Sub-20 fs visible pulses with 750 nJ energy from a 100 kHz noncollinear optical parametric amplifier

J. Piel and E. Riedle

Lehrstuhl für BioMolekulare Optik, Ludwig-Maximilians-Universität, Oettingenstrasse 67, 80538 München, Germany

L. Gundlach, R. Ernstorfer, and R. Eichberger

Hahn-Meitner-Institut, Abteilung SE 4, Glienicker Strasse 100, 14109 Berlin, Germany

Received November 2, 2005; revised January 29, 2006; accepted January 31, 2006; posted February 2, 2006 (Doc. ID 65780)

We demonstrate the operation of a 100 kHz noncollinear optical parametric amplifier that is pumped by just a few microjoules of 800 nm pulses with 50 fs duration. The device delivers sub-20 fs pulses tunable from 460 nm to beyond 1 μm and pulse energies up to 750 nJ when it is pumped with 7 μJ of energy. The design of the single-stage amplifier has been carefully optimized, and the design considerations are discussed.

© 2006 Optical Society of America

OCIS codes: 190.4970, 190.7110, 300.6530

For applications in physics, chemistry, and biology, tunable pulses with durations of less than 20 fs are needed to resolve the dynamics on the time scale of intramolecular nuclear motion. Many samples are damaged when they absorb strong laser pulses. Therefore measurements with weak pulses at high repetition rates are required. It is extremely hard to generate tunable sub-20 fs pulses at the 80 MHz repetition rate of Ti:sapphire oscillators. Near 1 kHz, nonlinear optical conversion is well established, and tunable sub-20 fs visible and even UV pulses can be generated. The low repetition rate combined with the necessary weak laser pulses for the samples mentioned above results in extremely long averaging times. An intermediate repetition rate of ~100 kHz is therefore of great interest. We investigated the scaling of a noncollinear optical parametric amplifier (NOPA) to pump energies of ~5 μJ available from 100 kHz Ti:sapphire amplifiers. Such amplifiers are routinely used to pump collinear optical parametric oscillators (OPAs); however, the OPA output cannot be compressed to much below 50 fs. Noncollinear operation permits an extremely wide spectral amplification range but has so far not been reported at 100 kHz.

In a preliminary experiment (see Fig. 1) we pumped a NOPA with 150 fs pulses from a 1 kHz Ti:sapphire amplifier (CPA 2001; Clark MXR, Inc.) A fraction of the 775 nm light was used to generate a seed continuum. A neutral-density filter was placed in the main beam to attenuate the pump. The position of the doubling crystal and the overlap in the β-barium borate (BBO) amplifier crystal were optimized. The energy of the green output scales according to the expected cosh² dependence on the blue pump energy (Fig. 1, inset) and stable amplification can be reached for a pump energy well below 1 μJ. A red pump energy of just a few microjoules should therefore facilitate the operation of a NOPA. We found that the low pump energy NOPA requires a significantly different beam geometry from the 1 kHz setups pumped by 200 μJ. In the latter, one can adjust the intensity at the amplifier BBO crystal to the desired value of >100 GW/cm² by placing the crystal some distance behind the focus of the blue pump light, similar to the situation reported for collinear OPAs. Alternatively, a telescope is used in some kilohertz systems; however, a variable attenuator is then needed, which will result in a loss of pump power that is unacceptable for 100 kHz systems. Unfortunately, positioning the BBO crystal close to the focus but still outside the Rayleigh range led to a poor spatial mode, instable operation, and low efficiency. The reason for this was not investigated in detail, but rather the beam geometry was modeled by Gaussian beam propagation and a setup developed that renders ~150 GW/cm² in the focal plane. This makes it possible to place the BBO crystal in the focus. The setup allows one to adjust the intensity at the amplifier crystal without moving it outside the Rayleigh range.

