TECHNICAL NOTE

Requirements and concept for the characterization of photon beam temporal properties at the SQS scientific instrument of the European XFEL facility

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at European XFEL
## Revisions

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Introduction

The Small Quantum System (SQS) scientific instrument is one of the six planned research end-stations of the European XFEL facility, which will become operational in 2017 and take great advantage of its extremely high number of light pulses (27 000 p/s) and of the two or three orders of magnitude higher average brilliance than that achieved at other free-electron laser (FEL) facilities. The European XFEL will thus enable novel experiments in atomic and molecular physics and, more generally, various new investigations in chemistry at the SQS instrument. The X-ray FEL pulses at the European XFEL are produced in 10 Hz bunch trains, each of which contains up to 2700 sub-pulses within the 600 μs time duration of the bunch train, corresponding to a 222 ns separation of individual light pulses or a 4.5 MHz repetition rate, as clarified in the SQS conceptual design report (CDR) and technical design report (TDR) [1-5].

Figure 1: Layout of the XFEL.EU SASE beamlines and photon beam transport

The layout of the SASE beamlines and photon beam transportation of the European XFEL is shown in Figure 1. Briefly, electrons generated from the electron gun are first accelerated by a linear accelerator to high energies and then manipulated to generate high-intensity X-ray laser pulses in different undulators. SQS receives the soft X-rays produced by the SASE3 undulator, whose parameters can be found in Ref. [1]. The simulated radiation parameters for SASE3 are listed in Table 1 [1]. The SASE 3 beamline is
dedicated to the transport of soft X-ray radiation in the photon energy range from 250 up to 3000 eV, enabled by tuning the SASE3 undulator gap and the incident electron energy. The pulse duration varies from ~ 100 fs down to an expected shortest length of 2 fs, depending on the FEL operation mode.

**Table 1: Simulated XFEL radiation parameters for SASE3, reproduced from Ref. [1]**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Photon energy</td>
<td>keV</td>
<td>0.28 0.496 1.55 3.1</td>
</tr>
<tr>
<td>Electron energy</td>
<td>GeV</td>
<td>10.5 14 14 14</td>
</tr>
<tr>
<td>Bunch charge</td>
<td>nC</td>
<td>0.02 0.25 1 0.02 0.25 1 0.02 0.25 1</td>
</tr>
<tr>
<td>Coherence time</td>
<td>fs</td>
<td>1.49 1.63 1.83 1.01 1.11 1.25 0.43 0.49 0.57 0.26 0.31 0.38</td>
</tr>
<tr>
<td>Pulse duration</td>
<td>fs</td>
<td>2 23 107 2 23 107 2 23 107 2 23 107</td>
</tr>
<tr>
<td>Spectra bandwidth</td>
<td>1E-3</td>
<td>6.9 6.4 5.7 5.8 5.3 4.7 4.4 3.9 3.3 3.6 3.1 2.5</td>
</tr>
<tr>
<td>Spectral bandwidth</td>
<td>eV</td>
<td>1.93 1.79 1.60 2.88 2.63 2.33 6.82 6.05 5.12 11.2 9.61 7.75</td>
</tr>
<tr>
<td>Peak power</td>
<td>GW</td>
<td>105 106 94 118 119 105 90 86 72 63 66 51</td>
</tr>
<tr>
<td>Average power</td>
<td>W</td>
<td>5 67 273 5 75 304 4 54 209 3 41 147</td>
</tr>
<tr>
<td>Source size</td>
<td>µm</td>
<td>51 65 79 43 56 68 39 50 60 36 46 56</td>
</tr>
<tr>
<td>S. divergence</td>
<td>µrad</td>
<td>33.9 29.6 26.0 23.1 20.0 17.5 9.7 8.1 6.9 5.6 4.6 3.9</td>
</tr>
<tr>
<td>Photon/ pulse</td>
<td>1E11</td>
<td>39 547 224 25 347 1420 6 81 312 2 31 109</td>
</tr>
<tr>
<td>Pulse energy</td>
<td>mJ</td>
<td>0.2 2.5 10.1 0.2 2.8 11.3 0.2 2.0 7.8 0.1 1.5 5.4</td>
</tr>
<tr>
<td>Peak brilliance</td>
<td>1E31*</td>
<td>7 7 7 16 17 17 49 54 53 98 104 93</td>
</tr>
<tr>
<td>Average brilliance</td>
<td>1E21*</td>
<td>3 46 212 7 109 498 22 338 1520 44 654 2680</td>
</tr>
</tbody>
</table>

*In units of photon / (mm²·µrad²·0.1% bandwidths)

SQS aims to investigate the nonlinear dynamics of atoms, ions, molecules, and clusters in intense fields, and one of the important research topics of SQS is related to time-resolved studies using two-colour pump and probe fields. Exploiting the high FEL peak brilliance, X-ray induced dynamical processes in atomic or molecular systems can be followed by applying X-ray–optical pump–probe schemes that are expected to open a new era for physics and chemistry. For example, time-resolved photo-fragmentation studies of small molecules can be performed by using e.g. an ultrashort optical infrared (IR) pump pulse and a time-delayed X-ray probe pulse that give access to experimental observations focused on the direct study of spatial structure in molecular systems. As a consequence, it is important to synchronize the X-ray pulses to those of the external optical pump pulses with high accuracy on the order of a few femtoseconds. Furthermore, the pulse duration and pulse structure have proved to be important parameters in X-ray–optical target interaction. By knowing accurately the pulse duration...
(together with the spatial dimensions of the beam), the peak intensities of the X-ray pulses in the interaction region can be obtained, which is of utmost significance for the investigation of non-linear processes and time-resolved measurements of the dynamics of photon–matter interaction processes.

