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TECHNICAL NOTE

Terahertz Science at European XFEL

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Abstract

Many excitation mechanisms of matter resonate in the terahertz (THz) regime. Recent progress in the generation of intense THz radiation enables driving these excitations far out of equilibrium, providing the tools to decouple various degrees of freedom and their impact on materials' properties. However, these sources are significantly limited in spectral tunability and efforts are ongoing to develop instrumentation for more versatile THz generation. This workshop was organized to obtain feedback from the THz community about the value of being able to generate and probe these out-of-equilibrium states with X-rays. Based on the scientific requirements and the present technological capabilities, this workshop concluded that laser-based sources are relatively inexpensive and have enabled novel experiments on specific sample systems, whereas only accelerator-based sources provide the wide tunability together with high intensity and repetition rates beyond 100 kHz, that will enable broad application at the European XFEL to the most interesting scientific problems in the field. Therefore, the suggested strategy, concluded during this workshop, is to follow two approaches simultaneously:
1) make available laser-based THz sources at every instrument at the European XFEL, based on the available pump–probe (PP) laser, and
2) evaluate two accelerator-based THz sources—one using the electron bunches from the main accelerator of the European XFEL and the other based on a separate accelerator—and ultimately develop and implement at least one of these accelerator-based sources.

Contents

1 Purpose of the workshop

Terahertz (THz)–based research has gained significant attention over the past 20 years as sources have been developed in what was formerly a dark region of the electromagnetic spectrum. This activity is reflected in the publication statistics from the “ISI Web of Knowledge”, which is plotted in Figure 1 and shows a steep increase around the year 2000, shortly after the technique of tilting the pulse-fronts to achieve phase matching in lithium niobate was first described [1, 2].

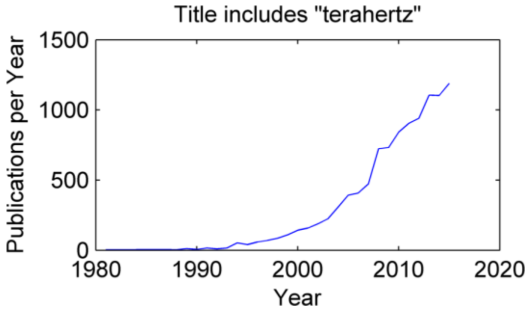


Figure 1. *The rate of publications concerning THz radiation increases steadily since the year 2000, which also coincides with the first reports of the tilted pulse-front technique. Data from the ISI Web of Knowledge.*

This workshop was organized to summarize the scientific opportunities enabled by THz provision as well as the requirements for THz provision at European XFEL and to evaluate the technical potential of various THz sources. The scientific objectives should be compatible with the X-ray instrumentation of the European XFEL, and the sources should enable the widest possible range of experiments. Given that users at the Linac Coherent Light Source (LCLS) at SLAC National Accelerator Laboratory in Menlo Park, California, USA, and the SPring-8 Angstrom Compact Free Electron Laser (SACLA) in Hyogo, Japan, cannot rely on sufficiently versatile THz sources, there is an opportunity to provide access to unique instrumentation.

2 Workshop agenda

The workshop took place at the headquarters of European XFEL in Schenefeld, Germany, on 1–2 June 2017 and centred around the presentations from 18 invited international scientists. The first day explored the scientific motivation for THz–X-ray experiments. The second day focused on the two main routes to THz generation in the frequency range from 0.1 to 20 THz (3 mm to 15 μm wavelength): on the one hand, state-of-the-art laser-based sources; on the other, accelerator-based sources. The final session of the workshop was an open discussion among the participants, initiated by a conclusion presented by the organizers.

Chapters 3 and 4 summarize all presentations given at the workshop with a focus on the scientific requirements for THz sources and the latest developments in THz generation. Chapter 5 summarizes the discussion following the workshop to reflect the evaluation of various generation techniques. Finally, Chapter 6 outlines a strategy to realize THz provision for user experiments at the European XFEL.

3 Motivation and requirements for THz–X-ray science

The first day of the workshop was focused on the application of THz science to condensed-matter physics, where its application is more prevalent than in other disciplines. While most science cases address questions related to solid samples, the enabling character of intense THz sources for novel experiments in the liquid state was outlined as well. It is worth mentioning that every invited speaker working in applied THz science stated that further activities, beyond that presented at the workshop, will be enabled by a tunable, more intense THz source.

3.1 Individual science cases presented at the workshop

Andrea Cavalleri discussed the transient reorientation of ferroelectric polarization in Lithium Niobat ($LiNbO_3$) by resonantly driving phonon modes around 10 THz (see Figure 2). The goal of this work is to demonstrate ferroelectric switching, i.e. an inversion of the polarization vector by coherently driving the respective phonon mode. His group needs higher THz field strengths from a tunable source to drive the polarization further out of equilibrium.

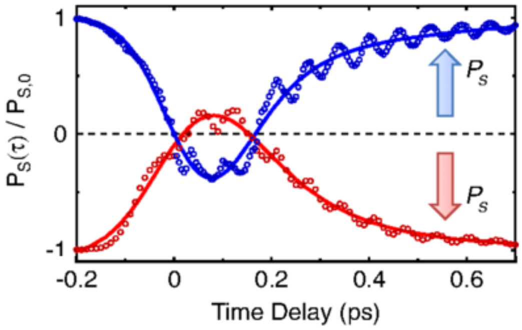


Figure 2. *The polarization of the ferroelectric material $LiNbO_3$ is partly inverted by the THz pulse and then relaxes back to the initial state. From Mankowsky et al., 2017 [3].*

Stefano Bonetti discussed the magnetization dynamics of crystalline (Fe) and amorphous (CoFeB) ferromagnetic materials. The magnetic field component of a single cycle THz pulse leads to precession of the magnetic moment in both materials, but only the amorphous sample is demagnetized (see Figure **3**). These results show that defects lead to enhancement of spin-lattice scattering [4]. His group intends to perform time-resolved X-ray diffraction and XMCD experiments after strong-field, tunable THz excitation to explore the structural and chemical origin, respectively, of this mechanism.

