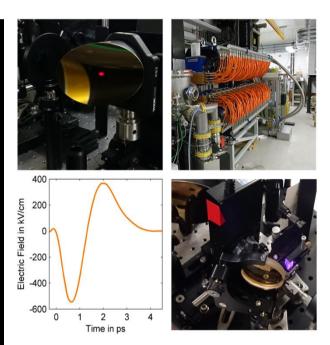
Terahertz science at European XFEL

01–02 June 2017 / European XFEL, Schenefeld, Germany

The interaction of terahertz radiation with matter excites novel states with unique properties. These non-equilibrium states will benefit from ultrafast characterization using the ultrashort and highly brilliant X-ray pulses available at FELs. With this in mind, we are pleased to announce a terahertz workshop, with the aim to find the most scientifically promising strategies to combine terahertz radiation with the unique X-ray pulses generated at European XFEL. The 2-day workshop will be led by 18 presentations from invited international scientists. The first day will explore the scientific motivation for THz - X-ray experiments. The second day will focus on the two main routes to THz generation in the frequency range from 0.1 to 20 THz (3 mm to 15 µm): On the one hand, state of the art laser-based sources; and on the other, undulator sources based on a second, smaller accelerator. Specific aspects will be compatibility with the MHz repetition rate of the European XFEL and novel opportunities for coherent control in the multi-THz regime. We intend to discuss how individual research projects can benefit from a combination of these sources.







Hosted by European XFEL

Organizers

Andreas Scherz (European XFEL, SCS) Robert Carley (European XFEL, SCS) Peter Zalden (European XFEL, FXE)

Program Committee

Robert Carley (European XFEL, SCS) Mikhail Krasilnikov (DESY, PITZ) Max Lederer (European XFEL, WP78) Mikhail Yurkov (DESY) Peter Zalden (European XFEL, FXE)



Programme

1st of June 2	2017		
09:00-10:30 Ge	et-together, Registration, Po	ster session	
10:45-11:00	Thomas Tschentscher (European XFEL)	Welcome Address	
11:00-12:30 Se	ession A: Correlated Materia	ls	
11:00-11:30	Andrea Perruchi (Fer- mi)	TeraFERMI: a superradiant beamline for THz nonlinear studies at the FERMI free-electron-laser facility	4
11:30-12:00	Stefano Bonetti (U. Stockholm)	THz-driven Ultrafast Spin-Lattice Scattering in Metallic Ferro- magnets	5
12:00-12:30	Adrian Cavalieri (CFEL)	Observation of coherent field-driven dynamics at XFELs	6
12:30-14:00 Lu	inch Break		
14:00-15:30 Se	ession B: Various Science C	ases #1	
14:00-14:30	Karsten Holldack (HZB)	Spins and Charges in Molecular-Nano Magnets and Phase- Change Materials Monitored by THz-Coherent Synchrotron Radiation	7
14:30-15:00	Dmitry Turchinovich (U. Duisburg-Essen)	Thermodynamic picture of Terahertz response in graphene	8
15:00-15:30 Co	· · · · · · · · · · · · · · · · · · ·		1
15:30-17:00 Se	ession C: Various Science C	Cases #2	
15:30-16:00	Aaron Lindenberg (Stanford, SLAC)	THz-driven structural dynamics in materials	9
16:00-16:30	Keith Nelson (MIT)	Nonlinear THz spectroscopy of molecules and materials: THz pump, THz-optical-xray probe	10
16:30-17:00	Louis DiMauro (Ohio State U.)	Intense mid-infrared laser interactions with clusters and solids	11
17:30-19:30 Pc	oster Session		÷
19:30 Joint Din	iner		

2nd of June	2017		
09:00-11:00 S	ession D: Laser-based Sour	ces	
09:00-09:30	Max Lederer (European XFEL)	Versatile Optical Laser for Experiments at the European X-ray Free-Electron Laser Facility	12
09:30-10:00	Matthias Hoffmann (LCLS)	THz-pump/X-ray probe experiments at LCLS	13
10:00-10:30	Christoph Hauri (PSI)	Versatile laser-based Terahertz source for strong-field THz physics	12
10:30-11:00	Andrea Cavalleri (MPSD)	Strong-field THz for nonlinear control	16
11:00-11:30 C	offee Break		
11:30-13:00 S	ession E: Undulator-based S	Sources	
11:30-12:00	Mikhail Krasilnikov (PITZ@DESY)	Tunable IR/THz source based on PITZ (-like) accelerator for pump probe experiments at the European XFEL	17
12:00-12:30	Michael Gensch (HZDR, TELBE)	High-Field High-Repetition-Rate superradiant compact THz (Undulator) sources for Selective THz Control of Matter	18
12:30-13:00	Nicola Stojanovic (FLASH)	Tunable High- field THz from 4th generation light sources: THz beamline at FLASH	19
13:00-14:00 L	unch Break		
14:00-15:30 S	ession F: Various Sources		
14:00-14:30	Franz Kärtner (DESY)	Single-Cycle and Multi-Cycle THz Generation with Lithium Niobate	14
14:30-15:00	Andrej Savilov (IAP, RAS, N. Novgorod)	Axial and Phase Stabilization of Short Dense Photo-Injector Electron Bunches as a Way for Spontaneous Coherent THz Emission from These Bunches	20
15:00-15:30	Zhirong Huang (Stan- ford, SLAC)	Generation of high-power, tunable THz radiation from laser interaction with a relativistic electron beam	21
15:30-16:00 C	offee Break		
16:00-17:30 O	pen discussions, Tours		

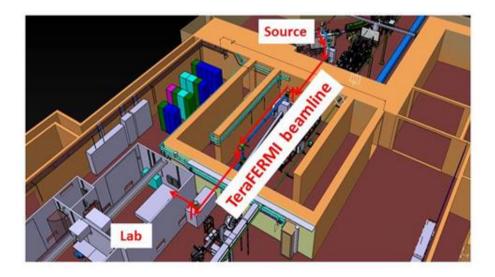


TeraFERMI: a superradiant beamline for THz nonlinear studies at the FERMI free-electron-laser facility

A. Perucchi¹

¹ INSTM Udr Trieste-ST and Elettra - Sincrotrone Trieste S.C.p.A., Area Science Park, I-34012 Trieste, Italy

TeraFERMI is the THz beamline at the FERMI seeded Free-Electron-Laser facility in Trieste. The beamline exploits the short electron bunch structure of the FERMI accelerator to generate THz subpicosecond pulses up to 100 mJ through Coherent Transition Radiation. Thanks to the high peak power achievable, TeraFERMI can be used to address non-linear THz spectroscopy and to achieve coherent control on material's properties. The TeraFERMI beamline will thus extend the free-electron-laser's capabilities in terms of peak power, and ultrafast temporal structure, into the THz spectral range. We illustrate here the beamline design and report on the TeraFERMI beamline status and commissioning results.





