Chirped-pulse Raman amplification for two-color, high-intensity laser experiments

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We report generation of millijoule-level Stokes (873, 961 nm) and anti-Stokes (738, 685, 639 nm) sidebands of 800 nm terawatt pulses by inserting a multistage Ba(NO$_3$)$_2$ Raman shifter-amplifier into a conventional Ti:sapphire chirped-pulse amplification system. The Raman subsystem requires no additional pump lasers and does not compromise the energy, duration, or beam quality of the main 800 nm pulses. The chirped first Stokes (870 nm) beam is compressed to transform limit (~100 fs) with several millijoules of energy and has high beam quality and is free of filamentation. Synchronized 800 nm and Raman sideband pulses are useful for a variety of two-color, high-intensity laser experiments.

1. INTRODUCTION

In many high-field experiments it is desirable to accompany the main terawatt (TW) pulse with a moderately powerful (~0.1 TW), temporally synchronized ultrashort pulse at a slightly shifted (Δλ ~100 nm) center wavelength outside the bandwidth of the main pulse. Such a secondary pulse can serve, e.g., as a spectrally distinct, low group-velocity-walkoff probe in centimeter-scale plasma waveguides [1], or induce beat-frequency modulation on the main pulse in order to seed the growth of a plasma wakefield via the forward Raman instability [2]. This pulse can also be used to realize two-color laser-plasma interaction schemes that underlie theoretical proposals to amplify [3], compress [4], and focus [5] intense laser pulses in plasmas. Three-wave parametric processes in nonlinear crystals cannot produce such small Δλ because of IR absorption of the idler wave. To avoid this problem, Zhavoronkov et al. [6] demonstrated the generation of 80 μJ, subgigawatt, 870 nm pulses by stimulated Raman scattering (SRS) of chirped 1.5 mJ, 800 nm pulses in barium nitrate, followed by recompression to ~190 fs. Because it uses chirped pulses to avoid damage and self-phase modulation in the Raman-active medium, this technique is well suited to TW-scale laser systems based on chirped-pulse amplification (CPA), but so far this has been demonstrated only on a much lower power system.

In this paper, we report scaling the chirped-pulse Raman amplification (CPRA) technique to a TW CPA laser system, resulting in 873 nm pulses up to 3 mJ, 0.03 TW synchronized with 800 nm, 5 TW pulses. The focused 873 nm output beam easily ionizes air and experiences ionization blueshifts. This energy was achieved purely by CPRA, without adding any pump lasers to the CPA system and without compromising the amplified energy of the main 800 nm pulses. Further amplification of the 873 nm output by conventional methods, e.g., a multipass Ti:sapphire amplifier with an additional pump laser prior to compression, appears straightforward. Scaling CPRA from microjoule to millijoule output is challenging because of the onset of beam filamentation and higher-order Raman processes in high-gain solid-state Raman amplifiers. We controlled these processes by distributing Raman gain among three passes, enabling spatial and spectral filtering between passes. As a result single-sideband, high-quality, unfilamented Gaussian pulses are generated. Another significant advance on the lower-power demonstration of Zhavoronkov et al. is the compression of the first Stokes output to near-transform-limited duration of 100 fs by custom-designing an optimized compressor for 873 nm pulses based on 830.8 lines/mm diffraction gratings. Other notable features of the Raman-amplified output are its high contrast compared with the main pulse, making it potentially useful as a pump in high-intensity, solid-target experiments, and the simultaneous production of a cascade of chirped anti-Stokes (738, 685, and 639 nm) and second-Stokes (961 nm) pulses as by-products of the chirped-pulse Raman amplifier, each compressible in principle with a suitably designed grating pair and potentially useful for synthesizing few-femtosecond and subfemtosecond optical pulses [7–9].