Even though the pulse length can be estimated theoretically by measuring the self-amplified spontaneous emission (SASE) FEL spectrum and by analysing the statistics of the pulse energies, direct time domain measurements are required to prove the theoretical considerations [6,7]. On the other hand, X-ray FELs have established themselves as essential and unique sources of ultrabright and tunable X-ray radiation. Similar to ultrashort optical laser pulses, ultrashort X-ray pulses are greatly appreciated as a key feature of FELs for material and biological characterization on single-shot basis with fs-scale temporal and nm-scale spatial resolution. Machine operation studies such as pulse length tuning will also need direct and precise characterization techniques to determine the longitudinal X-ray pulse profile and pulse duration [8, 9].

The main advantage of the European XFEL (SQS) is its extremely high number of photons per second and the two or three orders of magnitude higher average peak brilliance as compared with existing FEL facilities. Such a unique photon energy region and high repetition rate together with pulse duration and spectral bandwidth should be addressed for choosing and designing the timing tools for the European XFEL and the SQS instrument. One thus expects to develop an independent flexible universal timing tool for all X-ray photon energies and pulse durations on a shot-to-shot basis since the temporal structure of individual X-ray pulses are implicitly different from each other.

A detailed description of the temporal properties of European photon pulses and the requirements for the X-ray photon temporal diagnostics and candidate X-ray photon temporal diagnostic methods and techniques, including X-ray pulse length measurement and photon pulse arrival time measurement, can be found in Ref. [4].
2 Streaking electrons with THz radiation

Adapted from attosecond metrology, where the attosecond XUV pulses have been characterized by using a few-cycle near-infrared streaking pulse (Figure 2(a)) [10-12], the X-ray THz streaking technique is experimentally employed for measuring longer soft X-ray pulses on a shot-to-shot basis utilizing THz radiation from a THz undulator (U-THz) or an external laser-driven single-cycle THz source (L-THz) (Figure 2(b)) [13-16]. One needs to note that U-THz–based streaking does not permit one to access the timing information related to an external pump–probe (PP) laser system, while L-THz streaking overcomes the limitations and can provide the time information between PP laser and X-ray.

Figure 2: Schematic of (a) XUV pulse attosecond streaking and (b) X-ray pulse single-cycle THz streaking. Typical streaking spectrogram for attosecond and single-cycle THz streaking, respectively, are shown as insets.

Since the FEL pulse duration of the European XFEL is on the order of a few fs to 100 fs and typically has several tens of fs time jitter, long wavelength THz fields are chosen to fulfil the streaking criteria, i.e. the pulse length of the X-ray must be shorter than the half period of the streaking field. In principle, a longer streaking field would give rise to a longer linear streaking response and would be less sensitive to the X-ray time jitter.
Demonstration of a time-resolved THz streaking spectrogram is shown in Figure 3, with a streaking ramp of ~ 600 fs from the single-cycle THz field that is generated by optical rectification of femtosecond laser pulses [17-24]. As one can see from Figure 3, energy distribution of photoelectrons, liberated e.g. from noble gases under irradiation of X-ray pulses, can be broadened or shifted, depending on the relative time delay between the X-ray pulse and the THz field, i.e. the THz vector potential at the ionization time. The temporal properties of the X-ray pulse are thus mapped onto a photoelectron energy distribution that has an identical profile. By comparing the photoelectron energy distribution with and without an external THz field, one can in principle uniquely determine the X-ray pulse duration, temporal profile, and relative time delay to the THz field [14-16].

**Figure 3:** Demonstration of a scanned single-cycle THz streaking spectrogram, from which the X-ray pulse length, profile, and relative time delay can be derived.

In addition, this technique has been demonstrated to measure the X-ray chirp by either tuning the time delay between the X-ray and streaking pulse or by applying two electron time-of-flight spectrometers (eTOFs) that are parallel and anti-parallel to the streaking field [14,16].

The photoelectron streaking process can be described using a semi-classical model: X-ray photon absorption, photoelectron emission, and external field photoelectron streaking. Assuming a THz electric field described by $E_{THz}(t) = E_0(t)\cos(\omega_{THz}t+\phi)$ and assuming that the initial kinetic energy of the photoelectron without streaking is $W_0 = m_0v_0^2/2 = h\omega_x - I_p$, where $v_0$ is the photoelectron velocity and $\omega_x$ is the X-ray photon frequency, the modulated
photoelectron velocity in the presence of the THz field can be expressed as [16]:

\[ v(t) = -e/m_e A_t(t) + (v_0 + e/m_e A_i), \]

Here, \( A_t(t) \) is the vector potential of the THz field, and \( A_i \) is the instant vector potential at the ionization time. The external THz field modulates the kinetic energy and the propagation direction of the photoelectrons, whose momentum variation and final energy are given as:

\[
\Delta \vec{p}(t_i) = m_e (v - v_0) = e A_i
\]

\[
W_f = \sqrt{\left( \vec{P}_0 + \Delta \vec{p} \right)^2 / 2m_e} \left( \vec{P}_0 + \Delta \vec{p} \right)^2 / 2m_e.
\]

\( P_0 \) is the initial unstreaked momentum, and the final energy \( W_f \) can be expressed as:

\[
W_f = W_0 + 2U_p \cos 2\theta \cdot \sin^2 \varphi_i \pm \sqrt{\left( \frac{2U_p}{W_0} \right)^2 \sin^2 \theta \cdot \sin^2 \varphi_i \cdot \sqrt{8W_0 U_p \cos \theta \cdot \sin \varphi_i}}, \]

Here, \( \theta \) is the angle between the streaked electron velocity direction and the THz polarization direction, \( \varphi_i \) is the phase of the THz field at the ionization instant, and \( U_p \) is the ponderomotive energy of the THz field.

