Extension of a 1 kHz hard X-ray pump-probe setup by a few-cycle OPCPA pump at 11 μm

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A proven method to study ultrafast structural dynamics at atomic length scales is X-ray diffraction. Sources of hard X-rays driven by table-top ultrafast laser systems have been established in recent years for time-resolved X-ray absorption and diffraction studies [1]. The X-ray photon flux is generated by focusing pulses with few cycles and high energy onto metal targets. The electrons released are accelerated in the field of the laser driver. In order to increase the photon yield, pulses with a long optical period are advantageous [1]. This concept of scaling was impressively demonstrated using few-cycle pulses at 5 µm wavelength in a 1 kHz pulse train [2]. To perform time-resolved X-ray diffraction using this system a pump-probe (PP) line was implemented. With the purpose to investigate different excitation scenarios of the samples under study, pump pulses of different wavelengths are required.

Our four-stage mid-wave IR (MWIR) OPCPA delivers few-cycle pulses at 3.5 µm (signal) and 5 µm (idler) [3], which are direct available as pump pulses. The first successful PP experiments were carried out with a part of the idler at 5 µm and its SHG at 2.5 µm [4] as pump pulses. The availability of pump pulses in the long-wave IR (LWIR) with energies in the µJ range would enable interesting studies in the field of condensed matter physics, such as investigations of charge density wave states in rare-earth doped tellurite materials. To generate wavelengths beyond 10 µm, parametric down-conversion is typically the process of choice. Unfortunately, nonlinear crystals are only available to a limited extent for this spectral range. In addition, these crystals exhibit a comparatively low damage threshold. For this reason, the pulse energies for ultrashort pulse systems beyond 10 µm with kHz repetition rates have been limited to a few µJ so far [5,6].

Here we present the addition of a single-stage LWIR OPCPA as pump in a hard X-ray pump-probe setup. Few-cycle pulses at 11.2 µm containing 50 µJ energy are provided at a 1 kHz repetition rate. Utilizing the potential of our ZGP-based MWIR-OPCPA driver [3] the LWIR-OPCPA is implemented in the system. The still available residual energy of the 2.05 µm pump pulses after the fourth stage of the MWIR-OPCPA acts as pump for the LWIR OPCPA too. The signal pulses for the LWIR-OPCPA are provided by the SHG of the 5-µm idler applying GaSe. When using roughly 10% of the 3.4 mJ driver energy, the X-ray flux generated (Cu Kα-radiation) is not significantly affected. The generated LWIR-OPCPA signal pulses centered at 2.4 µm exhibit a bandwidth of 100 nm (FWHM). These pulses with energy of 40 µJ are stretched to 1.5 ps before parametric amplification to match the 2-µm pump pulse duration. GaSe is used as the nonlinear crystal in the single-stage OPCPA, which is designed in collinear geometry. The 2-mm thick crystal is implemented for Type-II phase matching profiting from the larger bandwidth compared to Type-I. To stay below the damage threshold of the GaSe crystal exhibiting 7 mm aperture, the maximum pump energy is limited to 4 mJ. Applying the latter an idler pulse energy of 70 µJ is generated. Its spectrum is centered at 11.2 µm and extends from 10.2 to 12.9 µm (1/e2 level). This corresponds to a Fourier-limited pulse duration of 140 fs. Anti-reflection coated ZnSe is used as compressor exhibiting a loss of 28The SHG frequency resolution optical gating technique (FROG) is used to characterize the pulses. The duration of the pulses was retrieved to be 180 fs, which corresponds to less than five optical cycles at the wavelength of 11 µm. This results in an impressive peak power of 300 MW which represents a record value for pulses beyond 10 µm wavelength. Finally, a further pump wavelength for fs hard X-ray PP experiments was thus successfully installed.

References

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