Femtosecond Kerr-lens mode locking with negative nonlinear phase shifts

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We report a Kerr-lens mode-locked Cr:forsterite laser operated with negative nonlinear phase shift. The nonlinear phase shift is induced by the cascade $\chi^{(2)}$:$\chi^{(2)}$ process in a lithium triborate crystal. Employing the cascade process at large phase mismatch produces a nearly linear frequency chirp. Transform-limited pulses as short as 60 fs are generated with positive cavity dispersion. © 1999 Optical Society of America

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The field of ultrafast science has been revolutionized by the development of Kerr-lens mode locking[1,2] (KLM) in solid-state lasers. KLM is based on the nonresonant Kerr nonlinearity, and for almost all materials the nonlinear index n_2 (and thus the nonlinear phase shift $\Delta\Phi^{\mathrm{NL}}$) is positive. In solitonlike pulse shaping, the frequency chirp induced by the positive nonlinearity is balanced by negative group-velocity dispersion (GVD), and prism pairs and chirped mirrors were developed to produce this balance. The signs of the nonlinearity and dispersion can be interchanged (Fig. 1), so if media with fast negative nonlinear refraction (i.e., self-defocusing) could be found, lasers could be operated with positive cavity dispersion. In either case some amplitude modulation is needed to stabilize the pulse formation. Operation at positive dispersion could simplify the design of compact and robust femtosecond lasers for applications.

Recently there has been a resurgence of interest in the effective third-order nonlinearity that arises from the cascading of $\chi^{(2)}$ processes. The renewed interest is based on the recognition that large effective third-order nonlinearities of controllable sign can be produced in the cascade process. Bakker and co-workers performed a systematic theoretical study of the phase shifts generated by three-wave interactions.[3] Large cascade nonlinear phase shifts were later measured in potassium titanyl phosphate.[4] Applications of the cascade nonlinearity include the mode locking of lasers, and this has been proposed and demonstrated.[5] Pulses of ~10-ps duration were generated; in this case the signs of $\Delta\Phi^{\mathrm{NL}}$ and cavity GVD have little influence on pulse shaping and laser performance.

Difficulties that must be addressed for the use of the cascade nonlinearity on a femtosecond time scale have been noted by previous workers. $\Delta\Phi^{\mathrm{NL}}$ is proportional to conversion efficiency η, and in general it is difficult to attain high efficiency in second-harmonic generation (SHG) with femtosecond pulses if the harmonic pulse is constrained to be no longer than the fundamental. Group-velocity mismatch (GVM) between the fundamental and the harmonic pulses and spatial walk-off will limit both conversion and backconversion efficiencies. These in turn limit the attainable nonlinear phase shift, and any residual second-harmonic light is a power-dependent loss that will destabilize KLM. A second complication is the fact that the phase shift produced by the cascade process can be modulated significantly owing to GVM.[3,5,6] These issues have led some workers[5,7] to conclude that it will be difficult to exploit the cascade nonlinearity with femtosecond-duration pulses.

Here we describe what is to our knowledge the first KLM laser operating with negative nonlinear phase shift. A lithium triborate (LBO) crystal inside a Cr:forsterite laser provides the effective nonlinearity by means of the cascade process. We believe that this is also the first application of the cascade process to mode locking with a broadband gain medium capable of supporting femtosecond pulses. We demonstrate that for a fairly large phase mismatch ($\Delta kL \sim 10\pi$) the frequency chirp generated in the femtosecond cascade process is approximately linear over the center of the pulse, similar to that produced by the electronic Kerr effect. The laser performs as expected theoretically, and nearly transform-limited pulses as short as 60 fs are generated without a prism pair in the cavity.