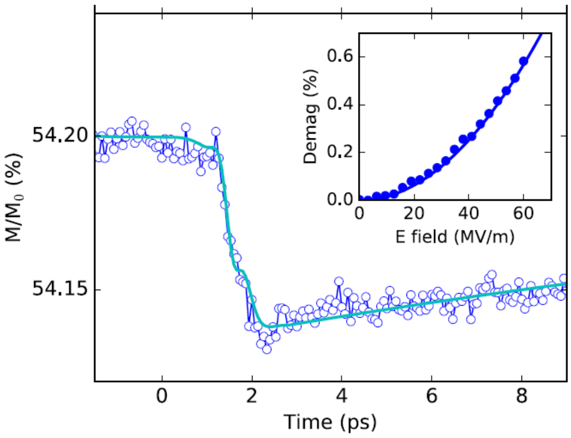


Figure 3. *Demagnetization of CoFeB by a THz pulse. The inset shows how the extent of demagnetization scales with the electric field strength of the pulse. From Bonetti et al., 2016 [4].*

Karsten Holldack presented the THz source at BESSY II that generates radiation from femto-sliced electron bunches passing through an undulator. This radiation was applied in Fourier domain, Fourier transform THz electron paramagnetic resonance (FD FT THz EPR) experiments and in time-resolved THz spectroscopy. The EPR experiments can be used to explore the coupling of spin states in transition metal ions of organometallic compounds from proteins (e.g. Fe(III) in Hermin [5]) and single molecule magnets. In time-resolved spectroscopy, the tunability of the THz pulse is combined with an optical laser pulse. In this case, THz pulses were used to probe vibrational modes in chalcogenide glasses (see Figure 4), whose sub-threshold reversible crystallization and amorphization dynamics [6, 7] are employed in novel electronic memory devices, which could possibly also be switched by sufficiently strong THz pulses, not presently available.

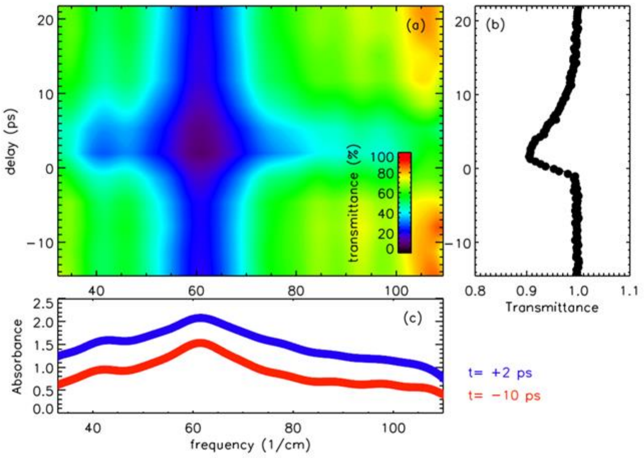


Figure 4. *a) Time and frequency PP maps of* Sb_2Te_3 *thin films upon pumping with 0.1 µJ* cm^{-2}*, 100 fs optical pulses at 800 nm. b) Delay trace of the total transmitted THz signal. c) Change of the absorbance spectrum around the E(u) TO phonon mode at 61* cm^{-1} *before and after time-zero. See also Ref. [6].*

Dmitry Turchinovich discussed non-linear THz spectroscopy experiments on graphene in which the strong THz pulse modulates and probes the electronic conductivity. The onset of non-linear effects is found already at field strengths below 10 kV/cm (see Figure 5) and is explained by carrier heating. It is of interest to see how this carrier heating interacts with the lattice of graphene at even higher field strengths.

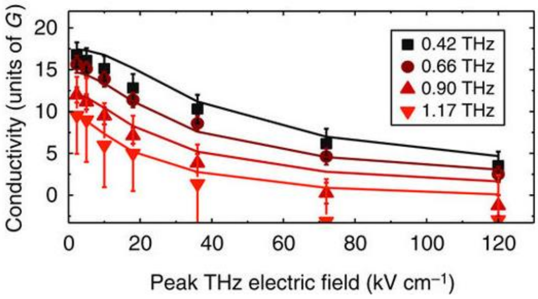


Figure 5. *The conductivity of graphene is affected non-linearly by even moderate field strengths in the THz regime. From Mics et al., 2015 [8].*

Keith Nelson presented evidence for a THz-induced metal-insulator transition in VO_2 [9]. To achieve the required field strength, resonator electrodes with a narrow gap of ~ 1 µm size were used. THz sources with sufficient field strength would resolve this issue. Furthermore, it was outlined how such sources could be used for time-resolved electrochemistry in liquid samples, e.g. water.

Aaron Lindenberg presented THz-induced switching experiments of ferroelectrics performed at the Linac Coherent Light Source (LCLS) at SLAC National Accelerator Laboratory in Menlo Park, California. Due to the absence of sufficiently strong THz pulses, he had to work on $BaTiO_3$ instead of the more interesting multiferroic $BiFeO_3$, which could be excited more strongly due to the lower coercive field. Furthermore, a THz-induced electronic breakdown mechanism, commonly referred to as threshold switching, was presented in the phase-change material $AgIn{:}Sb_2Te$ [10]. To induce this transition, electrode structures had to be employed, but further experiments using time-resolved X-ray experiments should resolve the subsequent THz-induced crystallization mechanism in detail.

In a departure from condensed matter physics, *Louis DiMauro* discussed scaling laws of THz-based experiments and how they lead to the expectation of performing atomic and molecular physics experiments at significantly lower pulse energies when the wavelength is increased into the IR and THz regimes [11]. Based on the scaling, it became clear that the very highest THz intensities of more than 300 GW/cm^2 would be required for these experiments.

Adrian Cavalieri discussed photoemission, in particular the different emission times of photoelectrons and Auger electrons, and how these can be measured by streaking them with a THz field [12]. The application of this technique in determining the phase of the THz pulse was discussed.