THz-driven Ultrafast Spin-Lattice Scattering in Metallic Ferromagnets

S. Bonetti¹

¹ Department of Physics, Stockholm University, Sweden

The interaction between magnetism and light is receiving considerable interest in recent years, after the groundbreaking experiments that showed that ultrashort (~100 fs) infrared light pulses can be used to demagnetize [1] or even switch [2] the magnetization of thin film ferromagnets. This has sparked great excitement towards the possibility of realizing ultrafast magnetic data storage controlled by light. However, to date no clear and commonly accepted understanding of the fundamental physical processes governing the ultrafast magnetization has been reached. Two main mechanisms have been put forward as possible ways of absorbing spin angular momentum: dissipation of spins through the electronic system, via the creation of superdiffusive spin-currents [3], or via the phonon bath, through spin- orbit scattering of Elliot-Yafet type [4]. Although experimental evidence for both mechanisms has been reported, their relative contributions to ultrafast demagnetization remain debated with the accurate modelling of the infrared fs laser-induced highly non-equilibrium state remaining a key obstacle.

In this talk, we present recent experiments where we used strong THz fields, rather than infrared pulses, to excite ultrafast magnetization dynamics in thin film ferromagnets, and probed it with the timeresolved magneto-optical Kerr effect. In one case [6], we compared the response from two films with remarkably different lattice structure: a crystalline Fe film and an amorphous CoFeB film. We observe Landau- Lifshitz-torque magnetization dynamics of comparable magnitude in both systems, but only the amorphous sample shows ultrafast demagnetization caused by the spin-lattice depolarization of the THz-induced ultrafast spin current. Quantitative modelling shows that such spin-lattice scattering events occur on similar time scales than the conventional spin conserving electronic scattering (~30 fs). This is significantly faster that optical laser-induced demagnetization, and THz conductivity measurements point towards the influence of lattice disorder in amorphous CoFeB as the driving force for enhanced spin-lattice scattering. In another experiment, we used the multicycle THz pulses produced at the High-Field High-Repetition-Rate Terahertz facility @ ELBE (TELBE) [7] to drive magnetization dynamics in the amorphous CoFeB sample. Our results show that demagnetization is strongly dependent on the frequency of the multicycle THz pulses, possibly illustrating the relation between chargeand spin-dependent scattering of conduction electrons. Further measurements and modeling are ongoing.

Finally, we will give an overview over the next challenges and opportunities that the use of THz radiation offers towards a deeper understanding of ultrafast magnetism, also in combination with the use of coherent x-ray radiation now available at free electron lasers.

References:

[1] E. Beaurepaire et al., Phys. Rev. Lett. 76, 4250 (1996).

- [2] C. D. Stanciu et al., Phys. Rev. Lett. 99, 047601 (2007).
- [3] M. Battiato et al., Phys. Rev. Lett. 105, 027203 (2010).

[4] B. Koopmans et al., Phys. Rev. Lett. 95, 267207 (2005). B. Koopmans et al., Nature Mater. 9, 259 (2010).

- [5] Z. Jin, et al. Nature Phys. 11, 761 (2015).
- [6] S. Bonetti et al., arXiv:1604.04077
- [7] http://www.hzdr.de/db/Cms?pNid=2609



Observation of coherent field-driven dynamics at XFELs

Adrian Cavalieri^{1,2}

¹ Max Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany ² The Hamburg Centre for Ultrafast Imaging, 22761 Hamburg, Germany

Our capability to observe structural dynamics in materials has progressed from the nanosecond timescale to the hundred-femtosecond timescale with the commissioning of X-ray free-electron lasers. Recently, few-femtosecond and potentially sub-femtosecond X-ray pulses have been delivered at LCLS. Further development will lead to routine delivery of attosecond X-ray pulses at XFELs in the future. However, despite these advances, the temporal resolution that can be achieved remains insufficient to measure vibrational motions and atomic rearrangements of most light atoms, including carbon and oxygen, which move on timescales of only a few femtoseconds. Furthermore, it is not yet possible to couple suitable, waveform or carrier-envelope phase stabilized optical-to-THz frequency light pulses to excite atomic, molecular and condensed matter systems.

To address these challenges, we pioneered the technique of laser-based THz streaking that is used to simultaneously clock the arrival time and measure the temporal profile of the X-ray pulse with tenfemtosecond precision. THz streaking effectively phase-locks a single-cycle THz pulse to the XFEL, allowing it to be used to pump vibrational modes in materials, for example. For even higher timeresolution and to initiate faster dynamics, infrared multi-cycle pulses are required. Yet, single-cycle waveform stabilized IR sources do not typically exist at user facilities and the presence of timing jitter would nevertheless hamper their application.

Therefore, we have also been working on a method of self-calibrated streaking spectroscopy with multi-cycle infrared fields. For proof-of-principle, we observed Auger emission from highly ionized neon atoms at LCLS. In these experiments, the lifetime of the core-hole state was observed with sub-femtosecond precision and the corresponding X-ray pulse that initiated the Auger decay was characterised with sub-femtosecond precision as well.

By exploiting the Gouy phase-shift, our streaking approach can be generalized to allow arbitrary sample systems to be coherently stimulated with infrared radiation. The subsequent dynamics could then be probed using X-ray spectroscopy and scattering with attosecond time-resolution and Angstrom spatial resolution. I will present preliminary results that demonstrate our ability to effectively phase-lock an infrared laser pulse to an XFEL, which is the key to many future experimental applications.



Spins and Charges in Molecular-Nano Magnets and Phase-Change Materials Monitored by THz-Coherent Synchrotron Radiation

K. Holldack¹, J. Nehrkorn³, V. Braggalia², R. Calarco², A. Schnegg¹

¹Helmholtz-Zentrum Berlin (HZB), Albert-Einstein-Str. 15, 12489 Berlin, Germany ²Paul Drude Institut für Festkörperelektronik, Hausvogteiplatz 5-7, 10117 Berlin, Germany ³University of Hamburg, Martin-Luther-King-Platz 6, 20146 Hamburg, Germany

THz spectroscopy in time and frequency domain provides unique information on charge transport and electron spin properties of a large variety of samples ranging from life sciences to semiconductor and spintronic research. Herein, the unique properties of THz-coherent synchrotron radiation are employed for studies in two different material classes. First, we report on high frequency electron-paramagnetic resonance (EPR) spectroscopy of paramagnetic transition-metal ions (TMI) in novel molecular-nanomagnets (MNM). Secondly, optical pump/THz probe experiments expitaxial films of state of the art phase-change materials (PCM) are presented.