DeSalvo *et al.* showed[4] that, for large phase mismatch ΔkL or low intensity or both, the nonlinear phase shift produced in the cascade process is approximately

$$\Delta\Phi^{\mathrm{NL}} \approx -\Gamma^2 L^2/\Delta kL\,, \tag{1}$$

where $\Gamma = (\omega d_{\mathrm{eff}}|E_0|)/(c\sqrt{n_{2\omega}n_{\omega}})$, E_0 is the incident fundamental field, and $\Delta k = k^{2\omega} - 2k^{\omega}$. This is the result of a plane-wave analysis that neglects (in addition to saturation of the cascade process) the

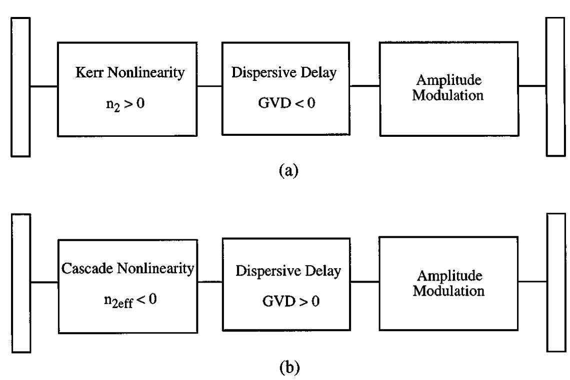

Fig. 1. Block diagrams of femtosecond KLM lasers: (a) traditional approach, (b) approach described here.

effects of GVM, which are generally significant for femtosecond pulses.

It is clear that highly efficient SHG is required for generation of a phase shift that will be useful for KLM. We recently showed that the GVM for frequency doubling 1.3-μm light is small in LBO, and because the phase matching is nearly noncritical the acceptance angles are large and the spatial walk-off is small. These properties allowed us to demonstrate 44%-efficient frequency doubling of 3-nJ pulses as short as 50 fs from a mode-locked Cr:forsterite laser[8] and are favorable for intracavity SHG.

The simplified analysis of Ref. 4 is adequate for estimation of the magnitude of the cascade phase shift inside a femtosecond laser. We assume that the pulse energy will be a factor of 30 higher than as given in Ref. 8. With 100-fs pulses of 100-nJ energy and an 8-mm LBO crystal the analysis of Ref. 4 predicts that $\Delta\Phi^{NL}$(cascade) ≈ -1.5 rad with $\Delta kL \approx 2\pi$. The Kerr nonlinearity in the doubling crystal contributes $\Delta\Phi^{NL}$(LBO) $\approx +0.3$ rad, for a net phase shift $\Delta\Phi^{NL} \approx -1.2$ rad. We have solved the coupled-wave equations for pulse propagation in the SHG crystal numerically, including GVM and intensity-dependent phase matching. The numerical solutions show that for $\Delta kL \geq 6\pi$ the simplified analysis accurately predicts the magnitude of $\Delta\Phi^{NL}$ if the GVM is not larger than the pulse duration. The temporal evolution of $\Delta\Phi^{NL}$ is generally distorted, so the frequency chirp across the pulse is nonlinear. However, for $\Delta kL \geq 6\pi$ the phase follows the pulse intensity envelope nearly ideally, as is illustrated in Fig. 2. $|\Delta\Phi^{NL}|$ decreases slowly with ΔkL, so little penalty is incurred by operating at large phase mismatch, where the frequency chirp is more desirable for KLM.

We used spectrally resolved two-beam coupling[9] to measure the electronic Kerr nonlinearities of LBO and Cr:forsterite and found that n_2(Cr:forsterite) = 2×10^{-16} cm^2/W and n_2(LBO) = $1.4 n_2$(Cr:forsterite). Measurement of a 0.5-mm length of LBO with $\Delta kL = +2\pi$ shows that $\Delta\Phi^{NL}$ is negative and larger in magnitude than the nonlinear phase shift in a similar length of Cr:forsterite. If we assume theoretical scaling with length, we conclude that $|\Delta\Phi^{NL}|$ produced by the 8-mm LBO crystal should be adequate to produce a net negative nonlinear phase shift in a Cr:forsterite laser for ΔkL in the range 2π–20π. Direct measurements of $\Delta\Phi^{NL}$ versus ΔkL in the long crystal will be valuable, but the 1.3-μm pulse energies required for Z-scan measurements[4] are currently not available in our laboratory.