Michael Gensch gave examples of samples and experimental approaches from the High-Field High-Repetition-Rate Terahertz facility @ ELBE (TELBE) at Helmholtz-Zentrum Dresden-Rossendorf (HZDR) in Germany. Techniques include THz TDS, Faraday/MOKE, THz emission, THz frequency mixing, coherent phonon spectroscopy, and transient reflectivity, which have been applied to samples such as graphene, CoFeB, $YFeO_3$, VO_2, LSMO, SCO, STO, SCVO, HoIG, water, carbon disulphide, ethanol, and others. However, in many cases, it was not clear if the non-linear regime could be reached under the present conditions of TELBE (1 μJ pulse energy). He furthermore explained how synchronization issues of accelerator and laser sources, operating at MHz repetition rates, have been solved for TELBE [13], concluding that this would not be an issue at European XFEL.

Experiences in laser-based THz sources coupled to ultrafast X-ray experiments at LCLS were the subject of the talk by *Matthias Hoffmann*. Topics included THz streaking, selective excitation of phonon modes, THz excitation of multiferroics, and driving the metal-insulator transition in VO_2.

Christoph Hauri discussed resonant and non-resonant excitation in the context of laser-based THz sources and showed an example of the highly nonlinear physics possible with intense THz pulses [14]. A broadband THz pulse focused to 25 MV/cm gave rise to a transient metal-like state in the semiconductor gallium phosphide. This was associated with strong alteration of the optical properties of the material, as witnessed by extreme cross-phase modulation of an optical laser pulse. The presentation emphasized the need for both broad- and narrow-band sources, as well as frequency tunability.

3.2 Summary of the technical requirements derived from the science cases

The above examples do not represent a complete overview of research involving THz radiation. Rather, they are intended to show that the broad and growing interest in THz radiation will be of interest to users at the European XFEL. Since X-rays are an ideal technique to probe e.g. atomic structure, the combination of THz pulses, inducing atomic and molecular dynamics with structural probes, is particularly promising. From these science cases, we can derive requirements for THz sources at the European XFEL, which are compiled also based on the contributions by Christoph Hauri (who hosted a similar workshop entitled “Workshop on an accelerator-based source for nonlinear THz science @ SwissFEL” on 3 October 2016) from the Swiss Free-Electron Laser (SwissFEL) at Paul Scherrer Institut (PSI) in Villigen, Switzerland, and Matthias Hoffmann from LCLS.

The following list summarizes all capabilities of an ideal THz source for time-resolved experiments at the European XFEL:

1. **Bandwidth:** Tunable bandwidth ΔE/E between 1 (single-cycle, shortest pulse possible) and 0.05 (multi-cycle, to coherently drive matter).
2. **Frequency:** Tunable centre frequency in the range 0.1 to 30 THz (3 mm to 10 µm wavelength). Within this range, 3 to 20 THz is the most difficult to cover by existing sources; at the same time, many vibrational resonances and relaxations in condensed matter occur at these frequencies.
3. **Pulse fluence/field strength:** More than 2 MV/cm, which corresponds to > 10 GW/cm^2. Pulses of 1 ps duration would then generate fluences of > 10 mJ/cm^2. Assuming a focus size with diameter of the wavelength, this requires pulse energies of 3 mJ at 0.1 THz and 30 µJ at 1 THz. At 10 THz, 0.3 µJ would be sufficient in principle, but the ideal focussing can most likely not be achieved and therefore a minimum of 10 µJ should ideally be achievable at all frequencies.

4 **Carrier envelope phase (CEP):** Should be either stable (i.e. each pulse has the same temporal electric field E(t)) or, alternatively, it must be measured for each pulse. The CEP-stable option simplifies data processing significantly.

5 **Repetition rate:** To make best use of the potential of the European XFEL, the source should operate at least at 0.1 MHz but ideally could follow the 4.5 MHz bursts.

6 **Synchronization:** Temporal jitter must be better than 0.1/frequency to resolve the electric field cycles, e.g. < 20 fs at 5 THz. This could be either the intrinsic jitter or the resolution of a timing measurement.

7 ***Optional:*** **Polarization control:** Could be achieved with optics after THz generation and does not need to be considered here.

Most likely, no single source exists that can cover the entire parameter range and all combinations. Therefore, it makes sense to categorize science cases further. Quite generally, one can distinguish two types of experiments using THz pulses as pump: resonant and non-resonant excitations. For **non-resonant** excitation, the main purpose of THz pulses is to provide an ultrafast electrical and/or thermal stimulus, which does not induce optical interband transitions, or specific vibrational modes. This requires the shortest pulses with the highest energy and a stable CEP, while spectral tunability is not critical. **Resonant** excitation of matter requires frequency tunability and, ideally, also bandwidth optimized to match the underlying mechanism. While the CEP must also be known for each pulse, the required pulse energies are less critical.

4 Technical solutions for THz provision

During the second day of the workshop, the talks were focused on THz generation using optical lasers and accelerators. A wide range of possible sources was discussed by leading scientists in the corresponding fields with the aim of evaluating the performance of each technique for application at the European XFEL. THz pulses can be generated most readily from the PP laser available at all instruments. Some of these techniques are well developed and understood, and require only minor efforts to deploy. Therefore, these sources should naturally be considered first. In the following sections, five different laser-based approaches are discussed. It turns out that laser-based sources cannot cover the entire frequency range given in Item 2 in Section 3.2, “Summary of the technical requirements derived from the science cases”. Therefore, we also seek to identify the ideal accelerator-based source to cover the missing parameter space.

4.1 Laser-based THz sources

Any laser-based THz provision at the European XFEL will be based on the existing optical PP laser. Based on this, we discuss five different techniques of laser-based THz generation and their possible implementation. It is worth mentioning that most of these sources, due to their low conversion efficiencies, are operated in laser labs at kHz repetition rates. For many of these techniques, the scaling to higher repetition rates has been hardly studied, and the exchange of this information was therefore a main topic of the workshop. It should be mentioned that two of these techniques—photoconductive switches and gas-plasma sources—are commonly used to generate THz pulses at high repetition rates, albeit with low damage threshold (photoconductive switches) and low conversion efficiency (gas-plasma sources). While they were not discussed during the workshop, key parameters of these techniques are included in this report. In particular, their potential application as complimentary probe sources, e.g. via a THz time domain spectroscopy (TDS) probe, should be considered by the instrument groups.