The above equation can be simplified to \( W_f \) when \( \theta \) equals 0 or 180 degrees, indicating that the final photoelectron velocity direction is either parallel or antiparallel to the THz field polarization direction.

\[
W_{f,\parallel} = W_0 + 2U_p \sin^2 \varphi_i \pm \sqrt{8W_0 U_p \sin \varphi_i},
\]

One can further simplify Eq. (4) by considering the fact that \( U_p \) in the THz region is negligible compared with \( W_0 \):

\[
W_{\text{streaking}} = W_{f,\parallel} - W_0 = \pm \sqrt{8W_0 U_p \sin \varphi_i} = \pm e \sqrt{\frac{2W_0}{m_e} A_i},
\]

\( A_i \) is the instant vector potential when ionization happens. Eq. (5) gives the description of the relationship between the THz vector potential and a time-dependent streaked photoelectron energy modulation.
One can, in turn, derive the THz vector potential, waveform, and peak electric field strength by scaling the streaking spectrogram according to Eq. (5) [25]. The X-ray pulse duration and profile can be determined by analysing the photoelectron spectral bandwidth and structure in a shot-to-shot basis.

Streaking speed $S$ is usually defined as the width of the streaked photoelectron energy distribution over the temporal width of the X-ray pulse:

$$S = \frac{\Delta W_{\text{streaking}}}{\Delta t} = e \sqrt{\frac{2W_0}{m_e}} E_{\text{THz}},$$  \hspace{1cm} (6)

For the value used in Ref. [15], $E_{\text{THz}}=165$ kV/cm, $W_0=233$ eV (He 1s, 4.8 nm, 258 eV), a streaking speed of 0.149 eV/fs can be derived. Increasing THz field strength or streak electrons with higher initial energy can give higher streaking speeds and higher temporal resolution as a result.
3 THz generation by optical rectification in LiNbO₃

The most commonly used method for laser-based THz generation is optical rectification in a quadratic nonlinear medium, which originates from a non-zero second order nonlinear optical susceptibility $\chi^{(2)}$. When an ultrashort laser pulse passes through such non-centro-symmetric mediums, different frequency generation (DFG) within each pulse produces low frequency radiation, i.e. the THz wave, as shown in Figure 4(a). Suffering from the phase mismatch between the pump pulse and the THz wave, THz conversion efficiency remains low and the THz waves propagate along a Cherenkov cone as demonstrated in Figure 5(b). The angle $\theta_c$ is determined by:

$$\cos \theta_c = \frac{v_{thz}}{v_{pump}}.$$  \hfill (7)

J. Hebling et al. proposed a tilted pulse front pumping scheme for efficient THz generation in LiNbO₃ (LN) [17], which qualifies the phase-matching condition along the THz propagation direction since the group velocity of the optical pump is larger than that of the THz pulse. This technique solves the problem of rapid dephasing between the optical laser and the THz pulse. In comparison with the Cherenkov configuration in Figure 4 (b), the tilted pulse front excitation geometry generates a nearly collimated THz beam with high conversion efficiency and allows an increase of the THz power by simply increasing the pump cross section and pump power. Up to now, THz field strengths up to MV/cm have been achieved by using different pumping wavelengths, energies, pulse durations, and focusing geometries [19-22]. Note that the generated THz electric field is intrinsically CEP-stabilized, since the two frequencies in DFG carry the same CEP offset.
According to Eq. (7), for efficient THz generation, the LN crystal should be specifically cut for the matching velocity of the THz and pump pulse inside the crystal. Since the pulse velocities are inversely proportional to the corresponding refractive index, we calculate $\gamma \sim 63^\circ$ using typical values of $n_{\text{pump}} = 2.25$ for 800 nm and $n_{\text{THz}} = 4.96$ for LN [26]. So the pump laser group velocity component (red dashed arrow in Figure 5) in the THz propagation direction is equal to that of the THz radiation (purple).

**Figure 5:** Typical LN crystal geometry used in the THz generation prototype. THz is emitted perpendicular to the out-coupling surface.

For Small Quantum Systems (SQS), we plan to use this tilted pulse front-based single-cycle THz generation for THz streaking measurement for the purpose of making the apparatus effective, compact, and mobile. As pointed out above, unlike the undulator-based THz generation, which is synchronized to the electron beam [14], optical laser-driven THz streaking can provide...
additional timing information (arrival time, time jitter) related to the external optical pump and probe laser system [15].

3.1 THz generation setup

The Optical Laser (OL) group (WP78) at European XFEL provides the future PP lasers for all instrument groups at European XFEL, including SQS [27-29]. The PP laser beam is delivered to SQS for ultrashort optical laser, FEL pump and probe studies of atomic, molecular, and cluster dynamics. Currently, the 1030 nm Yb fiber amplifier and 800 nm non-collinear optical parametric amplifier (NOPA) systems are under development by WP78, with the main target characteristics listed in Table 2.