2. EXPERIMENTAL PROCEDURE

Figure 1 presents an overview showing how the Raman shifter and amplifier is incorporated into, and temporally synchronized with, the overall Ti:sapphire CPA system. An amplified stretched laser pulse (wavelength λ = 800 nm, duration τ = 200 ps, energy $E = 180$ mJ, repetition rate 10 Hz) is derived from an intermediate amplifier stage of the Ti:sapphire CPA system. The chirped-pulse duration was measured by background-free autocorrelation using second-harmonic generation in a phase-matched beta barium borate crystal (see lower left inset of Fig. 1). This 800 nm pulse was split into two pulses with...
intensity ratio 15:85. The weaker pulse (≈30 mJ) was Raman shifted to 873 nm with 6 mJ energy, then compressed by a dedicated 873 nm compressor (830.8 lines/mm gratings) to 115 fs. The stronger pulse (≈150 mJ) was further amplified to ≈600 mJ—the same energy achieved in the absence of the CPRA subsystem—then sent to an 800 nm compressor (1200 lines/mm gratings) and compressed to 90 fs. Both pulses were combined by a dichroic mirror and sent to the experimental chamber. Because the 800 nm pump pulse generates the CPRA pulse, the two pulse envelopes are temporally synchronized. Synchronization was confirmed by generating their sum frequency as a function of time delay in a KDP crystal and by pump-probe experiments—e.g., defocusing of the 873 nm pulse induced by pump ionization of air. The cross-correlation widths extracted from these measurements were consistent with autocorrelation widths described below.

Figure 2 shows detail of the Raman shifter and amplifier. The incident 800 nm pump beam is split in the approximate ratio 1:100. After the beam splitter, the weaker portion (~300 μJ) is focused at f/20 into a Ba(NO₃)₂ crystal to generate an 873 nm first-Stokes seed, primarily by SRS. Crystal position is adjusted longitudinally to optimize gain and Stokes beam quality. The Ba(NO₃)₂ crystal, purchased from Marketech International Inc., was 5 cm long with a 1 cm × 1 cm cross section and had uncoated end faces normal to the pump and seed beams. Ba(NO₃)₂ is used because of its high gain coefficient in the stationary regime, compared with other solid-state Raman shifters [10]. Because the Raman mode dephasing time $T_2 ≈ 25$ ps in Ba(NO₃)₂, SRS shows characteristics of the transient regime [6,11] in our system.

The remaining 99% of the pump pulse (~30 mJ) passes through a delay line to synchronize its arrival at the second stage with the 873 nm seed pulse. The unfocused pump pulse enters the face of the second identical Ba(NO₃)₂ crystal at normal incidence, propagates through with Gaussian beam waist $w_0 = 0.3$ cm, and is then retroreflected for a second pass. The unfocused seed pulse propagates through this crystal with beam waist $w_0 = 0.25$ cm at a small angle (3 to 5°) to the pump path that is adjusted to optimize phase matching, and therefore gain. Phase matching is more important in the second stage than in the first because of the greater importance of four-wave mixing (4WM) relative to SRS in the amplification process. The seed pulse is then returned to the gain crystal for a second pass at an equivalent angle via a bow-tie configuration. The amplified 873 nm pulse is then sent to its custom-designed compressor.

Even though the chirped pump and seed pulses are of ~200 ps duration, the pump–seed delay $\Delta t$ in the second stage must be adjusted to an accuracy of ~1 ps, as illustrated in Fig. 3. First-Stokes gain is peaked narrowly around zero delay with a FWHM of 2.8 ps, consistent with the reciprocal line width of the Raman transition, taking into account gain broadening [10,12]. This narrow maximum demonstrates that the chirps of the seed and pump pulse are commensurate. Beyond the central peak, low-level gain is observed out to $\Delta t ≈ 60$ ps, evidently from the...
long tail of the Raman gain spectrum. When undesired pre- or post-pulses within tens of picoseconds of the primary pulse are present in the main 800 nm CPA system, they often appear as satellite peaks in the pump–seed delay scan, as illustrated by the peaks at $\Delta t \approx 10$ and 30 ps in Fig. 3. Usually such prepulses can be detected only after compression. Moreover, pre- and post-pulses separated by tens of picoseconds from the main pulse can be difficult to characterize, because these delays are too short for many fast photodetectors to resolve yet beyond the scanning range of most autocorrelators designed for femtosecond pulses. Satellite peaks in second-stage CPRA delay scan therefore provide a convenient built-in diagnostic of pre- and post-pulses at time delays that are critical for many high-intensity, solid-target experiments. Moreover, the narrow width of the satellite peaks demonstrates that prepulses in the 800 nm system are not transferred to the Stokes pulse when $\Delta t = 0$.

As 4WM increasingly dominates second-stage amplification, a cascade of higher-order Raman lines can be observed after each pass when the angle between the pump and incident seed beams is optimized to yield highest overall energy. A photograph of the Raman cascade viewed on a fluorescing screen following the first pass of the second stage is shown at the top of Fig. 2. The cascade consists of first (873 nm) and second (961 nm) Stokes and first (738 nm), second (685 nm), and third (639 nm) anti-Stokes radiation. The various wavelengths are spectrally dispersed at angles determined by phase matching in the crystal. Such cascades were observed neither from the first stage nor in previous work [6]. In the normal operation reported here, the pump–seed intersection angles in both stages, and the reflectivity spectra of the amplifier mirrors are chosen to optimize the first Stokes output. However, any of the other Stokes or anti-Stokes beams shown in Fig. 2 could be individually optimized and compressed in a similar manner. Using more elaborate multichannel phase control, it should be possible in principle to compress the entire cascade to synthesize ultrashort pulses [7–9].