4.1.1 Pump–probe laser at the European XFEL

Max Lederer described the design and operation of the PP laser, depicted schematically in Figure 6. The laser comprises a fiber oscillator synchronized to the photocathode laser of the free-electron laser (FEL), followed by an Yb-based amplifier chain operating at 1030 nm. The frequency-doubled amplifier output is used to pump a series of non-collinear optical parametric amplifiers (NOPA) configured to produce broadband pulses at 800 nm.

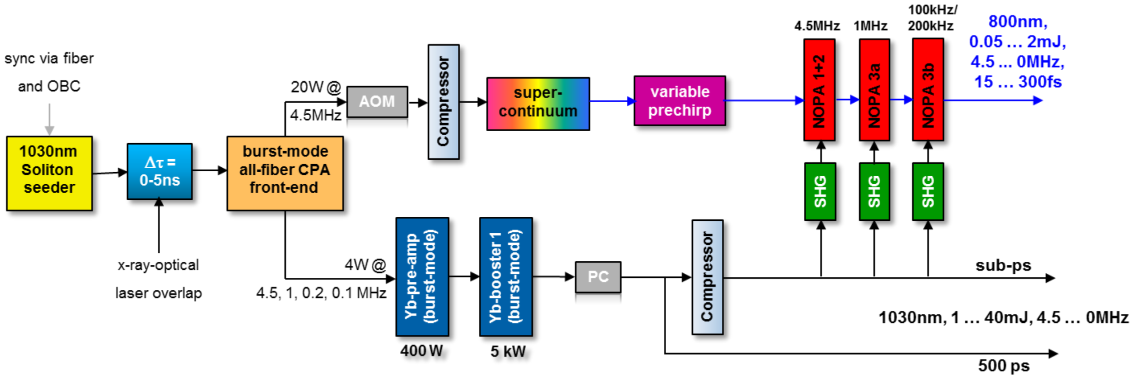


Figure 6. *Schematic of the PP laser at the European XFEL. See also Ref. [15].*

The laser has several operating modes to provide a variety of pulse parameters at key repetition frequencies of the FEL, as shown in Table 1.

Table 1. *Operation modes of the European XFEL PP laser. See also Ref. [15].*

				800 nm	1030 nm
Mode	**F_{rep} / MHz**	**Δt / ns**	**F_{eff} / Hz**	**E_{pulse} / mJ**	**E_{pulse} / mJ**
1	4.5	222	27000	0.05	1
2	1.13	886	6000	0.2	4
3	0.103	9750	600	2	40
			Pulse duration	15–300 fs	< 1 ps or 400 fs

4.1.2 Optical rectification in Lithium Niobate

Using optical rectification (OR) in $LiNbO_3$ with tilted pump pulse fronts, it is possible to generate broad-band, single-cycle pulses centred around 0.3 THz with pulse energies up to the mJ level [16]. These pulses are CEP-stable, but their centre frequency and bandwidth cannot be tuned. Preliminary in-house research at European XFEL, supported by Franz Kärtner's group, has shown that the 1030 nm output of the PP laser in Operation Mode 3 can be used to generate THz pulses of 70 µJ. This value is a factor of four below the required pulse energy in Item 3 in Section 3.2, “Summary of the technical requirements derived from the science cases”. Given the 4 kW average power in the pump burst, resulting in 7 W of THz radiation, this technique is operating close to its design limit due to heat dissipation in the $LiNbO_3$ crystal—as acknowledged also by the audience. Further increase of the repetition rate is possible only with reduced pulse energy, following the performance limit of the PP laser. These pulses are suitable for non-resonant experiments at 100 kHz and, in some cases, could even be used for resonant excitation when a suitable sample system is available. *Franz Kärtner* presented ongoing efforts in his group to characterize and optimize the efficiency of THz generation from this source. However, the output power of 7 W is not expected to improve by an order of magnitude. Many implementations of this technique were presented in the workshop, with different approaches to increasing the conversion efficiency.

4.1.3 Optical rectification in organic crystals

Using OR in various organic crystals, pulses with bandwidth of roughly $\Delta E/E = 0.1$ can be generated at fixed frequencies around 2 THz and pulse energies up to 10 µJ (see the emitted spectra in Figure 7). When used with a mid-IR (ca. 1.1 to 1.5 µm) driving laser, the conversion efficiency is approximately 2%, which is significantly higher than for $LiNbO_3$. The optical setup is also simpler: the generated THz is coaxial with the laser beam. Furthermore, the beam profile allows better focusing than with THz from $LiNbO_3$. On the other hand, the expensive crystals can be easily damaged by the driving laser and are known to degrade over time.