**Table 2: Target PP laser parameters for the European XFEL**

<table>
<thead>
<tr>
<th>PP laser</th>
<th>Basic property</th>
<th>Amplification scheme</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Repetition rate</td>
<td>Energy</td>
</tr>
<tr>
<td>800 nm</td>
<td>200 kHz</td>
<td>3 mJ</td>
</tr>
<tr>
<td></td>
<td>1 MHz</td>
<td>1 mJ</td>
</tr>
<tr>
<td></td>
<td>4.5 MHz</td>
<td>200 μJ</td>
</tr>
<tr>
<td>1030 nm</td>
<td>200 kHz</td>
<td>100 mJ</td>
</tr>
<tr>
<td></td>
<td>1 MHz</td>
<td>20 mJ</td>
</tr>
<tr>
<td></td>
<td>4.5 MHz</td>
<td>4 mJ</td>
</tr>
</tbody>
</table>

In brief, three fundamental repetition rates inside the burst—200 kHz with the highest pulse energies, 1 MHz with medium pulse energies, and 4.5 MHz with the lowest pulse energies at both 800 nm and 1030 nm—can be delivered. WP78 also provides a “pulse on demand” mode, i.e. the fundamental repetition rates can be further reduced by optical modulators with the same pulse energy. Future simultaneous delivery of 800 nm and 1030 nm to the SQS experiment hutch is planned.

In principle, PP lasers at either 800 nm or 1030 nm listed in Table 2 can be used for THz generation. Nevertheless, one should always keep in mind the minimal consumption of the PP laser for THz-based diagnostics since the PP
laser is mainly targeted for experiments. On the other hand, THz generation and streaking should be flexible for different repetition rates according to the SQS operation modes. Study of THz generation and THz streaking under different operation modes of the PP laser—i.e. its repetition rate, pulse duration, and pulse energy—are scheduled. However, a THz generation prototype has been already built and optimized by using a 3 mJ, 1 kHz, 100 fs commercial laser.

The prototype of THz source is schematically shown in Figure 6, which contains both THz generation and electro-optical sampling (EOS) THz diagnostics.

*Figure 6: Prototype of THz generation using tilted pulse front excitation and EOS THz profile detection*

A diffraction grating is used to tilt the pulse front of the pump laser pulse, and the diffracted beam is imaged onto the LN crystal by a 4-f telescope with adjustable focal length and a demagnification factor to achieve the velocity matching of the pump and THz pulses in the LN crystal [19]. The pump laser polarization, controlled by a half wave plate before the LN crystal, is paralleled to the crystal optical axis so as to maximize the THz generation efficiency.
Figure 7: Tilted pulse front setup for THz generation. The tilted pulse front is generated by a grating and imaging system, THz radiation is emitted perpendicular to the pulse front of the pump pulse, and a copper mirror with gold coating is applied for THz reflection and collimation.

The pulse front tilt angle $\gamma$ can be described as [19]:

$$
\tan \gamma = \frac{m \lambda_0}{n_p \beta_1 \cos \theta_d d},
$$

(8)

Here, $m$, $\lambda_0$, $\beta_1$, $\theta_d$, and $d$ is the diffraction order, central wavelength, magnification factor of the telescope, diffraction angle, and grating period, respectively. Angle $\gamma$ should have the same value as the LN cutting angle in order to achieving velocity matching.

Optimal THz beam and conversion efficiency can be achieved when the tilt angle $\theta$ of the grating image inside the LN crystal equals that of the pump pulse front $\gamma$.

$$
\tan \theta = n_{LN} \beta_2 \tan \theta_d
$$

(9)

$n_{LN}$ is the refractive index of the LN crystal. $\beta_2$ is the magnification factor for the grating image.
Figure 8 shows the calculated curves of the magnification factor as a function of diffraction angle for three different grating grooves 1500, 1800, and 2000 \( \text{l/mm} \) in the condition of an 800 nm pump pulse. The angle matching condition is fulfilled when \( \beta_1 \) equals \( \beta_2 \). As indicated in Figure 8, the magnification factor corresponding to the 1500, 1800, and 2000 \( \text{l/mm} \) gratings is 0.526, 0.584, and 0.623, respectively. Our prototype of the THz setup applies a 2000 \( \text{l/mm} \) grating and a telescope with a magnification factor of 0.5, which is not optimal but not far from optimum according to the calculation. Further optimization of THz output can be done by using different grooved gratings and different magnification factors.

\textbf{Figure 8:} Calculated magnifications of the pump for different grating grooves and grating image as a function of the diffraction angle under 800 nm excitation

On the other hand, one could think about producing THz radiation using a 1030 nm longer pulse since more pump power can be used. Similarly, 1030 nm with a longer pulse duration of sub-ps pumped THz generation scheme has been reported in Ref. [22]. For example, one can split 5 mJ out of 100 mJ at 200 kHz for THz generation, while delivering the remaining 95 mJ laser pulses to the NOPA for generating 800 nm. In principle, this has the advantage of operating the THz setup independently from the 800 nm PP laser, while it has the disadvantage of requiring accurate synchronization with the experimental PP laser. Preliminarily evaluation of the required energies
for efficient THz generation utilizing laser pulses from Table 2 can be found in Table 3.