For a given pump energy and pulse duration the desired intensity leads to a value of the pump focus diameter of 2w₀. Together with a predetermined distance of the amplifier crystal from the focusing mirror M, the necessary beam size 2wM at mirror M follows. Figure 2 shows the beam diameters as the...
The NOPA configured according to the considerations given above (Fig. 1) is pumped by a 100 kHz Ti:sapphire amplifier system (Mira seed laser pumped by Model Verdi V5 laser and RegA 9050 pumped by Model Verdi V10; Coherent, Inc.) delivering 7 μJ pulses at 800 nm, a spectral width of 27 nm, and a duration of 50–55 fs. Ten percent of the red light is used to generate a single-filament continuum in a 3 mm sapphire disk. The continuum is recollimated with a fused-silica lens and focused to a spot slightly before the NOPA amplifier crystal. A p-coated 300 μm thick, type I BBO crystal is used to double the main part of the red light. The resultant 2.7 μJ pulses at 400 nm with a bandwidth of 6.1 nm are collimated with lens L₂ and chirped to a length of ~140 fs with a pair of 0.25 in. (6.35 mm) fused-silica plates mounted at Brewster’s angle. This ensures good temporal overlap with the chirped seed light and helps to increase the effective amplification bandwidth. The blue beam is steered with a plane mirror and focused by mirror M placed a distance of 488 mm from lens L₂. The plane steering mirror is mounted on a translation stage for fine-tuning of the temporal overlap between the seed and the pump pulses and thereby tuning of the NOPA output. The two beams overlap in the 2 mm BBO crystal (cut for type I phase matching) at an internal angle of 3.7°. This noncollinearity permits effective group-velocity matching of signal and idler waves and consequently a large amplification bandwidth and sub-20 fs pulse lengths. The precise value of the noncollinearity angle is adjusted for the desired output wavelength.

Strong and stable output is obtained from the setup described. At the maximum of the tuning curve a pulse energy of 750 nJ is reached, corresponding to a quantum efficiency of more than 30% from the blue and an overall energy conversion efficiency (800 nm to green) of 10%. The efficiency falls off in the blue because of idler absorption in the BBO crystal and toward longer wavelength (see Fig. 3) because of
The NOPA can be tuned from 460 to 720 nm and from 850 nm to at least 1 μm (see Figs. 3 and 4). The near-infrared wavelengths are also generated on axis and utilize the near-infrared part of the seed light. The spectral bandwidth is sufficient for sub-20 fs pulses throughout the tuning range. With a fused-silica prism compressor we measure durations below 20 fs (Fig. 4) and a time–bandwidth product of ~0.5. The autocorrelator utilizes the two-photon-induced current in a SiC diode and second-harmonic generation in BBO crystals of 50 and 100 μm thickness. For visible operation the amplifier BBO crystal is not walk-off-compensated to prevent self-doubling of the 680 nm output. In the near infrared the walk-off-compensated orientation suppresses self-doubling of 880 nm light.

The 100 kHz NOPA has been used for an extended period for spectroscopic measurements. The BBO crystals and other optical components show no signs of aging. Its high stability and capability for rapid data averaging have made the setup a most valuable tool for the transient absorption measurements of photoinduced femtosecond electron transfer from the excited singlet state of a perylene chromophore into nanostructured anatase TiO₂. To provide the 15 fs pump pulses at 440 nm we tune the NOPA to 880 nm and double it in a 100 μm BBO crystal. In the future it should be possible to maintain the high efficiency and stability of the setup for even lower pump pulse energies and higher repetition rates. Such pump systems appear rapidly on the market and will quickly find their way into research labs. Adaptation to the various pump pulse parameters—including changed laser wavelengths—will be possible along the guidelines reported in this Letter. A reduced chirp of the seed light and optimized compression should lead to sub-10 fs pulses, as demonstrated for 1 kHz systems.

The authors thank Frank Willig for continuous support of the project. Financial support from the Deutsche Forschungsgemeinschaft is gratefully acknowledged. E. Riedle’s e-mail address is Eberhard.Riedle@Physik.uni-muenchen.de.

References