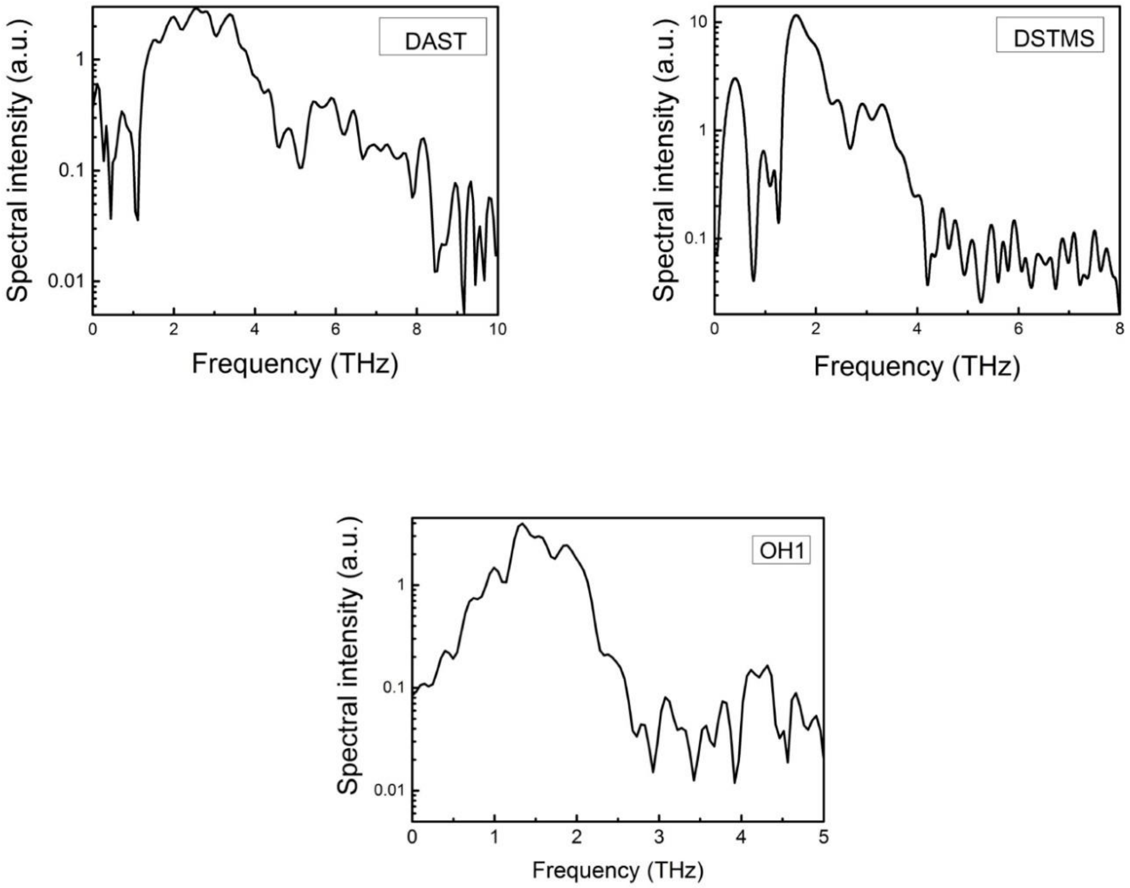


Figure 7. *THz spectra from OR in the organic crystals DAST, DSTMS, and OH1, all pumped at 1.25 µm wavelength. From Vicario et al., 2015 [17].*

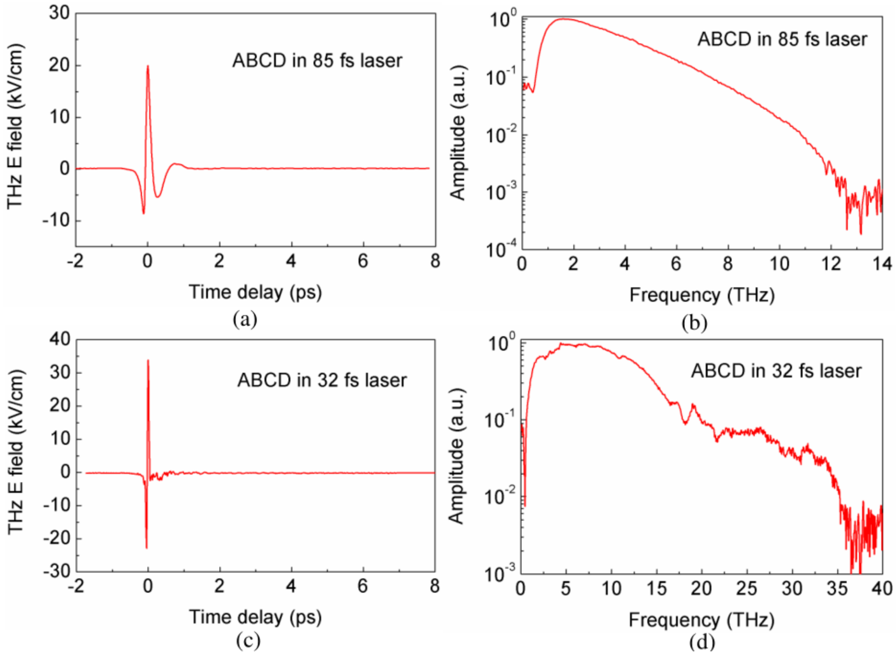
Operation of these sources at or below 10 µJ was demonstrated only up to a few tens of kHz (as presented by *Matthias Hoffmann*). The highest possible output pulse energy at 100 kHz or beyond is presently unknown but should exceed 1 µJ. At 3 THz, this is a factor of three below the requirement listed in Item 3 in Section 3.2, “Summary of the technical requirements derived from the science cases”. The spectral bandwidth (and therefore pulse duration) of these pulses can be selected by interferometric techniques (as presented by *Christoph Hauri*). Due to the lack of tunability, these pulses are suitable only for non-resonant experiments.

4.1.4 Difference frequency generation in organic crystals

Using difference frequency generation (DFG) in organic crystals, tunable, narrow bandwidth pulses between 4 and 18 THz have been generated very recently in the group of *Andrea Cavalleri* [18]. However, pulse energies were limited to 2 µJ at 1 kHz. While the scaling of this source to higher repetition rates is presently unknown, the current performance scales to 0.02 µJ at 100 kHz, which is more than an order of magnitude below the required pulse energy. Furthermore, this approach requires two optical parametric amplifiers (OPAs) operating at approximately 1.5 µm, so the laser setup is relatively complicated. In this context, the need for an R&D effort in the 5–15 THz range was noted during the workshop. The possibility to reconfigure the PP laser to pump two OPAs for THz generation by DFG at around 1.5 µm was discussed and could, in principle, start relatively soon.

4.1.5 Gas plasma sources and photoconductive switches

Optical rectification in a gas plasma creates ultrabroadband THz radiation, ideally suited for spectroscopic ultrafast probing (see Figure 8). This technique becomes efficient only with pulse durations shorter than 100 fs, because the pulse duration limits the peak frequency of the spectrum. Using 40 fs pulses, conversion efficiencies are on the order of 10^{-4} [19], so pulse energies of around 0.2 µJ are expected using the 800 nm pulses from the PP laser at 100 kHz. Peak field strengths of 2.5 MV/cm have been achieved [20], exceeding the requirement for nonlinear THz science at frequencies beyond 10 THz. While this source is not tunable and therefore suitable only for non-resonant experiments, it is expected to scale up to MHz repetition rates, provided that the PP laser could provide sufficiently energetic pump pulses.



