Efficient noncollinear parametric amplification of weak femtosecond pulses in the visible and near-infrared spectral range

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We report measurement of efficient amplification of weak femtosecond supercontinuum seed pulses by use of a noncollinear optical parametric process in BBO crystal pumped with 150-fs pulses from a frequency-doubled regenerative-amplified Ti:sapphire laser at 390 nm. The highest amplification factor, $10^8$, was achieved for $3 \times 10^{-16}$ J energy seed pulses at wavelength of 560 nm.

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 Optical parametric generation (OPG) and optical parametric amplification (OPA) are widely used for tunable wavelength conversion of different pulsed laser sources, including high-intensity ultrashort pulses from amplified solid-state lasers. It is well known that the birefringence and dispersion properties of common nonlinear crystals such as BBO, LBO, and LiIO$_3$ allow phase matching between the wave vectors of the pump and the signal (idler) beams to be achieved for collinear as well as for noncollinear propagation directions. For pulses with durations in the picosecond or the nanosecond range, it is usually observed that the efficiency of the collinear parametric process is higher than the efficiency of the corresponding nonlinear processes. This higher efficiency occurs because the interaction length of pump and parametric waves that are overlapping inside a nonlinear crystal is much longer for parallel beams than for beams that are crossing at an angle. Accordingly, efficient conversion of the pump wavelength into signal and idler wavelengths was achieved in long nonlinear crystals for picosecond and nanosecond pulses.

For high-intensity ultrashort pump pulses produced by amplified solid-state lasers it was observed that both collinear and noncollinear parametric processes can have high efficiency, resulting in transformation of a considerable fraction of the pump-pulse energy into the pulses at the signal and the idler wavelengths. This transformation is explained, on the one hand, by the fact that high-intensity femtosecond pulses achieve high parametric gain in nonlinear crystals within a relatively short interaction length of few millimeters. On the other hand, the highest parametric conversion efficiency of ultrashort pulses is achieved when the phase-matching condition is accompanied by matching of the group velocities of the interacting pulses. Especially in nonlinear crystals with large dispersion, and for pump-pulse wavelengths shorter than 400–500 nm, the matching of group velocities is of utmost importance. In particular, it may turn out that the condition for parametric generation is fulfilled preferentially for noncollinear interaction rather than for collinear interaction.

The ultrafast wavelength-conversion techniques that have been investigated so far have been designed to achieve the goal of increasing the efficiency of collinear OPG and OPA, often by means of suppressing the accompanying noncollinear process. Several suppression methods, such as seeding with white-light continuum pulses and multiple-pass amplification in two or more crystals, were investigated. We reported recently that efficient noncollinear parametric generation was produced in BBO and LiIO$_3$ crystals pumped by frequency-doubled pulses of an amplified Ti:sapphire laser at a wavelength of 390 nm. In this Letter we investigate one-pass noncollinear parametric amplification in BBO crystal by seeding with a femtosecond white-light continuum. For the first time to our knowledge, we accurately measure the amplification coefficient for noncollinear OPA in a broad visible spectral range. We demonstrate background-free detection of very weak ($3 \times 10^{-16}$ J) femtosecond signals with an amplification factor of $10^8$ and report detailed energy, spectral, and temporal measurements of the noncollinear OPA process.

A schematic of our experimental setup is shown in Fig. 1. The femtosecond laser source was a 1-kHz repetition-rate Clark MXR CPA-1000 Ti:sapphire regenerative-amplifier laser system. After frequency doubling the laser output in a 0.5-mm-thick BBO crystal, we obtained 200-fs pulses with 0.3-mJ energy at the 390-nm wavelength. For OPA we used a 1.5-mm-thick BBO type I crystal cut at $\theta = 31^\circ$. The 390-nm pump beam was focused to a 1.5-mm diameter by a $4 \times$ telescope. To produce a supercontinuum, we diverted a small fraction of the fundamental beam and focused it into 5-mm-thick fused-silica block. The supercontinuum beam was collimated with a lens and
Fig. 1. Experimental setup: SHG, second-harmonic generator (BBO crystal); OPA, optical parametric amplifier (BBO crystal); BS, dichroic beam splitter; SC, glass plate for supercontinuum generation; FP, Fabry–Perot etalon; T×4, telescope; AC, autocorrelator; PM, optical power meter; SP, grating monochromator; α, angle between the seed and the pump beams.