A laser designed to operate with $\Delta\Phi^{NL} < 0$ (Fig. 3) is quite simple: A fold for the SHG crystal is added to an ordinary Cr:forsterite laser.[10] The gain and SHG crystals are both 8 mm long. Mirrors with 10-cm radii of curvature are used in both folds, and the gain end of the cavity is imaged onto the SHG end such that conditions in the gain medium do not change with the addition of the SHG crystal. The repetition rate of the laser is 50 MHz. The GVD of the gain crystal is 400 fs^2, and that of the LBO crystal is -90 fs^2 at 1.27 μm. A pair of SF-6 prisms is set to permit variation of the cavity GVD from -3000 to $+2500$ fs^2, and a semiconductor saturable-absorber mirror[11] is used to start the mode-locking process while providing ~1% output coupling.

As a control experiment we translated the LBO crystal away from the beam waist and obtained normal KLM operation. The dependence of pulse duration τ on cavity GVD is plotted as open symbols in Fig. 4. From the slope of the graph at GVD < 0 we estimate the (round-trip) nonlinear phase shift $\Delta\Phi^{NL} \approx 0.4$ rad, and this agrees with both the value measured similarly for previous Cr:forsterite lasers and the value calculated by use of the measured n_2(Cr:forsterite) and the experimental intensity.

With the LBO crystal at the focus of the second fold, stable mode-locked pulse trains are generated in the range $6\pi < \Delta kL < 16\pi$. The broadest fundamental spectrum occurs with $\Delta kL \approx 10\pi$. With ΔkL fixed at this value, the pulse duration depends on GVD, as shown by the filled symbols in Fig. 4. Qualitatively, the trend is the reflection of normal KLM behavior about GVD = 0, as expected for $\Delta\Phi^{NL} < 0$. This includes the observation of an unstable region about GVD = 0, indicated by the dashed portions of the curves in Fig. 4. The time–bandwidth product is always less than 0.4 (a sech pulse shape is assumed) for $\Delta\Phi^{NL} < 0$ and GVD > 0. From the slope of the graph at positive dispersion we infer that $\Delta\Phi^{NL}$(total) ≈ -0.4 rad, which allows us to estimate $\Delta\Phi^{NL}$ produced by the cascade process:

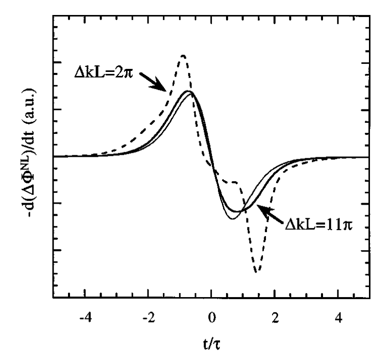

Fig. 2. Frequency sweeps generated in the cascade process with GVM equal to the pulse duration and $\Delta kL = 2\pi$ and $\Delta kL = 11\pi$. The magnitude of the phase shift produced with $\Delta kL = 2\pi$ is ~2.5 times as large as that produced with $\Delta kL = 11\pi$. The fine solid curve corresponds to an instantaneous nonlinear index of refraction.

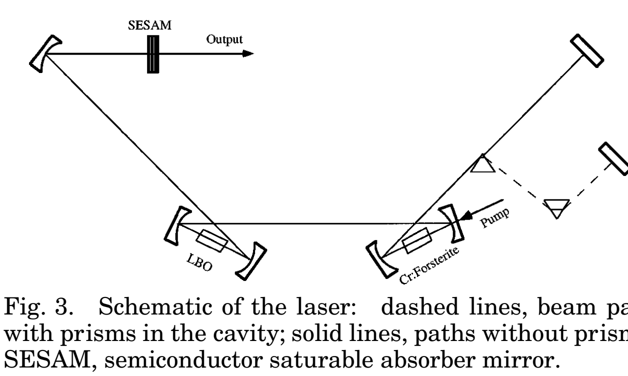

Fig. 3. Schematic of the laser: dashed lines, beam path with prisms in the cavity; solid lines, paths without prisms. SESAM, semiconductor saturable absorber mirror.

