Final Report

The results of our research have been published in 11 refereed journal papers with a cumulative impact factor of about 39. One of our papers has been selected for the cover picture of the high-impact (7.727) journal Optica. About 25 conference contributions were presented, out of which 3 were invited. At the time of this report, there are 2 manuscripts under review or in preparation. The submission of manuscripts on our experiments on THz-driven strong-field electron emission and on the measurement of the four-photon absorption coefficient in semiconductors can be expected in the near future.

Highly efficient semiconductor THz sources

The focus of our research was on the realization and investigation of novel semiconductor terahertz (THz) sources. This research was driven by the limitations in scaling the more conventional lithium niobate sources to high energies and field strengths (mainly caused by the large pulse-front tilt angle) and by the demand for compact and efficient high-field THz sources. The combination of infrared pumping at 1.7 μm wavelength and tilting the pump pulse front enabled us to substantially reduce the effect of two- and three-photon pump absorption and the associated free-carrier absorption at THz frequencies in semiconductor nonlinear optical materials, such as ZnTe and GaP. The additional advantage is the straightforward scalability to high THz energies and field strengths, enabled by the small pulse-front tilt angle of about 30° or smaller.

To exploit the potential of semiconductors, we demonstrated a ZnTe contact-grating THz source. This novel technology, proposed by us earlier, enabled the construction of a monolithic THz source, where no external pulse-front tilting optical system is needed. It was possible to work in a collinear pumping geometry. This feature is essential for enabling a straightforward increase of the THz energy by simply increasing the pumped area and for an excellent focusability of the THz beam to achieve highest field strengths. We experimentally demonstrated the focusability of the THz beam to a nearly diffraction-limited spot and an extremely high THz generation efficiency of 0.3%. This efficiency is two orders of magnitude higher than the highest value reported previously from the same material. We published our results in the high-impact journal Optica [**Optica 3 (2016) 1075**]. Our work has been selected for the title page of the journal (Fig. 1).

Fig. 1. Artistic view of the semiconductor contact-grating THz source.

Our simulations showed that the efficiency can be further increased by increasing the material thickness. In order to confirm the possibility of still higher conversion efficiencies we carried out experiments with a conventional pulse-front tilting setup, consisting of an external grating and imaging optics, used to pump a ZnTe prism with a larger average material thickness [**Opt. Express 24 (2016) 23872**]. We demonstrated a THz generation efficiency as high as 0.7%. This is similar to the highest values achieved with $LiNbO_3$ at high pulse energies and more than 200 times higher than the highest value previously reported by others in the same material. The achieved 14 μJ pulse energy is about one order of magnitude larger than any previously reported semiconductor THz source.

We investigated by numerical simulations optimal conditions for the efficient generation of high-field THz pulses by optical rectification in semiconductor nonlinear materials [**IEEE JSTQE 23 (2017) 8501508**]. The simulations included the effect of four-photon absorption on THz generation. The four-photon absorption coefficient, a very important material parameter for the design of THz sources, could be estimated from our THz generation experiments. To our knowledge, this was the first report on four-photon absorption coefficient for ZnTe determined from experiment. Basic design aspects of semiconductor THz sources were discussed, and a practical guideline was given on the selection of the nonlinear material, and the optimization of the crystal length, pump pulse duration, and pump intensity. The dependence of the spectral features and electric field waveforms on the phase-matching frequency and crystal length were analysed. Our work showed that semiconductor THz sources, pumped by infrared lasers or optical parametric amplifiers, can reach similar efficiencies than $LiNbO_3$ sources, but can surpass conventional technology in scalability to extremely high (tens-of-MV/cm) field strengths, as well as in versatility and THz beam quality.

For a more direct measurement of the four-photon absorption coefficient we carried out z-scan measurements in collaboration with University of Bern [**Monoszlai et al., ICOMP 2017 conference**].

Our results on semiconductor sources opened up a new route towards efficient generation of THz pulses with unprecedented energies and field strengths in the low- and mid-frequency (0.1-7 THz) part of the THz spectrum, which can be important for materials science and THz-driven particle acceleration.

