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

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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.

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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.[1] Near 1 kHz, nonlinear optical conversion is well established, and tunable sub-20 fs visible and even UV pulses can be generated.[2,3] 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 $\sim$100 kHz is therefore of great interest. We investigated the scaling of a noncollinear optical parametric amplifier (NOPA) to pump energies of $\sim$5 μJ available from 100 kHz Ti:sapphire amplifiers. Such amplifiers are routinely used to pump collinear optical parametric oscillators[4] (OPAs); however, the OPA output cannot be compressed to much below 50 fs.[5] 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.[6] 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 $\cosh^2$ 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/cm^2 by placing the crystal some distance behind the focus of the blue pump light, similar to the situation reported for collinear OPAs.[4,5] Alternatively, a telescope is used in some kilohertz systems[7]; 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 $\sim$150 GW/cm^2 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 $2w_0$. Together with a predetermined distance of the amplifier crystal from the focusing mirror M, the necessary beam size $2w_M$ at mirror M follows. Figure 2 shows the beam diameters as the

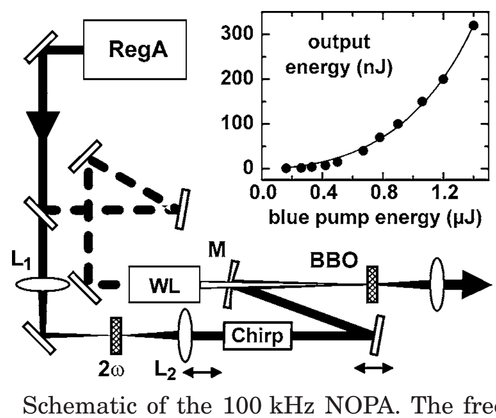

Fig. 1. Schematic of the 100 kHz NOPA. The frequency-doubled pump light is stretched to 140 fs with a chirper made from fused-silica plates. Inset, scaling of the output energy for 1 kHz pumping. Abbreviations defined in text.

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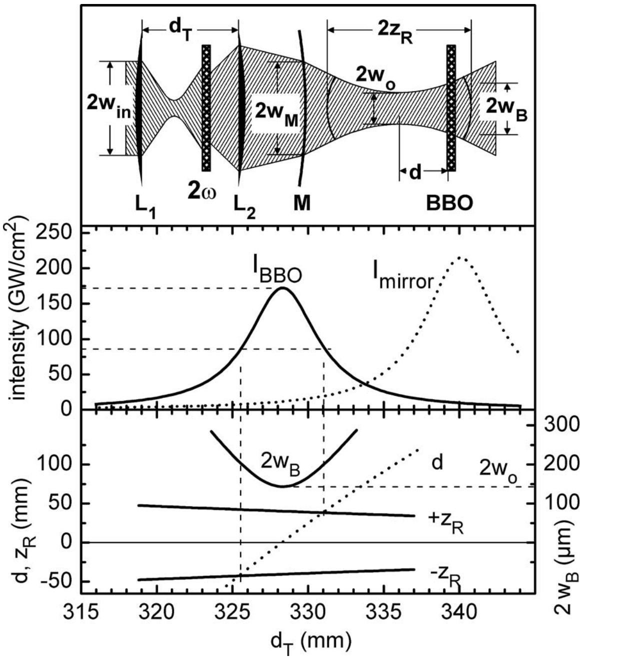

Fig. 2. Size of the pump beam and intensities within the NOPA. For details see text.

pump light propagates through the NOPA (cf. Fig. 1). The red input beam (diameter, 4 mm) is focused with antireflection-coated plano–convex BK7 lens L_1 ($f = 200$ mm). The BBO doubling crystal is placed slightly behind the focal plane to ensure good efficiency and no deterioration of the beam profile.[4,5] Compared to the red beam, the diameter of the blue beam and its divergence is smaller by the value of $\sqrt{2}$ expected for a quadratic process (confirmed by experiment). The plano–convex lens L_2 ($f=100$ mm; antireflection-coated fused silica) collimates the blue beam. The distance d_T between the two lenses can be adjusted with high precision and determines the value of $2w_M$. The radius of curvature of mirror M has to be chosen properly to focus the converging pump beam at the desired distance. We found a value of 1 m a good choice and therefore use it in the setup.