EPR employs electron spins as unique probes of the magnetic and electronic properties on the atomistic level. However, in the case of high-spin (HS, S > 1/2) TMIs and lanthanides, the building blocks of MNM, electron-spin couplings are oftentimes too large to be detected by conventional EPR spectrometers. To go beyond this limit, we recently constructed a synchrotron based Frequency-Domain Fourier Transform THz-EPR (FD-FT THz-EPR) spectrometer. This set-up allows for mapping of the EPR transition-energy/magnetic-field landscape, in the range from 0.1 - 5 THz at external magnetic fields from -11 T to 11 T and from room temperature to 1.5 K. FD-FT THz-EPR provides unique opportunities for ultra-broad band EPR detection, as will be demonstrated for integer and non-integer MNM [1-7].

Optical pump/THz probe time domain spectroscopy was carried out on amorphous and crystalline Ge₂Sb₂Te₅ and Sb₂Te₃ PCM as well as PCM super lattices. Very stable, single cycle naturally synchronized THz-pulses were obtained from laser-induced tunable single-cycle pulses of THz-coherent synchrotron radiation emitted from a laser-energy modulated relativistic electron bunch at the BESSY II storage ring. Intriguing ps structural- and carrier dynamics after fs-laser excitation was revealed separately by analyzing 2D time-frequency maps in the 0.3 - 3 THz range. Results are discussed together with fs time resolved soft X-ray diffraction data in view of future THz/X-ray FEL experiments [8-9].

- [1] E. A. Suturina, et al, Inorganic Chemistry, 56 (2017) 3102-3118.
- [2] A. Schnegg, Very-high-frequency EPR, EMAGRES, 6 (2017) 115-131.
- [3] L. Bucinsky et al., Inorganic Chemistry, 56 (2017) 4752-4769.
- [4] X. Feng, et al., Chemical Communications, 52 (2016) 12905-12908.
- [5] D. Pinkowicz et al., Journal of the American Chemical Society, 137 (2015) 14406-14422.
- [6] J. Nehrkorn et al, Journal of Physical Chemistry B, 119 (2015) 13816-13824.
- [7] J. Nehrkorn, A. Schnegg, K. Holldack, S. Stoll, Physical Review Letters, 114 (2015).
- [8] V. Bragaglia et al., Nature Scientific Reports, 6, 28560 (2016).
- [9] V. Bragaglia, A. Schnegg, R. Calarco, K. Holldack, Applied Physics Letters, 109 (2016) 141903.



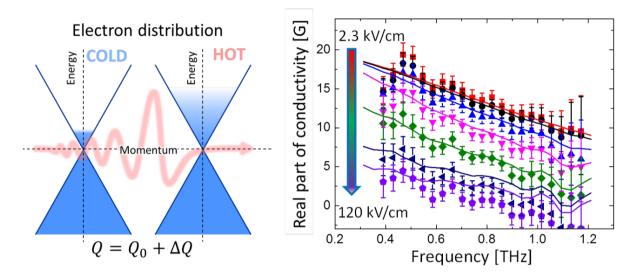
Thermodynamic picture of terahertz response in graphene

Dmitry Turchinovich^{1,2}

¹ Fakultät für Physik, Universität Duisburg-Essen, 47057 Duisburg, Germany

² Max Planck Institute for Polymer Research, 55128 Mainz, Germany

In this presentation we will show, that the terahertz linear, non-linear and photo-conductivity of graphene [1–3] can be accurately described within a simple picture, in which the free carrier population of graphene act as thermalized electron gas in or out of equilibrium with graphene lattice.



Within this simple thermodynamic picture, the electron population quasi-instantaneously increases its temperature by absorbing the energy of driving THz electric field or incident photon, and at the same time cools down via a time-retarded, few picosecond-long process of phonon emission. The asymmetry in electron heating and cooling dynamics leads to heat accumulation in the electron population of graphene, concomitantly lowering the chemical potential for hotter electrons, and thereby reducing the intraband conductivity of graphene – an effect crucially important for understanding of ultrafast graphene transistors and photodetectors.

References:

- Z. Mics, K.-J. Tielrooij, K. Parvez, S. A. Jensen, I. Ivanov, X. Feng, K. Müllen, M. Bonn, and D. Turchinovich, "Thermodynamic picture of ultrafast charge transport in graphene," Nat. Commun. 6, 7655 (2015).
- S. A. Jensen, Z. Mics, I. Ivanov, H. S. Varol, D. Turchinovich, F. H. L. Koppens, M. Bonn, and K. J. Tielrooij, "Competing Ultrafast Energy Relaxation Pathways in Photoexcited Graphene," Nano Lett. 14, 5839–5845 (2014).
- 3. I. Ivanov, M. Bonn, Z. Mics, and D. Turchinovich, "Perspective on terahertz spectroscopy of graphene," EPL Europhys. Lett. 111, 67001 (2015).



THz-driven structural dynamics in materials

Aaron M. Lindenberg^{1,2}

 ¹ Department of Materials Science and Engineering, Stanford University, Stanford, CA 94305, United States
 ² PULSE institute, SLAC National Accelerator Laboratory, Menlo Park, CA 94025, United States

I will describe recent experiments using single-cycle THz fields as an all-optical bias to enable measurements of ultrafast electric-field-driven processes in materials. These experiments have been coupled to a variety of optical and x-ray probes and I will describe recent efforts probing phase-change and ferroelectric materials and future related opportunities. Finally I will provide an update on recent complementary developments using ultrafast electron diffraction as a probe of THz-driven dynamical responses in materials.



Nonlinear THz spectroscopy of molecules and materials: THz pump, THzoptical-xray probe

Keith Nelson¹

¹ Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

Ever-improving methods for tabletop generation of strong THz fields are yielding an ever-widening range of THz-induced nonlinear responses being explored. Three sets of results will be reviewed: 2D THz spectroscopy involving either the THz electric magnetic fields; THz-induced electroluminescence and gigantic Stark shifts probed in the optical regime; and THz-induced phase transitions monitored through ultrafast x-ray diffraction. The results illustrate new prospects for THz spectroscopy and THz control of molecular and material behavior.