then directed into the BBO crystal at an angle α with respect to the pump beam. The duration of the seed pulses was measured to be 200 fs. The time delay between the pump and the seed pulses was controlled by an optical delay line. To measure simultaneously the energy, spectrum, and duration of the pulses, we split the beam at the output of the OPA among an optical powermeter, a grating spectrometer, and an autocorrelator.

When only the pump pulses were applied to the BBO crystal, parametrically generated signal and idler waves were observed at the output of the crystal as two cones centered around the propagation direction of the pump beam. Figure 2 shows the brightly colored ring that was produced when the signal wave was projected upon a screen placed behind the crystal. As we reported in Ref. 7, the OPG signal pulses in this case had a broad spectrum with a width of several tens of nanometers. Changing the phase-matching angle made the signal wavelength tunable in the range 460–780 nm, with the maximum of output energy at 560 nm. The second ring, corresponding to the idler wave, had a slightly larger cone angle, but it could not be photographed directly because its wavelength was tuned to the near-IR range of λ = 2.42 μm–820 nm. Our calculations show that this tunable range is determined by group-velocity-matching conditions for noncollinear OPG. Figure 2 shows the image of the signal wave when the white-light continuum seed beam was spatially and temporally overlapped with either (a) the signal wave or (b) the idler wave. Figure 2(a) shows that by choosing a proper propagation angle we could observe efficient parametric amplification of the seed for wavelength components near 560 nm, giving rise to a bright yellow spot in the direction of the seed beam. Note that in this experiment the parametric generation spectrum contained a broad interval of wavelengths over several tens of nanometers, whereas the seed spectrum covered essentially the whole visible and near-IR spectral range. The noncollinear parametric process efficiently amplified only those wavelength components for which the seed pulse had the same propagation direction as the corresponding spontaneously generated beam. Accordingly, in our experiment the spectral width of the OPA was limited to ~10 nm.

Figure 3 shows the energy of the amplified seed pulses at the output of the crystal as a function of the energy of the pump pulses. The integrated energy of the input seed pulses measured at the wavelength 560 nm and in the spectral interval Δλ = 10 nm was roughly 2 × 10⁻⁵ μJ. We observed that efficient amplification of the seed signal started at a pump energy of 5 μJ. In comparison, for the spontaneous OPG process the pump energy threshold was ~15 μJ. The seeding not only reduced the threshold of noncollinear generation by a factor of 3 but also increased the energy of the signal wave propagating in the direction and within the divergence angle of the seed by at least a factor of 20. In addition, the seeded parametric process yielded an improved pulse-to-pulse energy stability and a better spatial beam quality than that of conventional OPG in the same configuration.

We define the amplification factor of OPA as the ratio of the output and the input pulse energies at
the intensity of the seed pulses conveniently simply by chang-

Using this arrangement, we were able to vary the inter-

toral width of a Fabry–Perot etalon composed of two 50% reflectiv-

For this measurement we inserted into the seed beam energy not only of the pump but also of the seed pulses. This result corresponds well to the values reported in Refs. 1–7, in the range $K = 10^4$–$10^6$, which were obtained in various collinear OPA experiments by use of nanosecond, picosecond, and femtosecond pump pulses. However, this amplification coefficient is still orders of magnitude below the theoretically predicted value, which is as great as $10^{10}$–$10^{12}$. To clarify this point we carefully measured the amplification factor by varying in our experiments the angle $\alpha$ at which the seed beam is directed.

In conclusion, we have demonstrated that optical parametric gain in a BBO crystal pumped by ultrashort pulses can be as great as $10^8$. We have shown that high amplification is obtained by use of a seed in the signal or the idler beam. This method can be used for zero-background measurement of the time profile of weak ultrashort signals with energies as low as $10^{-16}$ J.

References