3. RESULTS AND DISCUSSION

A. First-Stokes Pulse Energy and Transverse Mode
The two main hurdles encountered in scaling CPRA from microjoule [6] to millijoule output energy are 1) onset of self-focusing and filamentation and 2) competition between first-Stokes and higher-order Raman amplification.

The first issue limits output energy of the first (SRS) stage to $\approx 0.1$ mJ. Below this threshold, a high-quality Gaussian mode can be achieved. Attempts to extract higher energy result in a filamented beam of limited usefulness, as shown in Fig. 4(a). Twenty percent conversion (i.e., $\approx 1$ mJ) to first Stokes is possible, but only with a pump that exceeds the critical power for self-focusing. In normal operation, we restrict first stage output to $\approx 20$ $\mu$J in order to completely eliminate filamentation.

Higher first-Stokes energy is then achieved by seeding a second Raman amplification stage with the high-quality output mode of the first stage. This reinitiates Raman amplification with the nonfilamented second-stage pump. The mode quality of the first stage output is then preserved through amplification to millijoule energies in the second stage, as shown in Fig. 4(b) for energy $\approx 1$ mJ. At this energy, Raman beam “cleanup” [12] occurs—i.e., the amplified Stokes beam assumes the pump-beam profile with smoothed irregularities. For example, the feature in the upper left corner of the profile in Fig. 4(b) was transferred from the pump, where it was much more prominent. To maintain good beam quality, overall gain in the seeded second-stage amplifier was kept $\approx 300$. Because a seed was injected, pump intensity could be kept below the level ($\approx 1$ GW/cm$^2$) required for single-pass Raman amplification to the same energy, thus preventing filamentation.

The second issue sets the primary limit on Raman gain in the second stage. Higher-order Raman processes (e.g., four-wave mixing) become important as the energy of the growing Stokes pulse approaches $\approx 10\%$ of pump energy. These competing processes deplete both first Stokes and pump energy, causing first-Stokes gain to saturate well below the theoretical quantum efficiency (91.6%) of SRS. To manage these saturation effects, we divided the second stage amplifier into two passes. After the first pass, the first-Stokes component of the incipient Raman cascade is spectrally and spatially filtered then preferentially seeds second-pass Raman gain. In this manner, first-Stokes gain saturates at a higher level than for a single pass through a crystal twice as long. Figure 5 shows typical Raman gain saturation for the complete two-stage Raman amplifier. Second-stage gain is observed above a threshold pump energy $\approx 12$ mJ. Saturation becomes significant above $\approx 16$ mJ pump energy, indicating that the amplifier

![Fig. 4. (Color online) Transverse profiles of first Stokes beam (a) produced by single-pass stimulated Raman amplification to $\approx 1$ mJ energy, showing filamentation and (b) similar energy produced by seeded Raman amplification in the second stage, showing nearly Gaussian mode. Pump intensity is much lower in the seeded amplifier.](image)

![Fig. 5. Stokes beam energy out of the seeded second-stage, two-pass Raman amplifier as a function of second-stage pump energy.](image)
is approaching optimum efficiency. Uncompressed Stokes pulse energy of 6 mJ is routinely achieved with pump energy of 25–30 mJ, sufficient for multigigawatt compressed power and $10^{16}$ W/cm$^2$ focused intensity.

B. Pulse Duration

Even though the CPRA pulse acquires the chirp of the main pulse, it is poorly compressed by 1200 lines/mm gratings matching those in the main pulse compressor and stretcher [6]. This is because the Stokes shift breaks the symmetry between stretching and compression mechanisms. Figure 6(a) shows the compressed first-Stokes pulse for our system calculated from a model of laser–Raman amplifier system, assuming 20 nm bandwidth, with an optimized compressor based on 1200 lines/mm gratings, using the ray-tracing program ZEMAX and a Fourier transform routine. The compressed first-Stokes pulse shows strong residual asymmetric chirp and a prepulse train extending over $\sim$1 ps.