Intense mid-infrared laser interactions with clusters and solids

Louis F. DiMauro

The Ohio State University, Columbus, OH 43210 USA

An intense laser-plasma interaction generates bright, energetic bursts of electrons, ions and photons that have been utilized to enable a variety of applications. Nanoplasmas, formed from nanometer-sized clusters under intense field conditions, are distinguishable from conventional plasmas from bulk solids or gases since they can absorb a large amount of laser energy. In this talk, we utilize the wavelength (λ) tuning of an intense mid-infrared source to expose and distinguish different cluster mechanisms. This unexplored territory of nanoplasma physics harnesses the l²-scaling of the ponderomotive energy to impact the electron dynamics, which in turn has profound ramifications on the collisional heating.

In a second topic, we describe harmonic generation from a ZnO crystal created by a mid-infrared pulse. The study focusses on the influence of direct electron seeding of the conduction band by a pump pulse on the harmonic generation processes.



Versatile Optical Laser for Experiments at the European X-ray Free-Electron Laser Facility

Max. J. Lederer, Guido Palmer, Moritz Emons

European XFEL GmbH, Holzkoppel 4, 22869 Schenefeld, Germany

The European XFEL will start its operation in 2017, enabling innovative atomic-scale measurements at high repetition rates and with femtosecond time resolution. Helping to facilitate such experiments are highly specific, synchronized, ultrafast optical lasers to excite specimens before they are probed with x-rays (pump-probe). A development project for such a laser source aimed at the MHz / milijoule / femtosecond performance level, which is substantially outside the realm of commercial lasers.

Adapted to the temporal x-ray emission pattern of the facility, the pump-probe laser provides 10 Hz bursts of up to 600 µs duration with intra-burst pulse frequencies as high as 4.5 MHz. In this mode, we have generated pulses shorter than 15 fs at 350 W average power during the burst from a twostage non-collinear parametric amplifier (NOPA). This is, to the best of our knowledge, the highest power to date of a few-cycle laser operating at a center wavelength of 800 nm. Important for experimental flexibility, the laser can be configured in various unique ways, enabling, e.g., energy scaling to >3 mJ per pulse through a frequency change down to 100 kHz and the generation of nearly transform limited pulses between 15 fs and 300 fs. In addition to the 800 nm femtosecond beam line, a synchronized long pulse (0.8 ps) 1030 nm beam can be utilized, offering up to 4 kW burst average power, i.e. up to 40 mJ per pulse at 100 kHz. Efficient nonlinear wavelength conversion and tuning through intrinsic and external means further enhance the capabilities of the laser. Finally, first experiments using the tilted phase-front 0.8ps / 1030nm beam line for optical rectification in LiNbO₃ were performed to generate high energy THz pulses.

We will present the main features of the prototype laser with first results of conversion experiments and give account of the status of laser installation at SASE 1.



THz-pump/X-ray probe experiments at LCLS

Matthias C. Hoffmann¹

¹Linac Coherent Light Source, SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, USA; hoffmann@slac.stanford.edu

With the availability of the first generation of x-ray free electron lasers, pump-probe measurements with femtosecond resolution and high brilliance are now possible. For condensed matter systems, a wealth of modes in the mid-infrared (MIR) and terahertz (THz) regime determine the physics, such that targeted excitation with ultrashort pulses at long wavelength becomes an important tool. Since THz and MIR pulses usually have photon energies much below band gap, parasitic excitation pathways are eliminated compared to experiments with conventional femtosecond laser wavelengths. Additionally, single or few-cycle THz pulses offer the capability to exert control over matter with the electric or magnetic field component of the radiation, while probing the sample response on a subcycle time scale.

We will discuss the methodology of THzpump/X-ray probe experiments at the Linac Coherent Light Source (LCLS) and present recent examples from atomic and condensed matter physics at LCLS. We will highlight recent results from experiments at the XPP beam line, where hard x-ray pulses were used to probe the nonlinear dynamics of the crystal lattice when infrared active phonon modes are excited resonantly with THz pulses. Figure 1 shows the response of (2 -2 3) Bragg peak in SrTiO₃ when the soft mode phonon at ~1.5 THz is pumped with THz pulses at 1 MV/cm field strength. At these high pump fluences it is possible to transfer energy from the fundamental soft mode into higher frequency phonon modes through the large anharmonicity of the soft mode potential. This example demonstrates that coherent control of the lattice is possible with THz pulses through nonlinear phonon processes. This technique may lead to a large number of applications in ferroelectrics, multiferroics or correlated electron material systems where the electronic band structure is closely tied to the

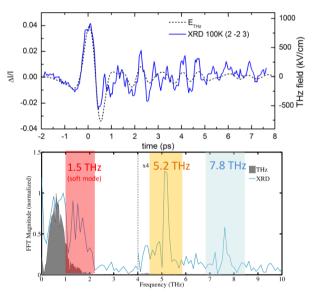


Figure 1: upper panel: X-ray diffraction intensity change of (2 -2 3) Bragg peak in SrTiO₃ at 100K (blue) overlaid with the THz pump electric field (dotted line). Lower panel: Fourier transform reveals energy transfer from the low frequency soft mode phonon at 1.5 THz into the higher phonon modes at 5.2 and 7.8 THz through nonlinear phonon dynamics mediated by the large anharmonicity of the soft mode potential.

underlying lattice configuration and small changes in atomic position can lead to phase transitions. We conclude with an outlook of future developments both in laser-based THz sources and future experiments at LCLS and other FEL light sources.



Versatile laser-based Terahertz source for strong-field THz physics

Christoph P. Hauri

SwissFEL, Paul Scherrer Institute, Switzerland

The production of bright Terahertz radiation with capabilities of tuning the spectral bandwidth and central frequency in the THz gap (0.1-10 THz) has been a formidable challenge in the past.

Here we present recent progress towards the realization of a versatile THz source based on optical rectification of a femtosecond laser pulse in organic crystals. We discuss the production of intense single-cycle pulses reaching atomically strong fields close to 1 V/Å as well as the tailoring of narrowband radiation with a minimum spectral bandwidth of 2.5% (Fig. 1). We discuss the implementation of this type of Terahertz source at complex user facilities like FELs which require the THz source to be robust, simple, easy to operate and to be located in close vicinity of the experimental stations. We finally will present first strong-field THz experiments [3] corroborating the potential of this source for future THz pump, x-ray probe experiments at FELs.

