Abstract

Numerous applications, starting from linear and non-linear spectroscopy and extending to extreme non-liner optics and high field physics would benefit from using mid-IR high-energy, ultrashort pulse laser sources. The first group of the applications is related to the availability in this spectral range of a variety of wavelength-selective vibrational transitions while the second exploits the dominating role of tunneling ionization as well as high ponderomotive energy released during the ionization electrons.

However, due to vibrational transitions being dominant in this spectral range there is a lack of laser media, especially with broadband resonances corresponding to an ultrashort time response. Therefore the most popular approach to generate ultrashort mid-IR pulses is a nonlinear-optical frequency conversion by employing optical parametric (chirped pulse) amplifiers (OP(CP)A)s pumped by near-IR laser pulses. As long-wavelength as possible near-IR laser pump sources are demanded, because of both 1) the transparency of mid-IR crystals, effectively limited by two photon absorption; and 2) unfavorable quantum efficiency, while shifting to longer wavelengths. Yet development of such laser systems is challenging due to specific drawbacks of laser media lasing in the 1.9 – 3 μm spectral region: ions of Holmium support rather narrow emission line; laser media doped with Chromium typically have poor thermal conductivity; Thulium-based active materials possess low emission cross-sections, making energy extraction difficult; Erbium has a short upper state lifetime making energy storage inefficient. Among above mentioned laser materials Holmium-doped media are most attractive for the generation of pump pulses for driving OP(CP)As since they enable high energy storage and extraction. The narrow amplification bandwidth of Holmium is not an obstacle if a broadband coherent seed can be generated and amplified in an optical

parametric chirped pulse amplifier pumped by a relatively narrow-bandwidth output of a Holmium-based laser system.

This work is devoted to the development of an OP(CP)A system for the generation of high-energy ultrashort mid-IR pulses and application of mid-IR sources in non-linear spectroscopy and for remote atmospheric sensing. Prior building mid-IR OPCPA operating in the vicinity of 6 μm, we concentrated our efforts on the development of femtosecond Ho:YAG chirp pulse amplifier (CPA), which is the first to our knowledge reported femtosecond multi-millijoule 2-μm laser system. The key elements of the CPA system are ring-cavity configuration of the regenerative amplifier, mechanical shaper for pre-compensation of the gain narrowing and Acousto-Optic Programmable Dispersive Filter (Dazzler) for the pre-compensation of the phase shift introduced by the amplification in the presence of neighboring laser transitions. Employing either a carrier envelope phase stable OPA or 2.1-μm fiber oscillator - amplifier as a seeder we generated femtosecond multi-millijoule pulses at kilohertz repetition rate that opens prospects for generation of high energy broadband mid-IR pulses in OP(CP)As.

The developed Ho:YAG laser was employed for both seeding and pumping a cascaded mid-infrared OPA based on potassium titanyl arsenate (KTA) and zinc germanium phosphate (ZGP) nonlinear optical crystals. A crucial part of the OPA is the generation of coherent broad-band seed supporting few-optical cycle pulse duration. The seed in principle can be generated by an independent laser source, however it demands an external electronic synchronization with the main pump laser; therefore we aimed for all optical synchronization of the system. Since it is difficult to produce a seed in the vicinity of either 6-μm (idler wavelength) or 3-μm (signal wavelength) directly from the 2.09-μm radiation, we opted for cascaded seed generation. For the generation of white-light seed 2.09-μm laser radiation was frequency doubled, which due to non-linear conversion resulted in even shorter 1.045-μm pulses; thus, allowing stable white light generation in yttrium aluminum garnet (YAG) crystal in the vicinity of 1.5 μm. Generated 1.5-μm signal pulses were amplified in a KTA crystal pumped by the remaining fraction of the 1.045-μm radiation, which produced idler radiation in the vicinity of 3 μm. The 3-μm pulses further were used as a seed (signal) pulses for a mid-IR ZGP OPA pumped by 2-μm output of the Ho:YAG laser system. Proposed approach enables generation of coherent hundreds micro-joule mid-IR pulses supporting few-cycle pulse duration.

Recent studies showed that filaments ignited by multi-millijoule mid-IR lasers can emit coherent back-propagating radiation, which can be used for remote atmospheric sensing. Here, by employing high energy 4-μm OPCPA system, we demonstrate a proof-of-principle scheme for the detection of atmospheric pollutants in a remotely located small volume in a completely free-space configuration. The key element of the system is an atmospheric nitrogen laser remotely ignited by an ultrashort-pulse mid-IR laser. Inversion population in a filament generated in N_2 environment leads to the emission of narrowband back-propagating sub-nanosecond μJ-level pulses which, in combination with tunable UV pulses, generated in a synchronized OPA, can be used as a stimulated Raman scattering probe for atmospheric gas detection. We have developed a prototype which in laboratory conditions is capable of detecting methane in variable pressure cell with rather high temporal and spectral sensitivity.