(a) (b) (c) (d)

Figure 8. *Electric field waveform and spectral composition of THz radiation obtained from air-based plasma generation. From Ho et al., 2010 [19].*

Photoconductive switches generate the THz pulse energy from an externally biased capacitor structure, which is short-circuited by the photoexcited carriers in a semiconductor in contact with both electrodes. These devices were shown to generate THz pulses with peak field strengths of up to 100 kV/cm, limited by saturation of the semiconductor material [21]. Due to the simple operation of these devices and the efficient conversion at low pump fluences, this technique is commonly used in turn-key THz TDS systems based on oscillator pump sources. Due to their low peak field strength, these systems are not suitable as pump sources at the European XFEL.

4.2 Accelerator-based THz sources

The discussion at the workshop confirmed that there is no laser-based source in sight that can fulfil Items 1–5 in Section 3.2, “Summary of the technical requirements derived from the science cases”. Therefore, accelerator-based THz sources need to be considered to fill the remaining THz gap and to make such pulses available for the first time at any hard X-ray FEL source.

4.2.1 Additional MeV-scale accelerator

The idea of installing an additional MeV-scale accelerator and undulator for THz generation at the European XFEL was already evaluated and proposed several years ago [22]. Ongoing R&D work on a prototype IR/THz source at the Photo Injector Test facility at DESY, Zeuthen (PITZ), was presented by *Mikhail Krasilnikov*. The proposed design uses electron bunches with 4 nC and 20 ps pulse duration to generate tunable radiation between 3 and 15 THz using an APPLE II undulator. This source could operate at 4.5 MHz to match the X-ray bursts of the European XFEL. Pulse energies of a few mJ are expected. However, due to the SASE process (the electron bunch length being longer than the inverse frequency of the THz radiation), this source is no longer CEP-stable. Microbunching techniques were discussed to recover CEP stability, but these mostly require a tunable THz source to begin with.

CEP-stable pulses can be generated with superradiant sources, such as the TELBE facility [23] introduced by *Michael Gensch.* Electron bunches of sub-ps duration and 27 MeV generate THz radiation in an undulator with 300 mm period. Because the bunches are shorter than the inverse frequency of 0.3 to 1.1 THz generated at this source, they do not experience microbunching and therefore generate CEP-stable radiation. The resulting pulses are depicted in Figure 9. Timing jitter between the accelerator source and an optical laser used for PP experiments can be effectively measured on a single-shot basis using a separate source of coherent transition/diffraction radiation (CTR) in the electron path [13] (see also Figure 10). Therefore, temporal synchronization does not induce any limitation on the combination of an X-ray FEL with a THz FEL.

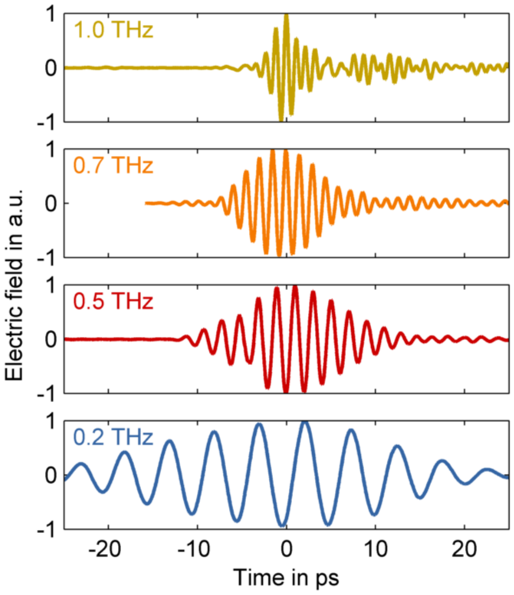


Figure 9. *Tunable, CEP-stable THz pulses, generated at TELBE at a 100 kHz repetition rate and around 1 µJ pulse energy (limited by the bunch charge of only 0.1 nC) [24].*

Extending this concept to higher THz frequencies and pulse energies, *Nikola Stojanovic* presented THz activities at the Free-Electron Laser in Hamburg (FLASH) at DESY in Germany, where a THz afterburner undulator situated after the XUV undulators produces radiation between 2 and 12 THz with 100 µJ pulse energy. Compared to TELBE, FLASH employs higher electron energies and bunch charges, up to 1.25 GeV and 1 nC, respectively.

Aiming for even shorter electron bunches even in the MeV regime, *Andrej Savilov* described the theoretical analysis of THz generation from a negative mass undulator. Using this technology, one could produce CEP-stable THz pulses up to higher frequencies than using a regular undulator [25, 26]. The approach would require technical evaluation and R&D effort, but might be considered as an upgrade.

Zhirong Huang described a proposal to use a train of laser pulses to modulate a MeV-scale electron bunch such that a subsequent undulator emits CEP-stable THz radiation. A similar approach has been demonstrated in the XUV at the Free Electron Laser for Multidisciplinary Investigations (FERMI) at Elettra Sincrotrone Trieste in Italy [27]. The scheme is expected to generate tunable radiation between 1 THz and 10 THz (possibly up to 20 THz) at high repetition rates with pulse energies of tens to hundreds of microjoules.

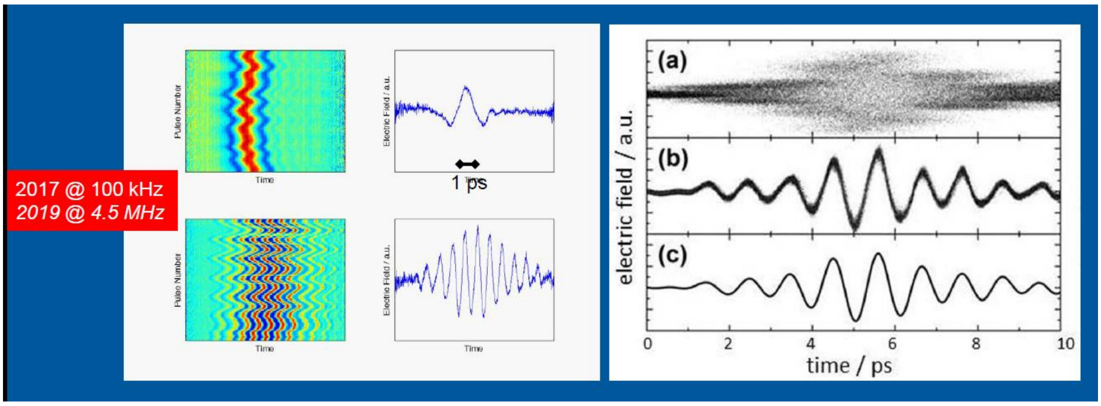
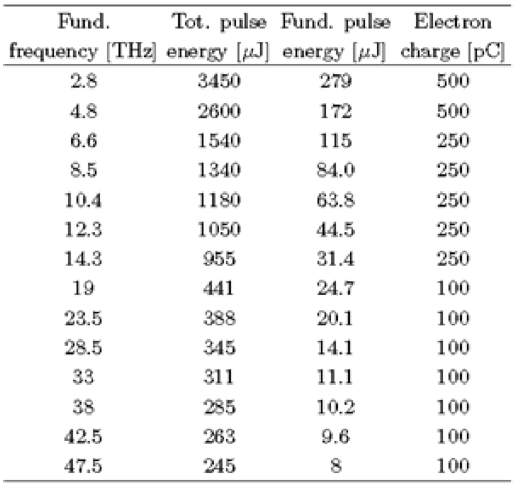