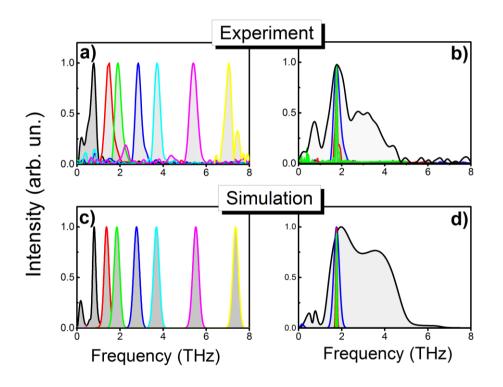


Fig. 1. Frequency and bandwidth tunability of strong-filed THz pulses.

[1] M. Shalaby and C.P. Hauri, "Demonstration of a low-frequency three-dimensional terahertz bullet with extreme brightness," Nature Commun. 6, 5976 (2015) doi:10.1038/ncomms6976

[2] C. Vicario, C.P. Hauri et al. "Generation of strong-field spectrally-tunable Terahertz pulses," Nature Commun. accepted (2017)

[3] C. Vicario, M. Shalaby and C.P. Hauri, "Sub-cycle extreme nonlinearities in GaP induced by an ultrastrong Terahertz field," Phys. Rev. Lett. 118,083901 (2017)

[4] M. Shalaby, C. Vicario and C.P. Hauri, "Extreme nonlinear terahertz electro-optics in diamond for ultrafast pulse switching," APL Photonics 2, 036106 (2017)



Single-Cycle and Multi-Cycle THz Generation with Lithium Niobate

F. X. Kärtner

Center for Free-Electron Laser Science, DESY, Hamburg, Germany, Physics Department and The Hamburg Center for Ultrafast Imaging, University of Hamburg, Hamburg, Germany franz.kaertner@cfel.de

We report on experimental results and numerical studies to produce intense single-cycle and multicycle terahertz pulses via optical rectification or difference frequency generation in Lithium Niobate using the tilted-pulse front technique or periodic poling.

Summary

Strong-field THz physics is a very active area of research with implications from fundamental physics to technology, chemistry and biology. We are especially interested in the conctruction of a terahertz (THz) driven linear accelerator to utilize the high electric fields and field gradients achievable with ultrashort and intense THz pulses to generate low emittance electron beams that can be compressed via velocity compression close to femtosecond duration. Then even pC-electron bunches do show high enough peak current to drive a Free-Electron Laser Process to produce sub-femtosecond hard X-ray pulses from potentially very compact machines. Ideally, the necessary high energy THz pulses on the order of 1-10 mJ single- and multi-cycle can be directly generated with high energy optical pulses via difference frequency generation (DFG) and optical rectification (which is a special case of DFG). To accelerate bunches with a charger higher than 1-10 pC, we prefer THz wavelengths in the range 0.1 – 0.5 THz. In this wavelength region Lithium Niobate is the best choice for efficient optical to THz conversion. Over the last years, we investigated THz generation with Lithium Niobate systematically. Cooling of the material to cryogenic temperatures reduces the THz absorption significantly by a factor of 3-5 in the frequency range below 0.5 THz with the corresponding increase in optical to THz conversion efficiency.¹ For the generation of single-cycle THz pulses, optical to THz conversion efficiencies up to 1% have been demonstrated to date.² The observed efficiencies are limited by pulse break-up in Lithium Niobate which results from the large pulse front-tilt angle necessary to achieve phase matching in Lithium Niobate in combination with the necessary cascading to achieve high conversion efficiency. This was shown by numerical modelling.^{4,5}

These limitations of non-collinear approaches can be overcome when using periodically poled Lithium Niobate (PPLN) for multi-cycle generation. Here, all waves are interacting collinearly and limitations are only due to the THz absorption and damage threshold of the material. Due to the enhanced interaction length, higher conversion efficiencies are possible, limited only by the damage threshold achieveable in the material. Multi-cycle THz is generated in multi-period PPLN using either two or multiple single frequency lines and cascaded DFG or cascaded optical parameteric Generation^{6,7}. The later may be easier to implement for high energy THz pulse generation due to the easier construction of a single-frequency high energy pulsed laser system. Several percent of optical to THz conversion efficiency seems to be possible by these techniques.⁸

References

- [1] F. X. Kärtner et al. doi:10.1016/j.nima.2016.02.080.
- [2] X. Wu, Ch. Zhou, W. R. Huang, F. Ahr and F. X. Kärtner, Opt. Exp. 23:(23) 29729-29737 (2015).
- [3] S.-H. Huang, E. Granados, W. R. Huang, K.-H. Hong, L. E. Zapata and F. X. Kärtner, Opt. Lett. 38:(5), 796-798 (2013).
- [4] K. Ravi, W. R. Huang, S. Carbajo, X. Wu and F. X. Kärtner, Opt. Express 22, 20239 (2014).
- [5] K. Ravi, W. R. Huang, S. Carbajo, E. A. Nanni, D. Schimpf, E. Ippen and F. X. Kärtner, " Opt. Exp. 23:(4) pp. 5253 (2015).
- [6] K. Ravi, D. Schimpf and F. X. Kärtner, Opt. Lett. 42:(22), p. 25582 (2016).
- [7] K. Ravi, M. Hemmer, G. Cirmi, F. Reichert, D. N. Schimpf, O. D. Mücke and F.X. Kärtner, Opt. Lett. 41, pp 3806 (2016).

[8] F. Ahr, S. W. Jolly, N. H. Matlis, S. Carbajo. T. Kroh, K. Ravi, D. N. Schimpf, J. Schulte, H. Ishizuki, T. Taira, A. Maier and F. X. Kärtner, accepted in Optics Letters.



Strong-field THz for nonlinear control

Andrea Cavalleri

Max Planck Institute for the Structure and Dynamics of Matter, Hamburg GERMANY

In this talk, I will discuss how coherent electromagnetic radiation at infrared and TeraHerz frequencies can be used to drive collective excitations nonlinearly, and to create new non-equilibrium states of interest. I will discuss experiments in which phonons, plasmons and magnetic excitations are driven, and will discuss how femtosecond x-ray beams from free electron lasers are integral to these studies, to image structures during these non-equilibrium processes.