Optimal compression of the CPRA pulse requires gratings mismatched to those in the main pulse compressor and stretcher. We investigated alternative first-Stokes compressors using ZEMAX, focusing on commercially available gratings that could be made large enough to be useful in a high-energy system. For each line density, grating separation and beam angle were adjusted to optimize compression with group-velocity dispersion (GVD) and third-order dispersion (TOD) zeroed. Residual fourth-order dispersion (FOD) could be minimized by further compressor adjustments. Residual FOD in the main pulses proved to be the main limit to compressed first-Stokes pulse duration. Figure 6(b) shows a calculated 20 nm bandwidth, 60 fs FWHM profile, compressed with 830.8 lines/mm gratings separated by 225.6 cm, with the 873 nm pulse incident at 53° on the first grating. The asymmetric prepulse train is eliminated and peak power improved fivefold compared with compression with matched gratings [Fig. 6(a)].

On the basis of these calculated results, we constructed a CPRA compressor using 830.8 lines/mm gratings purchased from Richardson Grating Laboratory. A single-shot autocorrelator measured compressed pulse durations with the Ti:sapphire and CPRA systems operating at approximately 10 nm bandwidth. As a result, measured 873 nm pulse durations were approximately twice as long as for our system calculated from a model of our laser–Raman amplifier system, assuming 20 nm bandwidth, with an optimized compressor based on 1200 lines/mm gratings, using the ray-tracing program ZEMAX and a Fourier transform routine. The compressed first-Stokes pulse shows strong residual asymmetric chirp and a prepulse train extending over $\sim$1 ps.

Fig. 6. Temporal profiles of compressed first-Stokes beam. The top two profiles are calculated from a model of laser–Raman amplifier and optimized compressor with (a) 1200 line/mm and (b) 830.8 line/mm gratings based on the ray-tracing program ZEMAX. Residual fourth-order dispersion is $1.9 \times 10^6$ fs$^4$, with $2.5 \times 10^4$ fs$^4$ in the incident main pulse. (c) Measured single-shot, background-free SHG autocorrelation trace of first-Stokes pulse from compressor with 830.8 line/mm gratings separated by 225.6 cm, with pulses incident on first grating at 53°. The trace corresponds to a pulse of FWHM=115 fs, assuming a sech$^2$ pulse shape.

Fig. 7. (Color online) Illustration of focused intensity. (a) Intensity profiles of first-Stokes and fundamental pulses at focus of f/6 parabola. (b) Schematic (top) and digital (bottom) photographs showing the incident focused Stokes beam fluorescing on a screen (left), the laser produced spark at focus (center), and the ionization defocused beam after the focus (right). Ionization blue-shifted light from the IR Stokes beam is visible.
as the calculated pulse in Fig. 6(b), as illustrated by the experimental single-shot autocorrelation in Fig. 6(c). Assuming sech$^2$ pulse shape, this trace corresponds to a temporal FWHM of 115 fs. Even at 10 nm bandwidth, this compression is significantly better than previous results with matched gratings in the 800 nm and 873 nm systems [6].

C. Focused Intensity
Figure 7(a) shows images of the spot profile of the CPRA (873 nm) and main (800 nm) pulses in vacuum after focusing by an f/6 off-axis parabolic mirror. The two pulses focus to very similar spot sizes and shapes, confirming that the profile of the pump pulse is transferred to the CPRA pulse with high fidelity. In vacuum, the focused, compressed CPRA pulse reaches an estimated peak intensity $>10^{18}$ W/cm$^2$ with $\sim 3$ mJ energy. In air, the same pulse produces a bright ionization spark [Fig. 7(b), center]. After the spark, the pulse experiences plasma lens defocusing, and an ionization blueshift to visible red wavelengths, as shown in Fig. 7(b), right. Relativistic intensity should be achievable with wider bandwidth and modest further amplification of the CPRA pulse in Ti:sapphire.

4. CONCLUSION
We have augmented a TW-scale 800 nm CPA system with a solid-state Raman amplifier subsystem that produces 873 nm, $\sim 100$ fs, sideband pulses at the millijoule level with high beam quality without adding pump lasers and without diminishing the output of the primary system. Raman gain is distributed between a seed-pulse generation stage in which SRS dominates and a two-pass power amplifier stage in which 4WM dominates. This division enables control of filamentation, generation of multiple Stokes and anti-Stokes sidebands, and channeling of second-stage gain into a selected sideband. Further compression of the sideband pulses in wider bandwidth systems, and further amplification in conventional multipass Ti:sapphire amplifiers with additional pump lasers appear straightforward. These synchronized 800 nm and Raman sideband pulse trains should be useful for a variety of high-intensity, two-color experiments.

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