta\Phi^{\text{NL}} < 0$ exhibit none of the spatial distortions observed with $\Delta\Phi^{\text{NL}} > 0$. Given the complications involved in designing chirped-pulse amplifiers that are capable of producing sub-50-fs pulses with millijoule energies,¹² a 100-fs amplifier plus a quadratic compressor seems to offer a useful alternative approach.

Inasmuch as the cascade nonlinearity saturates, increasing the pulse energy does not increase the compression ratio as effectively as does the use of longer and (or) more nonlinear crystals. For example, calculations show that 200-fs pulses at $1.55 \mu\text{m}$ may be compressed by a factor of 10 by use of a 2-cm piece of periodically poled LiNbO₃ (Fig. 3). Owing to the limitations of GVM, compression by a factor of >10 appears to be difficult.

In conclusion, we have demonstrated that pulse compression with negative phase shifts produced by quadratic nonlinearities is effective and offers some advantages over existing techniques. The simplicity of this compressor should be attractive for many applications.

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Note added in proof. By minimizing spatial and temporal chirp on the input beam, we observe auto-correlations without excess energy in the wings of the pulse.

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