Tunable IR/THz source based on PITZ (-like) accelerator for pump probe experiments at the European XFEL

Mikhail Krasilnikov

(DESY) for the PITZ Team

The Photo Injector Test facility at DESY, Zeuthen site (PITZ), develops high brightness electron sources for modern free electron lasers like FLASH and the European XFEL. High performance of the photo injector has been demonstrated at PITZ. Currently extensive studies on even further improving the photo injector stability, reliability and performance are ongoing. Besides this, a concept for a source of powerful, coherent and tunable IR/THz radiation for pump-probe experiments at the European XFEL based on a linear accelerator similar to that in operation at PITZ is being developed. It consists of an RF gun and a warm accelerating section (energy up to 25 MeV). The time structure of the THz source and x-ray FEL are perfectly matched since the THz source is based on the same technology as the injector of the European XFEL and they will both be synchronized to the same master clock. The radiation is assumed to be generated in an APPLE-II type undulator, thus providing polarization control. Radiation with wavelength between 20 and 100 um is generated using the SASE FEL mechanism by applying 4 nC electron bunches. Start-to-end simulations using ASTRA and GE-NESIS1.3 codes yield radiation pulse energies of up to the mJ-level. Feasibility of generating such high charge electron bunches has been proven recently at PITZ by the experimental characterization of 4 nC bunches. For comparison SASE FEL simulations based on the experimentally obtained electron beam properties have been performed and show similar results. Several possibilities to improve the stability of the generated THz pulses (e.g. pre-bunched electron beam from the photo injector) as well as more sophisticated schemes for THz pulse length control (e.g. single cycle production using a pre-bunched chirped electron beam in a tapered undulator) are also under study at PITZ now.



High-Field High-Repetition-Rate superradiant compact THz (Undulator) sources for Selective THz Control of Matter

M. Gensch

HZDR, Bautzner Landstrasse 400, 01328 Dresden

In this contribution the design, performance and scientific case of TELBE [1], the worldwide first THz user facility based on superradiant THz emission from a compact quasi-cw superconducting radiofrequency driven linear accelerator, are presented. Based on example benchmark experiments from the 1st year of friendly user operation, the achievable time resolution and dynamic range of multicolor ultra-fast experiments is presented. As will be shown novel pulse-resolved detection schemes operating at high repetition rates in the few 100 kHz regime allow to achieve time-resolution of better than 30 fs and a dynamic range of better than 10^6 [2]. The final design parameters aim at providing pulse energies of beyond 100 µJ at flexible repetition rates of up to 100 kHz in the lower THz frequency range (< 3 THz). Fundamental limits of the achievable frequency range, given by the moderate beam energy of 40 MeV available from the compact ELBE accelerator, will be discussed.

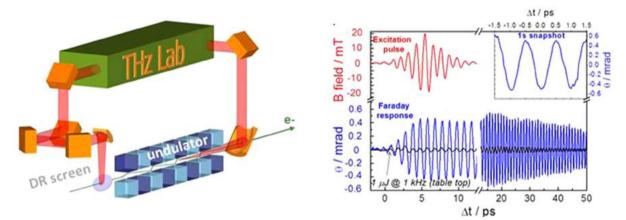


Figure: Arrangement of the superradiant THz sources at TELBE (left) and transient Faraday measurement of a selectively excited spin wave in NiO [1]. Note that the 2 THz sources (diffraction radiator – single cycle and undulator – multi cycle) can be operated in parallel without any observable degradation of the THz pulse properties.

[1] B. Green et al., High-Field High-Repetition-Rate Sources for the Coherent THz Control of Matter, Sci. Rep. 6 (2016), 22256.

[2] S. Kovalev et al., Probing ultra-fast processes with high dynamic range at 4th-generation light sources: Arrival time and intensity binning at unprecedented repetition rates, Struct. Dyn. **4** (2017), 10159.



Tunable High- field THz from 4th generation light sources: THz beamline at FLASH

N. Stojanovic

Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany nikola.stojanovic@desy.de

Last decade witnessed the rise of new type of accelerator light sources based on linear accelerators. Like synchrotron sources, these large-scale facilities have the appeal of their broad spectral range (from THz to X-rays) as well as tunability. However, their advantage over synchrotrons lays in the ability to generate ultrashort pulses with peak intensities many orders of magnitude higher.

In THz range, the ability of 4th generation sources to generate pulses with electric field strength in the GV/m range, enabled number of exciting experiments, especially in the field of non-linear THz spectroscopy and THz control experiments. A variety of genesis concepts allows shaping the THz pulses from single-cycle (broadband) pulses to many-cycle narrow-bandwidth pulses and polarizations ranging from radial to linear. The main advantage of accelerator-based THz originates from the fact that the THz generation process does not take place in a medium but in the accelerator vacuum, and therefore the THz pulse energy can be scaled up much easier than in case of the table top sources available today. In addition, it has been demonstrated recently that coherent THz radiation can be generated along femtosecond X-ray pulses in 4th Generation X-ray Light sources such as FLASH [1, 2, 3] and LCLS [4]. This opens up the exciting opportunities for naturally synchronized THz pulse for the THz facility at FLASH/DESY and experimental opportunities and challenges are discussed with the example of recent experiments.

References

[1] M. Gensch et. al., New infrared undulator beamline at FLASH, Infrared Phys. Technol. 51 (2008), 423.

[2] U. Fruehling et. al., Single-Shot THz-field-driven X-ray streak camera, Nat. Photon. 3 (2009), 523.
[3] F. Tavella, N. Stojanovic, G. Geloni, M. Gensch, Few-Femtosecond timing at Fourth-Generation X-ray Light sources, Nat. Photon. 5 (2011), 162.

[4] D. Daranciang et. al., Single-cycle terahertz pulses with > 0.2 V/angstrom field amplitudes via coherent transition radiation, Appl. Phys. Lett. 99 (2011), 141117.



Axial and Phase Stabilization of Short Dense Photo-Injector Electron Bunches as a Way for Spontaneous Coherent THz Emission from These Bunches

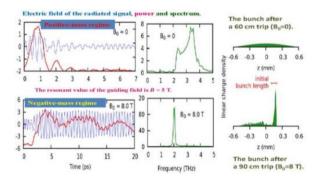
Ilya V. Bandurkin¹, Vladimir L. Bratman^{1,2}, Ilya S. Kurakin³, Yulia S. Oparina³, Nezah Balal², Yury Lurie², Andrey V. Savilov^{1,3}

¹Institute of Applied Physics, Nizhny Novgorod, Russia ²Ariel University, Ariel, Israel

³Lobachevsky State University of Nizhny Novgorod, Russia

I. UNDULATOR EMISSION IN THE NEGATIVE-MASS RE-GIME

Laser-driven photo-injectors are capable to form very compact and dense electron bunches with a particles energy of 3-6 MeV, picosecond and subpicosecond duration, and charge of the order of 1 nC. When moving in the periodic field of the undulator with a period of a few centimeters, such bunches can generate coherent radiation in the near-terahertz range. The power and duration of such a generation is limited by spreading of the electron bunch under the action of the Coulomb repulsion, which under normal conditions does not allow for the effective implementation of such a scheme. However, providing of a special configuration of the magnetic field, when a so called regime of negative electron mass is realized[1,2], can significantly slow down the Coulomb repulsion. Such a regime is realized, when electrons move in a combination of periodic undulator field and relatively strong homogeneous axial magnetic field, and the cyclotron frequency corresponding to the axial field is slightly higher than the undulator bounce-frequency. In this situation, an increase/decrease in the electron energy leads to a decrease/increase in its axial velocity. This is due to the dependence of the undulator (transverse) electron velocity on the cyclotron frequency (and, therefore, on the energy). This means that the Coulomb electric field leads not to repulsion but to attraction of electrons.