Once the optical components are chosen and the amplifier crystal is positioned 145 mm from the focusing mirror, only the distance d_T is changed for fine tuning. This does not significantly alter the diameter of the focus or Rayleigh length z_R but repositions the focus along the beam axis. An increase of d_T moves the focus closer to mirror M and leads to an increase of distance d between the focal plane and the BBO crystal. A change of d_T of the order of 1 mm significantly changes d (cf. Fig. 2, bottom), the diameter $2w_B$ of the pump beam at the fixed-position amplifier crystal, and the intensity I_{BBO}. This variation is used for optimization of the NOPA; at $d_T = 328$ mm, the highest intensity, 172 GW/cm^2, is found. The model also shows that the intensity at the focusing mirror is still quite low, and we have not observed damage of the dielectric coating. All parameters deduced from the modeling were confirmed to high accuracy by measurements on the operational NOPA.

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[2,4–6] 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 L_2 and chirped to a length of ~140 fs with a pair of 0.25 in. (6.35 nm) 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.[8,9] The blue beam is steered with a plane mirror and focused by mirror M placed a distance of 488 mm from lens L_2. 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[1] and consequently a large amplification bandwidth and sub-20 fs pulse lengths.[3,6] 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

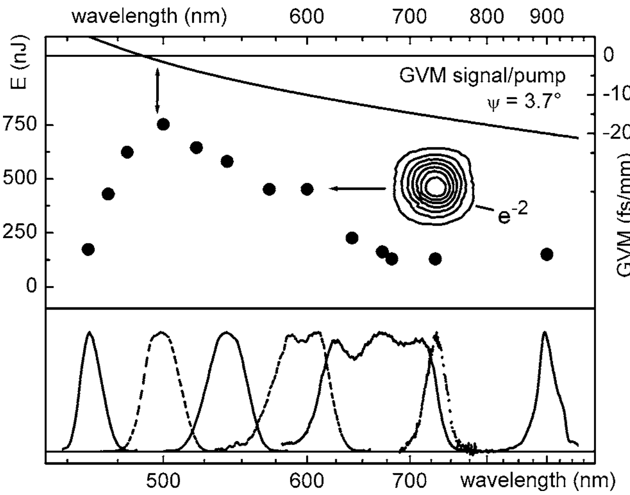

Fig. 3. Output energy and spectra. The spatial mode is shown in the inset, and the group-velocity mismatch (GVM) between the 400 nm pump light and the visible signal is given for a noncollinearity of 3.7°.

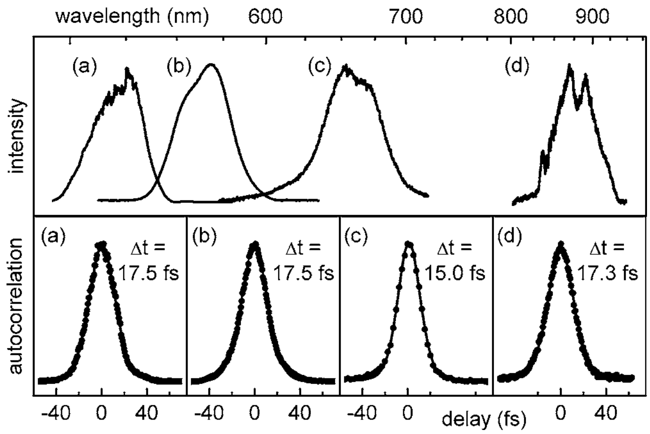

Fig. 4. Spectra and intensity autocorrelation traces (together with the $sech^2$-fitted pulse width) for pulses at four central wavelengths.

signal–pump group-velocity mismatch. The group-velocity mismatch is zero for signal pulses near 490 nm and increases to 150 fs/mm for 700 nm (Fig. 3, top). The effective overlap of a given spectral component with the 400 nm pump pulses decreases correspondingly to only 1 mm out of the 2 mm amplifier crystal. Even at the unfavorable wavelength of 880 nm we find a rms fluctuation of only 0.87%.

Compared to a two-stage collinear OPA,[4,5,10] the single-stage NOPA produces considerably more output energy and a much broader bandwidth (see below) and is more stable. The decreased number of components greatly simplifies the alignment. To set the proper phase matching angle, the various colors of the visible parametric fluorescence ring have to be collapsed. The overlap between the seed and the pump beams can then easily be found if the seed beam is spatially enlarged on the BBO crystal. It might be helpful if a 1 kHz system were used for prealignment and the beam positions are fixed with additional irises. As shown in the inset of Fig. 3, a nearly perfect Gaussian mode can be obtained at 100 kHz operation.

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.[11] 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 $\sim$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 TiO_2.[12,13] 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.[7,14–16]

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