**Table 3: Requirements for efficient THz generation and THz streaking by using the laser pulses listed in Table 2**

<table>
<thead>
<tr>
<th>PP laser</th>
<th>Basic property</th>
<th>Requirement</th>
<th>Expected THz field (kV/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>800 nm</td>
<td>Rep. rate 200 kHz</td>
<td>Energy 3 mJ</td>
<td>Duration 80 fs</td>
</tr>
<tr>
<td>1 MHz</td>
<td>Energy 1 mJ</td>
<td>Energy 80 fs</td>
<td>-----</td>
</tr>
<tr>
<td>4.5 MHz</td>
<td>Energy 200 μJ</td>
<td>Energy 80 fs</td>
<td>-----</td>
</tr>
<tr>
<td>1030 nm</td>
<td>Rep. rate 200 kHz</td>
<td>Energy 100 mJ</td>
<td>Energy 800 fs</td>
</tr>
<tr>
<td>1 MHz</td>
<td>Energy 20 mJ</td>
<td>Energy 800 fs</td>
<td>1–2 mJ, cryogenic cooling</td>
</tr>
<tr>
<td>4.5 MHz</td>
<td>Energy 4 mJ</td>
<td>Energy 800 fs</td>
<td>1 mJ, tight focus, cryogenic cooling</td>
</tr>
</tbody>
</table>

The same angle matching plot for 1030 nm is shown in Figure 9. The magnification factor corresponding to the 1500, 1800, and 2000 l/mm gratings is 0.60, 0.67, and 0.73, respectively.

**Figure 9: Calculated magnifications of the pump for different grating grooves and grating image as a function of the diffraction angle under 1030 nm excitation**
3.2 Considerations for further THz generation improvement

Optical rectification can be treated as different frequency generation between different spectral components, with the efficiency for phase-matched conditions described as follows [23]:

\[
\eta_{THz} = \frac{2\omega^2 d_{eff}^2 L^2 I}{\varepsilon_0 n_p^2 n_{THz}^2 c^3} \exp \left[ -\frac{\alpha_{THz} L}{2} \right] \frac{\sinh^2 \left[ \frac{\alpha_{THz} L}{4} \right]}{\left[ \frac{\alpha_{THz} L}{4} \right]^2},
\]

(10)

Here, \( \omega \) is the THz frequency, \( d_{eff} \) the effective nonlinear coefficient, \( I \) the intensity of the pump-pulse, \( \varepsilon_0 \) the vacuum permittivity, \( c \) the velocity of light in vacuum, \( L \) the length of the nonlinear crystal, and \( \alpha_{THz} \) the absorption coefficient of THz radiation, with the group refractive indexes of the pump \( n_p \) and THz \( n_{THz} \).

Eq. (10) can be simplified to Eq. (11) and Eq. (12) in case of low or large absorption [23]:

\[
\eta_{THz} = \frac{2\omega^2 d_{eff}^2 L^2 I}{\varepsilon_0 n_p^2 n_{THz}^2 c^3},
\]

(11)

\[
\eta_{THz} = \frac{8\omega^2 d_{eff}^2 I}{\varepsilon_0 n_p^2 n_{THz}^2 c^3 \alpha_{THz}^2}.
\]

(12)

Figure of merit (FOM) of nonlinear crystal that characterizes the relative energy conversion efficiency for optical rectification is defined as follows for low or large absorption crystal:

\[
FOM = \frac{d_{eff}^2}{n_p^2 n_{THz}^2} \quad \quad FOM_A = \frac{4d_{eff}^2}{n_p^2 n_{THz}^2 \alpha_{THz}^2}.
\]

(13)

Table 4 gives the relevant parameters for commonly used optical rectification crystals: LiNbO\(_3\), DAST, and ZnTe. Note that the energy conversion efficiency is proportional to FOM.
Table 4: Properties of selected optical rectification materials: $r =$ electro-optical coefficient, $d_{\text{eff}} =$ nonlinear optical coefficient for optical rectification, $n_{\text{r}800} =$ group refractive index at 800 nm, $n_{\text{THz}} =$ refractive index at 1 THz, $\alpha_{\text{THz}} =$ THz absorption coefficient, $\text{FOM/FOM}_A =$ figure of merit.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>$r_{\text{eff}}$ (pm/V)</th>
<th>$d_{\text{eff}}$ (pm/V)</th>
<th>$n_{\text{r}800}$</th>
<th>$n_{\text{THz}}$</th>
<th>$\alpha_{\text{THz}}$ (cm$^{-1}$)</th>
<th>FOM (pm$^2$/V$^2$)</th>
<th>FOM$_A$ (pm$^2$cm$^2$/V$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LN(300K)</td>
<td>30.9</td>
<td>168</td>
<td>2.25</td>
<td>4.96</td>
<td>17</td>
<td>1170</td>
<td>18.2</td>
</tr>
<tr>
<td>LN(100K)</td>
<td>30.9</td>
<td>168</td>
<td>2.25</td>
<td>4.96</td>
<td>4.8</td>
<td>1170</td>
<td>48.6</td>
</tr>
<tr>
<td>DAST</td>
<td>77.0</td>
<td>618</td>
<td>3.39</td>
<td>2.58</td>
<td>150</td>
<td>28000</td>
<td>4.8</td>
</tr>
<tr>
<td>ZnTe</td>
<td>4.04</td>
<td>68.5</td>
<td>3.13</td>
<td>3.17</td>
<td>1.3</td>
<td>180</td>
<td>426</td>
</tr>
</tbody>
</table>