One can propose a design in which the electron bunch with the particles energy of 5.5 MeV (mass factor of 12) scatters the undulator field with a period of 2.5 cm into the wave with a frequency of about 2 THz. The negative mass regime is realized at the axial field from the range near 8 T and the undulator field amplitude of about 0.2 T. The efficiency of this process (the share of the initial electron energy passed to the radiated wave) can be as high as 20%.

II. NEGATIVE-MASS COMPRESSION

Effective coherent radiation is possible if the axial bunch size is shorter than the radiation wavelength. If the duration of the initial bunch exceeds this wave period, it should be precompressed. Simulation show that the compression can be based on the negative mass regime. This may be either a system with a constant undulator period, in which the output power gradually increases as the compression of the bunch evolves, or a system with a varied undulator period. In the latter case, in the first section, the period of the undulator corresponds to a small negative mass of the particles, so that a high rate of the bunch compression is achieved [3].

III. SUPER-RADIATIVE SELF-COMPRESSION

A disadvantage of the negative-mass undulator is the requirement of a strong axial magnetic field. An alternative method for the bunch compression can be the super-radiation of a long-wavelength wave in additional (auxiliary) long-period undulator [4]. If the bunch is short enough, then the bunch front is placed close to the maximum of the decelerating phase of the radiated wave, whereas the tail is placed close to the "zero" wave field. This effect can be used for creation of a "bicolor" THz source based on the spontaneous emission from a short bunch, so that the super-radiation of the auxiliary long-wavelength wave is used to compress the bunch down to a size shorter than the wavelength of the operation shortwavelength wave.

IV. CYCLOTRON SUPER-RADIATION

On more approach can be based on the use of the spontaneous cyclotron emission from the short electron bunch [5]. If the emission is carried out in the super-radiation regime (when the axial electron velocity coincides with the wave group velocity), then the Coulomb repulsion leads to an increase in the axial bunch size, but this does not result in an increase of the phase size of the bunch with respect to the radiation wave and, therefore, does not stop the spontaneous coherent radiation.

[1] N. Balal, I. V. Bandurkin, V. L. Bratman, E. Magory, and A. V. Savilov. "Negative-mass mitigation of Coulomb repulsion for terahertz undulator radiation of electron bunches." Appl. Phys. Let. 107, 163505 (2015).

[2] Yu. Lurie, V.L. Bratman, and A. V. Savilov. "Energy enhancement and spectrum narrowing in terahertz electron sources due to negative mass instability." Phys. Rev. AB 19, 050704 (2016).

[3] I. V. Bandurkin, I. S. Kurakin, and A. V. Savilov. "Compression of a photoinjector electron bunch in the negative-mass undulator." Phys. Rev. AB 20, 020704 (2017) (2017).

[4] I. V. Bandurkin, Yu. S. Opatina, and A. V. Savilov. "Super-Radiative Self-Compression of Photo-Injector Electron Bunches." Submitted to Applied Physics Letters, (2017).

[5] Yu. S. Oparina, and A. V. Savilov. "Spontaneous Coherent Cyclotron Radiation from Short Dense Electron Bunches." Submitted to Physical Review AB (2017).

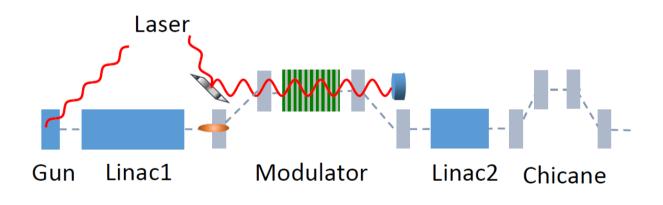


Generation of high-power, tunable THz radiation from laser interaction with a relativistic electron beam

Zhirong Huang

SLAC, Stanford University

We propose a method based on the slice energy spread modulation to generate strong subpicoseond density bunching in high-intensity relativistic electron beams [1]. A laser pulse with periodic intensity envelope is used to modulate the slice energy spread of the electron beam, which can then be converted into density modulation after a dispersive section. It is found that the double-horn slice energy distribution of the electron beam induced by the laser modulation is very effective to increase the density bunching. Since the modulation is performed on a relativistic electron beam, the process does not suffer from strong space charge force or coupling between phase spaces, so that it is straightforward to preserve the beam quality for terahertz (THz) radiation and other applications. We show in both theory and simulations that the tunable radiation from the beam can cover the frequency range of 1-20 THz with high power and narrow-band spectra, and discuss recent experimental studies of this scheme.

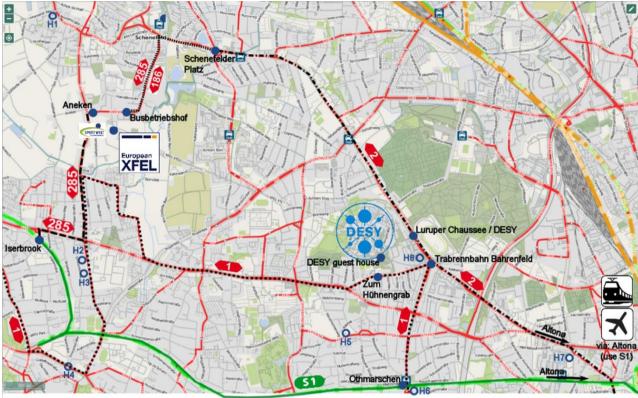


References: [1] Z. Zhang et al., Phys. Rev. Accel. Beams 20, 050701 (2017).