Multicycle THz pulses

Also intense multicycle THz pulses are of significant interest for linear and nonlinear THz spectroscopy in materials science and for driving compact THz particle accelerators. For example, waveguide, resonator, or some dielectric structures proposed for electron acceleration can be more efficiently driven by multicycle THz pulses. Therefore, we have investigated by numerical simulations multicycle THz pulse generation in semiconductors [**Nugraha et al., submitted**]. Our study was also motivated by the promising advancement in efficient infrared-driven semiconductor THz-source technology (see above) and a new concept of efficiently producing intensity-modulated infrared pump pulses with flexible shaping capability (see below). Our results show that the semiconductor nonlinear material GaP, pumped at an infrared wavelength of 1.7 μm or longer, can be an efficient and versatile multicycle THz source with up to about 5% conversion efficiency and a tuning range from below 1 THz up to about 7 THz (Fig. 2). For an optimal combination of THz frequency, pump intensity, and crystal length, efficiencies even beyond 10% can be expected. For example, with 40 mJ of intensity-modulated pulse energy it is feasible to produce multicycle THz pulses with >1.5 mJ energy from a GaP contact-grating source of about 1 cm^2 area. The contact-grating technology for pulse-front tilt can ensure an excellent focusability and

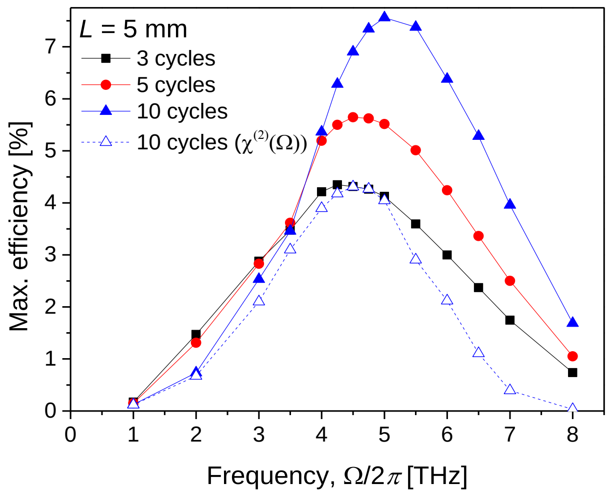

Fig. 2. Predicted THz generation efficiency for multicycle pulses as function of the phase-matching THz frequency in GaP.

high electric field strengths. THz pulses with desired waveforms and fixed or broadband-tuneable carrier frequencies can be produced, covering the low- and significant part of the mid-frequency THz range up to about 7 THz. The accessible frequency range can be extended to fully cover the mid-frequency THz band up to about 20 THz by using organic nonlinear crystals for THz generation.

$LiNbO_3$ THz sources

Besides developing the new semiconductor THz source technology, we also worked on improving THz generation in $LiNbO_3$. We have demonstrated in collaboration with the Technical University of Vienna that the THz generation efficiency can be increased 3× (to about 1%) when the pump pulse duration is increased from 200 fs to 500 fs [**Monoszlai et al., EOS TST 2016 conference**].

Theoretical and experimental work by other groups and us revealed that further increasing the THz pulse energy and field strength beyond state-of-the-art is subject to fundamental limitations, related to the large (~63°) pulse-front-tilt angle required for phase matching. In case of a large beam size, required for a large pulse energy, the interaction length can be nonuniform, resulting in a strongly nonuniform THz beam. Such a beam is disadvantageous for applications and it is difficult to focus it to a spot size close to the diffraction limit, which restricts the achievable peak field strength. Furthermore, the distortions introduced by the imaging optics used for tilting the pump pulse front limit the useful pumped area and the THz pulse energy.

We have proposed possible solutions to these problems. One possibility is to reduce the errors of the imaging system. Calculations were carried out for the optimization of the tilted-pulse-front THz excitation setup containing telescope [**J. Infrared Millim. Te. 38 (2017) 22**]. A hybrid tilted-pulse-front excitation scheme, containing imaging optics and a contact grating, was also investigated by calculations [**Opt. Express 24 (2016) 8156**]. A new concept was proposed, which can provide a uniform interaction length across a large pump beam and is scalable to high energies [**Opt. Express 25 (2017) 29560**]. In this case, the pulse-front tilt is generated by an external grating, and a stair-step echelon is fabricated on the input side of the LN crystal of uniform average thickness. This new concept can lead to extremely strong THz fields as high as 10 MV/cm in the 0.1 to 1.5 THz low-frequency range.