As one can see from Table 4, organic electro-optic crystals like DAST have very large second order nonlinear electric susceptibilities and have recently become promising sources of THz generation. The first THz generation from DAST was reported in Ref. [30], where a THz generation with 20 THz bandwidth from a 100 μm DAST crystal was reported. In spite of the extraordinarily large FOM value of DAST, this material is less attractive than LN since the effective length of DAST is only several tens of micrometers. On the other hand, the multi-cycle THz field generated by DAST is more complicated in both temporal and frequency domains as compared with the single-cycle THz from LN [30, 31]. Besides, organic crystals have a large natural birefringence, which complicates their application. However, A.L. Cavalier successfully demonstrated [to be published] THz streaking measurement at LCLS at 120 Hz using THz radiation from DAST.

For the THz generation prototype and THz streaking at SQS, we will mainly focus on THz generation based on tilted pulse front excitation in LN and will later explore the possibility of using organic crystals like DAST, pumped by near-infrared laser pulses (1.3–1.5 μm, model: HE-TOPAS-prime). However, thermal concentration–induced damage may happen in organic crystal due to the high repetition rate of the PP laser.

Optical resistance to laser damage of congruent LiNbO$_3$ (cLN) and stochiometric LiNbO$_3$ (sLN) is greatly improved by MgO doping, typically 6.0 mol % for cLN and 1.0 mol % for sLN to avoid photo-refraction and reduce
THz absorption. Previously, sLN doped with 0.6/1.3 mol % MgO and cLN doped with 6.0 mol % from different manufacturers were used for THz generation [19, 20, 22].

Currently, it is difficult to grow large-size, high-quality sLN, while relatively larger cLN crystal is available. The difference between sLN and cLN can be found in Ref. [32], from which the absorption curve of cLN and sLN in temperatures of 10 K and 300 K are shown in Figure 10. It is evident that the sLN absorption coefficient is much smaller than that of its cLN counterpart in the frequency region above 70 cm\(^{-1}\) (2.1 THz), while it becomes comparable at 10 K and 300 K in the lower frequency region (< 2.1THz). Below 1 THz (33.3 cm\(^{-1}\)), the larger cLN crystal could be used since it potentially supports larger pump spot sizes and higher pumping energies. For the prototype of THz generation, we compared the performance of cLN (6.0 mol % doping) and sLN (0.6 mol % doping) at room temperature; the results are shown later.

**Figure 10:** Comparison of the absorption coefficient of MgO-doped cLN and sLN at (a) 10 K and (b) 300 K

THz pulse energy conversion efficiency by pumping a 300 K room-temperature LN crystal remains so far as low as 0.25%. Recent theoretical and experimental studies show that THz generation in LN can be further improved by optimizing longer transform-limited pump–pulse duration (~ 500 fs) and reducing the photorefractive losses in LN crystals by cooling down to cryogenic temperatures [22]. Three to four times higher conversion efficiency was experimentally demonstrated. A cooling Dewar with ~ 100 K cooling capability and a small crystal chamber for protection of the cooled crystal against humidity damage by condensation are constructed.
Furthermore, Fresnel reflection losses (45%) at the input surface of the crystal should be taken into account, as the refractive index of LN is much larger than the refractive index of air in the THz region (~ 5.0 at 1 THz). Anti-reflection (AR) coating on the incident surface results in higher THz output. A three-sided polished LN prism geometry, illustrated in Figure 11, has been used for the THz generation prototype. The optical axis of the LN crystal is parallel to the polarization direction of the pump source, with two calculated angles of 63 degree and AR coating on the incident surface.

Figure 11: LN geometry for THz generation prototype

To make the THz streaking more applicable, this setup should give an optimal THz beam and a maximized THz peak field under a certain fixed amount of excitation energy, thus a detailed analytical study of the tilted pulse front–induced THz generation should be performed, with the main issues listed as follows:

1. Test pump intensity dependence to understand the LN saturation behaviour.
2. Test THz spot size as a function of pump spot size, to figure out how the THz beam profile changes with respect to the pump spot size and shape.
3. Try different imaging approaches to see how different imaging methods affect the THz efficiency, beam profile, and divergence angle.
Similar to the imaging scheme listed in Ref. [21]:

a One lens with plane convex lens
b One lens with plane convex achromatic lens
c Two-lens telescope with spherical lenses
d Two-lens telescope with cylindrical lenses
e Telescope with reflective optics

4 Figure out how imaging aberrations affect the THz generation efficiency. Programmable deform mirror can be applied for manipulating the imaging aberrations.

5 Compare different crystals with different doping and geometry from different manufacturers.

6 Study pump pulse temporal profile dependence, i.e. pulse duration, linear and higher order chirp.

7 Study pump pulse spectral profile dependence, as the optical rectification originated from the DFG of different spectral elements of the pump. Either simple nonlinear effect–induced spectral modulation or spectral domain shaping could be applied.