Figure 10. *Single shot electro-optic sampling of THz pulses from the undulator at TELBE, showing resolution down to tens of femtoseconds. CTR is shown in the top left panels and the undulator radiation in the lower-left panels. The source is CEP-stable and, measured in this way, also has low femtosecond level jitter, so it fulfils Items 4 and 6 in the Section 3.2, "Summary of the technical requirements derived from the science cases". See also [23].*

4.2.2 Using an additional undulator in the GeV bunches

Following an alternative approach, the use of the GeV electron bunches of the European XFEL was discussed for THz generation. *Gianluca Geloni et al.* suggested an approach in which a superconducting undulator would be installed in the existing SASE3 tunnels. With a long period of 1 m and being based on a novel superconducting technology, it could generate CEP-stable THz pulses in the range from 3 to 50 THz. Table 2 [28] shows that the bunch charge has to be reduced to < 100 pC to access the > 10 THz regime. While this would also affect the X-ray generation, it was proposed to inject additional pulses with less charge 21.5 ns before the main bunch, employing a double pulse scheme recently demonstrated at FLASH to generate the THz pulses. Because the electron bunches in SASE3 are dumped 150 m before the experiment hall, the THz beam would need to be transported through vacuum tubes with a predicted transmission ratio of 60%. The X-ray–THz time delay could then be realized and adjusted by an optical delay line in the THz beam path and/or by the chosen time separation of the "THz" and "X-ray" bunches in the double pulse scheme.



Fund. frequency [THz]	Tot. pulse energy [μJ]	Fund. pulse energy [μJ]	Electron charge [pC]
2.8	3450	279	500
4.8	2600	172	500
6.6	1540	115	250
8.5	1340	84.0	250
10.4	1180	63.8	250
12.3	1050	44.5	250
14.3	955	31.4	250
19	441	24.7	100
23.5	388	20.1	100
28.5	345	14.1	100
33	311	11.1	100
38	285	10.2	100
42.5	263	9.6	100
47.5	245	8	100

Table 2. *THz generation from a THz afterburner undulator at GeV electron bunches of the European XFEL. Note that, while the total pulse energy, including the higher harmonics, can be in the few mJ regime, the pulse energy in the actual fundamental is between 15 and 3 THz scaling from 30 to ~ 300 µJ. [28]*

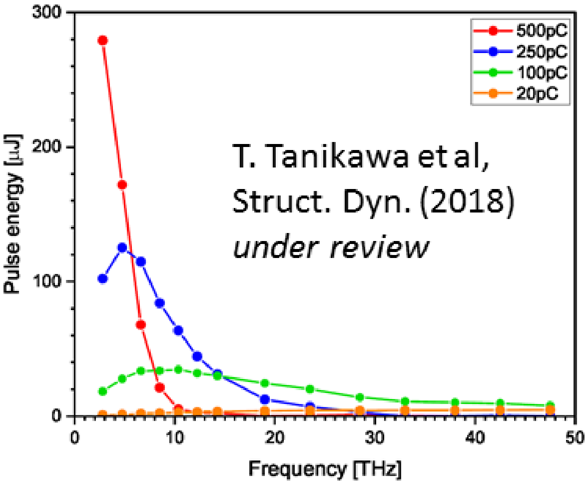


Figure 11. *THz generation from a THz afterburner undulator at GeV electron bunches of the European XFEL. Note that, while the total pulse energy, including the higher harmonics, can be in the few mJ regime, the pulse energy in the actual fundamental is between 15 and 3 THz, scaling from 30 to ~ 300 µJ. [28]*

4.2.3 Coherent transition radiation

An alternative approach is to use CTR for THz-based experiments. Such a source is operational at TELBE. *Andrea Perucchi* described the technical implementation and commissioning of TeraFERMI, a CTR source of sub-picosecond THz pulses at 1–5 THz incorporated into the FERMI LinAc. While this source provides intrinsically broadband THz radiation, its centre frequency cannot be tuned. Therefore, plans to modulate the electron bunches to generate narrow-band, tunable THz radiation are ongoing.

5 Summary of the discussion

The discussion at the end of the workshop was held to evaluate the potential of the various techniques for THz generation presented earlier. Direct feedback from potential users in the audience was provided and considered in deriving the suggested strategy for THz provision at the European XFEL.

From the talks about facilities already offering THz radiation, we can conclude that THz provision by the European XFEL will be of strong interest to users. It was concordantly agreed that laser- and accelerator-based THz sources are complementary approaches, because laser-based sources can cover only a limited fraction of the required parameter range. The enabling character of accelerator-based sources became apparent also from the scientific talks during the first day, which reported efforts mostly limited by the availability of suitable sources.

The potential scalability of laser-based sources was discussed for all approaches. While OR in LN can just provide sufficient pulse energies at 100 kHz repetition rates, there is no chance to scale it to higher repetition rate and to achieve frequencies different from 0.2–0.3 THz.

Christoph Hauri and Matthias Hoffmann stated that they expect OR in organic crystals to be limited to repetition rates at or below 10 kHz without compromising the THz pulse energy of few µJ, which at the same time is required to enable THz excitation. Difference frequency generation in organic crystals is expected to have the same limitation. Despite its frequency tunability it is expected not to be able to achieve the required pulse energies at repetition rates beyond 10 kHz. It is also important to consider the maximum available size of these specific organic crystals of about 10 mm, which prohibits further scaling of this technique.