Another work was aiming at the development of an efficient THz source for small and medium (0.1-10 µJ) pump energies to enable linear and nonlinear THz spectroscopic studies at high (100 MHz-100 kHz) repetition rates. A significant >20× increase in efficiency, as compared to that in bulk material, was predicted by proposing the novel concept of an absorption-reduced planar waveguide structure. This scheme acts as waveguide for both the pump and the THz pulses. Simulation results on such a THz source were published [**Appl. Phys. Lett. 107 (2015) 233507**].

Pump sources for THz generation

The demonstrated semiconductor THz sources required cutting-edge pump sources. A unique multi-mJ infrared optical parametric amplification system, developed at the Technical University of Vienna, has been used in these experiments. This amplifier was pumped by a 100-mJ-class high-energy $Yb:CaF_2$ pump laser system [**Opt. Express 24 (2016) 28915**], developed by the same institute in close collaboration with us. This high-energy pump laser is now installed in our laboratory.

In case of phase-matched optical rectification, the THz waveform is determined by the intensity shape of the pump pulse. Thus, for a multicycle THz pulse source, one of the main technical challenges is to create suitably shaped intensity-modulated pump pulses, ideally with a versatile shaping capability. Previously proposed schemes had serious limitations in their pulse-shaping capability. We have proposed an efficient method to generate high-energy infrared pump pulses with a constant intensity modulation period for multicycle THz pulse generation [**Opt. Express 25 (2017) 28258**]. The method is based on the interference of chirped signal and idler pulses generated in an optical parametric amplifier (OPA) driven by chirped pump pulses (dual-chirped OPA, DC-OPA). Easy tuneability of the modulation frequency (which determines the THz frequency) is enabled by introducing a time delay between the pump and the signal pulses. The pulse duration of the intensity-modulated pulse (and hence that of the generated THz pulse) can be set by the chirp of the signal and the pump. An efficiency as high as ~50% was predicted for 40 mJ output pulse energy at a wavelength of 2 µm. Such flexibly shapeable pulse sources, operating near 1.6 µm or 2 µm and pumped by compact Ti:sapphire or Yb-doped lasers, can be ideally suited to drive organic or semiconductor THz sources. The realization of such a flexible pump source is planned in our laboratory, pumped by our high-energy 1-µm laser.

We also made a proposal, based on a detailed numerical study, for a tuneable high-power broadband optical parametric chirped-pulse amplifier system utilizing angular dispersion of the signal in lithium niobate [**Opt. Commun. 370 (2016) 250**]. An optimized double-grating setup can provide the required angular dispersion. Such a system can be suitable for high-power few-cycle pulse generation in the 1.4 to 2.1 µm wavelength range. Calculations predict 16.8-fs (3-cycle) pulses with 13 TW peak power. Further scalability of the scheme towards the 100-TW power level is feasible by using efficient cost-effective diode-pumped solid-state lasers for pumping directly at 1 µm, without second-harmonic generation.

Materials in strong THz fields

A THz pump—THz probe system, driven by the 7-mJ 200-fs pulses of our $Yb:CaF_2$ laser, was designed and built up (Fig. 3). As a unique feature, the setup contained two independent lithium niobate THz sources, enabling measurements near zero delay. The pump THz source delivered up to 10 µJ pulse energy. The THz probe pulses, after transmission through the sample to be investigated, were characterized by electro-optic sampling. This system served as prototype for the commercial-grade THz pump—THz probe system developed in our laboratory for ELI-ALPS. This advanced system contains two cryogenically cooled THz sources

First user samples were investigated with the prototype pump-probe setup, in the frame of a collaboration with the Technical University of Budapest (S. Bordács, I. Kézsmárki), aiming at the investigation of THz-induced phase transition in GaV_4S_8. We plan to continue this study with better quality samples. As it was necessary to concentrate the available human and infrastructure resources on the large-scale research and development project for the ELI-ALPS device, further investigations of materials had to be delayed. The systematic measurements on a variety of samples can be continued at ELI-ALPS after the commissioning of the device in the near future.