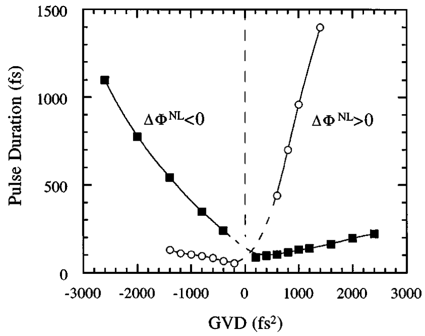

Fig. 4. Pulse duration plotted versus cavity dispersion. Open symbols, ordinary KLM; filled symbols, KLM with the cascade nonlinearity. The curves are to guide the eye, and dashed portions of the curves indicate unstable regions.

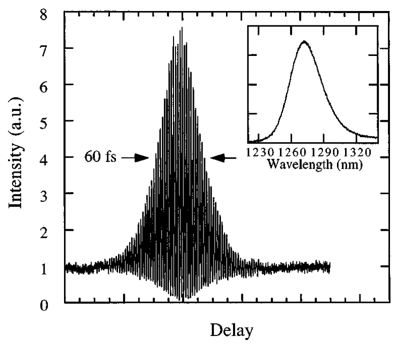

Fig. 5. Autocorrelation and power spectrum (inset) of pulses generated with prisms removed from the laser. The time–bandwidth product is $\tau\Delta v = 0.4$.

$$\begin{aligned}\Delta\Phi^{NL}(\text{cascade}) &= \Delta\Phi^{NL}(\text{total}) \\ &\quad - \Delta\Phi^{NL}(\text{Cr:forsterite, Kerr}) \\ &\quad - \Delta\Phi^{NL}(\text{LBO, Kerr}) \\ &\approx -0.4 - 0.4 - 0.6 \approx -1.4 \text{ rad.}\end{aligned}$$

The single-pass nonlinear phase shift generated in the cascade process is therefore -0.7 rad. This number agrees reasonably well with the value -0.6 rad calculated numerically or estimated from relation (1) with the experimental drive $\Gamma^2 L^2 \approx 18$.

We also obtain stable mode locking with $\Delta kL \approx -2\pi$, where $\Delta\Phi^{NL}$(cascade) is theoretically positive and near its maximum value. With positive GVD a narrow spectrum and long, highly chirped pulses are indeed observed, as expected for $\Delta\Phi^{NL} > 0$ and GVD > 0.[2,12]

The broad bandwidth of the femtosecond pulses precludes perfect backconversion to the fundamental, and the residual second harmonic constitutes a nonlinear loss that works to destabilize KLM. From the generated harmonic beam we estimate the nonlinear loss that is due to SHG as 1%. This amount of loss is just accommodated with the saturable-absorber mirror, which provides a fractional reflectivity increase of 0.8% at the intracavity pulse intensity. The laser is self-starting, but marginally so. If we adjust ΔkL to reduce the conversion efficiency, completely reliable self-starting operation is obtained.

The generation of transform-limited pulses in cavities with positive GVD potentially simplifies the design of mode-locked lasers. With the prism pair removed from the laser, the cavity dispersion is $+310$ fs^2. Under these conditions we observe transform-limited pulses as short as 60 fs (Fig. 5). In this demonstration the benefit of removing the prisms is offset by the addition of the second fold, but adding that fold may not be essential.

Finally, the laser is stable and already offers useful performance. Energies of $\sim$1 nJ are available for the shortest pulses, although the output power has not been optimized. The residual second-harmonic pulses have similar energy and should be useful either by themselves or in applications that need synchronized femtosecond pulses at 1270 and 635 nm. With $\Delta kL \approx 6\pi$, 130-fs pulses with 2-nJ energy are generated, but the harmonic pulse energy decreases to 0.2 nJ.

We have demonstrated the operation of a KLM laser in which the nonlinear phase shift is negative. Crucial in this development is the recognition that undistorted phase shifts can be generated by the cascade process at large values of ΔkL. The laser's performance agrees qualitatively with the theory of KLM, and pulses as short as 60 fs are generated with positive cavity dispersion. This approach allows us to envisage simple, compact femtosecond lasers consisting of gain and second-harmonic crystals in a single confocal cavity with ordinary laser mirrors.

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