With these limitations to laser-based THz sources, it becomes apparent that accelerator-based sources can out-perform laser-based sources in pulse energy, tunability and repetition rate, and the discussion therefore focused mostly on the various accelerator-based concepts (see Figure 12).

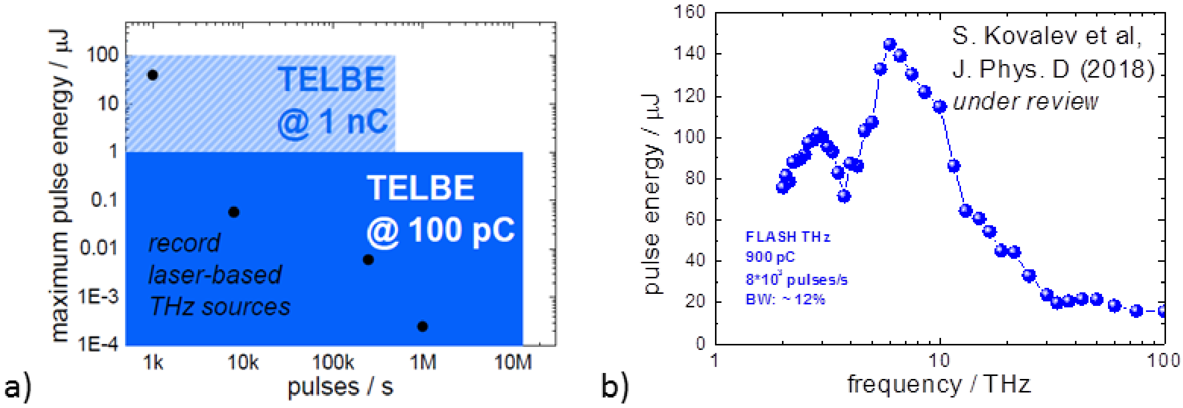


***Figure 12**. a) Design parameters of the THz sources at TELBE compared to laser-based sources [23]. The final available THz frequency range should be 0.1 to 3 THz. It should be noted, however, that the full extent of this parameter space (the 1 nC operation) has yet to be demonstrated. Figure courtesy of M. Gensch.*
b) Experimentally demonstrated pulse energies at the THz undulator afterburner facility at FLASH [30]. Figure courtesy of N. Stojanovic.

There were concerns as to the whether the design pulse energies of accelerator-based sources could be achieved. We note, however, that at FLASH at least 40 µJ THz pulses at 1 MHz repetition rate have been successfully generated, fully meeting the requirements listed in Section 3.2.

With the above accelerator-based presentations in mind, the following two approaches to accelerator-based THz generation at the European XFEL could cover all requirements listed previously. We note that both approaches require further R&D.

1 An additional injector gun as developed at PITZ with an accelerator that reaches electron energies up to around 30 MeV and the capability to compress electron bunches to sub-ps duration. Together with an Apple II-type undulator, this source would enable operation in the superradiant regime, with the shortest electron bunches generating CEP-stable pulses below an upper limit of at least 3 THz and, in the SASE regime, using uncompressed electron bunches, generating THz pulses of up to 15 THz.

At higher frequencies, CEP stability could be achieved by:

  - **a** Optimized compression of electron bunches
  - **b** Modulation of the electron current within the burst (see talk by Zhirong Huang)
  - **c** Seeding (ongoing work at PITZ)
  - **d** Dielectric-lined waveguides [31]
  - **e** Negative mass undulator design (see talk by Andrey Savilov)

2 An additional long-period, superconducting undulator installed in the existing electron tunnels. While this source would fulfil all requirements for THz-generation, the R&D involved in the design of a 10 m long superconducting undulator is not yet clear.

It should be mentioned that CTR radiation from high-energy electron bunches passing through e.g. a metal foil produces THz radiation. However, the radial polarization of this radiation makes it rather complicated to handle and it lacks the required tunability and the option for generation of narrow-bandwidth THz pulses, which is why this type of source is not considered further for implementation at the European XFEL.

6 Suggested strategy for THz provision at European XFEL

Based on the arguments in Chapter 5 above, we can conclude that THz radiation sources will be a popular and powerful adjunct to X-ray experiments at the European XFEL. The facility should aim to provide a full range of THz radiation as soon as possible.

Bearing in mind the different timescales and budgetary requirements of laser-based and accelerator-based sources, the versatility of THz sources at the European XFEL should be increased sequentially in three stages:

1. **$LiNbO_3$-based THz sources driven by the 1030 nm output of the PP laser.** This source was shown to deliver up to 40 µJ pulses at a 0.3 THz and 100 kHz repetition rate. It can be made available where desired as soon as the corresponding SASE1, SASE2, and SASE3 PP lasers are operational. Several of the experiments that require “incoherent” THz excitation are expected to be possible with this source. Likewise, OR in organic crystals can be done with the 1030 nm beam of the PP laser or by using the output of a commercial OPA at 1.1 to 1.5 µm. Such an OPA has been tested with the PP laser and found to perform well at 100 kHz. However, the use of organic crystals is expected to require further R&D to test the performance at repetition rates beyond 1 kHz, which has not been reported in literature. This effort does not require additional funding or personnel.
2. **DFG-based sources producing 4–18 THz:** Two dedicated OPAs driven by the 1030 nm output of the PP laser perform difference-frequency generation in organic crystals. This setup could be developed and built in a preparation environment and deployed at the instruments as required. To overcome the pulse energy limitation of 1 µJ seen in Ref. [18] and explore the available repetition rates, significant R&D effort is to be expected.

3 **An accelerator-based source** to enable “coherent” excitation experiments that require a tunable THz source. Technical details and funding options are still to be clarified. The accelerator could be installed in the hall or in one of the existing as-yet-unused tunnels next to an X-ray beam pipe. The alternative concept of a superconducting undulator, driven by the GeV electron bunches of the European XFEL, could be installed in SASE3 without modification to the existing electron beam dump.

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