8 Test THz generation with a different pump source with a different repetition rate, wavelength, and spectral bandwidth.

9 Compare THz generation with and without cryogenic cooling.

3.3 THz radiation characterization

As discussed in Section 2, the time-dependent streaking photoelectron energy modulation by the external THz field is closely related to the THz electric field strength and its vector potential. The THz pulse properties like pulse energy, cross section profile, divergence angle, polarization, pulse shape and duration, and field strength should be known before a THz streaking experiment is performed.
The pulse energy of the THz radiation is measured by a calibrated pyroelectric detector (MicroTech Instruments, 0.02–3 THz). A 2 mm thick Teflon plate is placed in front of the detector and serves as a spectral filter that blocks the excitation pulse. The modulated voltage signal of the detector is monitored by an oscilloscope, and the THz energy is calculated from the amplitude of the THz-induced voltage modulation.

As schematically shown in Figure 12, electro-optical sampling (EOS) is used to measure the time-dependent THz electric field.

*Figure 12:* Balanced detection scheme for electro-optical sampling. THz and probe pulses are spatially and temporally overlapped in an EO crystal, and the unbalanced probe signal is analysed by a quarter–wave plate and polarizer.

Briefly, the THz beam is collimated by a Teflon lens and then focused by a 90 degree off-axis parabolic mirror to an EO detection crystal. The probe beam, whose temporal decay in respect to the THz radiation is controlled by a delay stage, is focused onto the EO crystal, with its focus spatially overlapped to that of the THz beam. A quarter–wave plate (λ/4) is used to generate circularly polarized probe light. A polarizer is used to separate the two equal orthogonal components, whose intensities are recorded by two identical photodiodes A and B. THz can imprint a birefringence on the EO crystal that changes the polarization of the probe beam. Phase modulation of the probe beam is converted to an intensity modulation of the two orthogonal polarized
components, thus one can determine the phase shift as well as the peak electric field of the THz pulse from the modulation depth of the balanced photodiodes A and B in the EOS system by [24]:

\[
\frac{A - B}{A + B} = \frac{2\pi}{\lambda} n_0^3 r_{41} E_{THz} L
\]

(14)

where \( n_0, r_{41}, \) and \( L \) present the refractive index, electro-optical coefficient, and thickness of the EO crystal.

**Table 5:** Properties of typical EO materials: \( r = \) electro-optical coefficient, \( \text{GVM} = \) group velocity mismatch.

<table>
<thead>
<tr>
<th>Material</th>
<th>( r_{41}(\text{pm/V}) )</th>
<th>Surface orientation</th>
<th>( \text{GVM}(\text{ps/mm}) )</th>
<th>Experimental bandwidth (THz)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZnTe</td>
<td>4.04</td>
<td>110</td>
<td>1.1</td>
<td>2.5</td>
</tr>
<tr>
<td>GaP</td>
<td>0.97</td>
<td>110</td>
<td>—</td>
<td>7.0</td>
</tr>
<tr>
<td>GaSe</td>
<td>14.4</td>
<td>—</td>
<td>0.10</td>
<td>14.4</td>
</tr>
</tbody>
</table>

An important condition for detection of THz radiation is the phase matching between the optical probe and THz inside the EO crystal. Crystals with a larger EO coefficient and a lower group velocity mismatch (GVM) are highly appreciated, and typical EO crystals are listed in Table 5. Moreover, detectability of an EO crystal is reversely proportional to its thickness, according to Eq. (14). A 200 \( \mu \)m thick GaP with a detection range of 425 kV/cm is chosen for THz EO sampling.

An off-axis parabolic mirror is used to focus the THz radiation, whose peak field strength is closely related to the focused THz beam size. We pre-aligned the parabolic mirror by using a collimated visible alignment laser (He-Ne, 632.8 nm). The alignment of the Teflon lens can be done by maximizing the voltage modulation in Pyroelectric detector located at the focal point of the parabolic mirror.

EO sampling gives accurate spatial and temporal overlap between the THz and probe and is extremely important in the streaking measurement. A typical single-cycle THz waveform and its converted vector potential and spectrum
are shown in Figure 13. Optimization schemes proposed in Section 3.1 will improve the THz peak electric field strength and shorten the rise time of the THz vector potential. The results in Figure 13 show a single-cycle THz pulse with a period of ~4 ps, centred at 0.5 THz with a vector potential rise time of 700 fs.

**Figure 13:** Typical (a) single cycle THz waveform, (b) vector potential, and (c) frequency spectrum detected by EO-sampling
4 Temporal resolution of THz streaking

4.1 Definition of temporal resolution

Temporal resolution $\tau$ is an important parameter of THz streaking, and it is usually defined as the minimal temporal intervals that can be resolved by streaking. $\tau$ is strongly related to THz field strength, the bandwidth of the FEL pulse, and the energy resolution of the eTOF spectrometer.

Streaking speed $S$ is defined in Eq. 6 as the width of the streaked photoelectron energy distribution over the temporal width of the X-ray pulse. The temporal resolution can thus be calculated as:

$$\tau = \frac{\Delta W_{\text{unstreaked}}}{S} = \frac{\Delta W_{\text{unstreaked}}}{e \sqrt{\frac{2W_0}{m_e} E_{\text{THz}}}}. \quad (15)$$