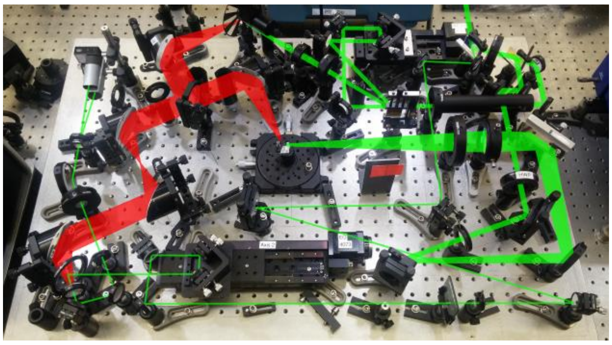

Fig. 3. Photograph of the prototype THz pump—THz probe setup. Red: optical beam path, green: THz beam path.

In a measurement series related to the development of the pump-probe setup, we investigated the influence of a strong-field THz pulse on the output characteristics of the THz source itself. Our near- and far-field measurements of the THz beam profile revealed a strong dependence of the beam size and divergence on the pump intensity [**New J. Phys. 17 (2015) 083041**]. This effect could be explained, with the help of numerical simulations, by the influence of the intense THz field on the optical pump pulse owing to their nonlinear interaction. Our findings can be important for the design of THz sources and application setups including focusing or beam transport lines and measurement setups with variable pump intensity.

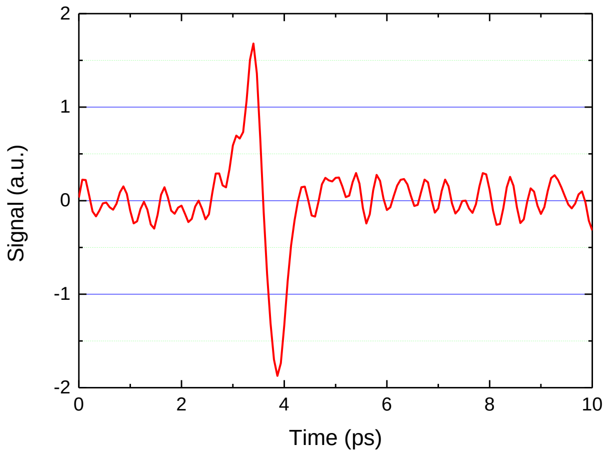

Fig. 4. THz signal measured from an oriented bacteriorhodopsin sample.

In an ongoing measurement series, carried out in collaboration with the Wigner Research Centre for Physics (P. Dombi, Gy. Farkas), THz-induced electron emission was observed from a gold surface irradiated by single-cycle THz pulses with up to 300 kV/cm peak electric field [**Lombosi et al., CLEO/Europe 2015 and ICOMP 2017 conferences**]. The transition regime between multiphoton emission and strong-field emission was studied. The polarization dependence of the electron emission probability has been observed. It was shown that reversing the polarity of the field significantly affects electron emission.

THz spectroscopy of light-harvesting systems

A time-resolved THz spectroscopy setup has been designed and built up to study light-induced ultrafast charge transfer dynamics in light-harvesting biological systems (bacteriorhodopsin, type-I plant photosynthetic centre). Dried thin layers of the purple membrane of bacteriorhodopsin (bR), oriented by an external electric field, were prepared on indium tin oxide substrates from a suspension by Géza Groma et al. (BRC, Szeged). Pump pulses of 515 nm wavelength were produced by second-harmonic generation of the laser pulses. The coherent THz signal from the sample was detected by electro-optic sampling. Specific balanced photodiodes were available in a collaboration with E. Riedle (LMU, München). As an improvement over previous work, we adopted a technique enabling a signal enhancement up to about one order of magnitude [Brunner at al., J. Opt. Soc. Am. B 31 (2014) 904]. With this sensitive setup we were able to detect the extremely weak THz signal from the bR sample (Fig. 4). Our first experimental results were presented by **Polónyi et al.** at the **Workshop "Photonics Meets Biology" (2017)**. Systematic measurements are ongoing. A modification of the setup to detect the longitudinal component of the THz signal in the near field is in preparation, as well as the extension of the studies to type-I plant photosynthetic centre (PSI).