A Traveller's Guide to

European XFEL



© OpenStreetMap

O H1	Hotel Klövensteen
O H2	Hotel Blankenese
O H3	Hotel Hesse
O H4	Hotel Behrmann
O H5	Landhaus Flottbek
O H6	Hotel Schmidt
O H7	Gastwerk Hotel Hamburg
O H8	Hotel Mercure

Hauptstraße 83, 22869 Schenefeld Schenefelder Landstraße 164, 22589 Hamburg Schenefelder Landstraße 139, 22589 Hamburg Elbchaussee 528, 22587 Hamburg Baron-Voght-Straße 179, 22607 Hamburg Reventlowstraße 60, 22605 Hamburg Beim Alten Gaswerk 3, 22761 Hamburg Albert-Einstein-Ring 2, 22761 Hamburg

How to get to XFEL

Bus from DESY Guesthouse / Hotel Mercure

From *Luruper Chausse* board Bus **(2)** direction *Schenefeld, Achterndiek* travel 8 stops (approx. 12 minutes) until *Schenefelder Platz*. At *Schenefelder Platz* change to Bus **(186)** (direction *Schenefeld Mitte*). Travel 5 stops (approx. 5 min) to *Busbetriebshof*. Walk along *Osterbrooksweg* the direction the bus drove and turn the next street left onto Holzkoppel. Go straight until you reach the XFEL Gate.



Alternatively, take the Bus **1** at *Trabrennbahn* or *Zum Hünengrab* (DESY) in direction *Rissen*. Get out at *Holtbarg* and change to bus **285** direction *Pinneberg* and get out at *Aneken*. Walk along Osterbrooksweg in the same direction as the bus left but turn the next right on to Holzkoppel. Go straight until you reach the XFEL gate.

Malking from Hotel Klövensteen (ca. 25 min)

Follow Hauptstraße right hand side till crossing Blankeneser Chaussee. Turn right and follow Blankeneser Chaussee till Osterbrooksweg (ca. 1,5 km). Turn left in Osterbrooksweg till Holzkoppel. turn right into Holzkoppel. European XFEL is at the end of the street.

S-Bahn from station ALTONA

Take S-Bahn **S1** from station *Altona* in direction *Wedel* (goes every 20 min) travel until *Iserbrook* (S1 will change direction at station *Blankenese*, don't worry you are still going the right way). At *Iserbrook* walk down the stairs to the bus stop on the right. Take Bus **285** direction *Pinneberg* travel 2 stops and get out at *Aneken*. Walk along Osterbrooksweg in the same direction as the bus left but turn the next right on to Holzkoppel. Go straight until you reach the XFEL gate.

XFEL -> Hotel Klövensteen

Bus (ca 15 min)

Walk along *Holzkoppel* until *Osterbrooksweg*, turn right and walk to bus stop *Busbetriebshof*. Take bus **285** in direction *Bf. Pinneberg*, travel 6 stops to *Uetersener Weg*. Walk a bit back until you reach the traffic light. You should see the hotel across the roard (Peter's Bistro).

Alternatively, walk along Holzkoppel until Osterbrooksweg, turn left and walk to the bus stop *Aneken*. Take bus **186** direction *Schenefeld Mitte*, and travel 3 stops to *Schenefeld Mitte*. Here you can change to bus **285** in direction *Bf. Pinneberg* and travel 1 stop until *Uetersener Weg*, or you can walk back to *Hauptstraße* turn right and follow *Hauptstraße* until you reach the Hotel. Walking from *Schenefeld Mitte* to Hotel Klövensteen should take approx. 6 min.

🕈 Walking (ca. 25 min)

Walk along *Holzkoppel* until *Osterbrooksweg*, turn left and go straight until you reach a traffic light, at the traffic light turn right onto *Blankeneser Chaussee*. Follow *Blankeneser Chaussee* for 1.1 km until you reach *Hauptstraße*, then turn left and follow *Hauptstraße* until you reach the Hotel (400m).

XFEL → DESY Guesthouse/Hotel Mercure

Walk along *Holzkoppel* until *Osterbrooksweg*, turn right and walk to bus stop *Busbetriebshof*. Take bus number **186** direction *S Othmarschen* travel 4 stops until *Parkstieg*. Change to bus **2** direction *Bf Altona* travel 9 stops until *Luruper Chaussee (DESY)*. Walk for approx. 200m in the direction the bus left until you reach Hotel Mercure. Or for DESY Guesthouse after you reach *Luruper Chaussee (DESY)* walk back approx. 100m until you reach the DESY side gate entrance.

Alternatively, Walk along Holzkoppel until Osterbrooksweg, turn right and walk to bus stop Busbe-



triebshof. Take bus number **(285)** direction *S Iserbrook*, travel for 2 stops until *Holtbarg*, change to bus **(1)** direction *Bf. Altona*, travel for 12 Stops until *Bahrenfeld, Trabrennbahn*, walk back to the traffic light and head straight for approx. 300m on *Luruper Chaussee* until you reach Hotel Mercure. Or for DESY Guesthouse get out 2 stops earlier at *Zum Hünengrab (DESY)*, and walk 100m to the DESY main Entrance.

XFEL → Hamburg Airport / Main Train station / Altona

Walk along *Holzkoppel* until *Osterbrooksweg*, turn right and walk to bus stop *Busbetriebshof*. Take bus **285** in direction S *Iserbrook* (goes every 20 min). Travel 3 stops to station *S Iserbrook*, walk up the stairs to the trains and take the S-Bahn **51** to *Altona* (5 stops) or *Hauptbahnhof* (main train station) (11 stops), or continue on to Hamburg Airport (55 min, 22 stops). Note: for Hamburg airport please board one of the **last** three trains of six available (at S Bahn *Iserbrook* or at *Blankenese*), as only these go straight to the airport.

WiFi access at European XFEL

1) Eduroam

For guests, whose home institutes are participating in the project eduroam, we offer this wireless guest network connection. For registered user this is a quick and easy way to obtain a secure access to the Internet without any further registration.

2) XFEL-Guest

Registration and Log in Procedure:

- 1. Enable Wi-Fi on your device and Connect to the XFEL-Guest network. You will be redirected to your browser, if not...
- Open your browser and call up any unencrypted webpage (http://... NOT https://...), except desy.de (your browser will display the DESY homepage even if you have not completed the registration for the XFEL-Guest network).
- 3. You should have been redirected to the XFEL-Guest portal (<u>https://guestnet-portal.desy.de</u>).
- 4. Click on "Receive your credentials by E-mail".
- 5. Fill in your **First** and **Last name** and **Email**, accept the DESY data protection provisions and click on Register.
- 6. You now have **5 minutes access** to retrieve your *User Name* and *Password* from the Email address you entered.
- 7. After retrieving your login data go back to <u>https://guestnet-portal.desy.de</u> and disconnect the temporary connection.
- 8. You can now log in to the XFEL-Guest network with the credentials you received per E-mail.
- 9. Fill in "Login" (User name) and "Password" and accept the "DESY Condition of Use".
- 10. The account is now valid for 10 days and can be used with 5 different devices at the same time.