Eq. 15 indicates the dependence of temporal resolution $\tau$ on the bandwidth of the unstreaked photoelectrons $\Delta W_{\text{unstreaked}}$, THz field strength $E_{\text{THz}}$, and initially photoelectron energy $W_0$. Ultimately, temporal resolution is limited by $\Delta W_{\text{unstreaked}}$, which is the convolution of the initial X-ray bandwidth, the natural width of the photoelectron lines, and the energy resolution of the eTOF spectrometer. The measurement in Ref. [15] was performed with a 165 KV/cm THz field, and the bandwidth of the field-free photoelectrons is 7.2 eV, with a nearly Gaussian shape. The eTOF spectrometer resolution was calculated to be 6.75 eV by deconvoluting the X-ray bandwidth (2.5 eV), and the corresponding temporal resolution is estimated to be 45.3 fs [15, 33]. For a fixed eTOF spectrometer, the temporal resolution can be improved by either applying a more intense THz field or streaking more energetic photoelectrons. It is stated that the temporal resolution could be further improved to $\sim$ 10 fs.
For SQS instrumentation, pulses with duration as short as 2 fs in the low charge mode (20 pc) or up to 100 fs in the nominal bunch charge mode (1 nc) can be selected (see Table 1). The development of THz streaking for SQS is expected to be versatile in the broad range of X-ray photon energies (250 eV – 3.0 keV) and to have the flexibility of measuring X-ray pulse lengths under different operation modes.

### 4.2 Candidate sample under different SQS photon energy and required energy resolution of eTOF spectrometer

Table 6 from Ref. [3] lists all relevant potential atomic energy levels of the rare gases that can be used for streaking in the SASE3 energy region from 250 to 3000 eV. The binding energy, photoionization cross section, and corresponding photoelectron energy for different energy sub-regions are listed.
Table 6: Summary of the core levels of the rare gases Neon (Ne), Argon (Ar), Krypton (Kr), and Xenon (Xe) in photon energy ranges of SASE3. Combinations of the highlighted atomic orbitals are suggested for photon diagnostics in order to satisfy the requirements discussed in the text.

<table>
<thead>
<tr>
<th>SASE 3 0.25 - 3 keV</th>
<th>Photon energy range</th>
<th>Atomic level</th>
<th>Binding energy</th>
<th>Photoionization cross section</th>
<th>Kinetic energy</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>keV</td>
<td></td>
<td>eV</td>
<td>Mb = 10^{-22} m²</td>
<td>eV</td>
</tr>
<tr>
<td>Min</td>
<td>Max</td>
<td>From</td>
<td>To</td>
<td>Min</td>
<td>Max</td>
</tr>
<tr>
<td>0.25</td>
<td>0.9</td>
<td>94</td>
<td>95</td>
<td>4</td>
<td>0.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Xe 4s</td>
<td>213</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Kr 3p</td>
<td>214</td>
<td>0.2</td>
<td>0.8</td>
</tr>
<tr>
<td>0.28</td>
<td>0.9</td>
<td>251</td>
<td>248</td>
<td>3</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Kr 3s</td>
<td>293</td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Ar 2s</td>
<td>326</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>0.7</td>
<td>1.75</td>
<td>676</td>
<td>689</td>
<td>3</td>
<td>&lt;0.5</td>
</tr>
<tr>
<td>0.9</td>
<td>1.75</td>
<td>870</td>
<td></td>
<td>0.3</td>
<td>&lt;0.07</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Xe 3p</td>
<td>1002</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Xe 3s</td>
<td>1149</td>
<td>0.06</td>
<td></td>
</tr>
<tr>
<td>1.75</td>
<td>3.25</td>
<td>1678</td>
<td>1730</td>
<td>0.6</td>
<td>72</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Kr 2s</td>
<td>1921</td>
<td>0.06</td>
<td></td>
</tr>
<tr>
<td>3.25</td>
<td>Ar 1s</td>
<td>3206</td>
<td></td>
<td>0.075</td>
<td>44</td>
</tr>
</tbody>
</table>

The energy shells proposed for diagnostics are highlighted in Table 3. Proper gas targets and energy shells can be chosen according to the applied X-ray photon energy. Based on the samples and energy shells listed in Table 6, we estimate the streaking speed $S$ and required related eTOF spectrometer energy resolution for a given pulse duration. Here, the peak THz E-field strength is set to 300 kV/cm; the results are listed in Table 7.
Table 7: Calculated streaking speed $S$ (eV/fs) and corresponding required eTOF spectrometer resolution $\delta_{\text{eTOF}}$ (eV) and $\delta_{\text{eTOF}}/W_0$ for given X-ray pulse parameters: photon energy, pulse length: 10 fs, 100 fs. The proper energy shell and its binding energy and natural bandwidth for different photon energy region are listed. $W_0$ is the unstreaked photoelectron energy.

<table>
<thead>
<tr>
<th>$E_{\text{x-ray}}$ (eV)</th>
<th>Target and parameter</th>
<th>$E_{\text{binding}}$ (eV)</th>
<th>$W_0$ (eV)</th>
<th>$S$ (eV/fs)</th>
<th>Pulse length (fs)</th>
<th>X-ray bandwidth (eV)</th>
<th>Resolution limit (fs)</th>
<th>$\delta_{\text{eTOF}}$ (eV)</th>
<th>$\delta_{\text{eTOF}}/W_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>250</td>
<td>Kr 3d</td>
<td>0.101</td>
<td>94</td>
<td>155</td>
<td>0.222</td>
<td>10</td>
<td>1.80</td>
<td>8.1</td>
<td>2.22</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.101</td>
<td>95</td>
<td>805</td>
<td>0.505</td>
<td>100</td>
<td>1.60</td>
<td>7.2</td>
<td>22.2</td>
</tr>
<tr>
<td>900</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>100</td>
<td>4.50</td>
<td>8.9</td>
<td>5.05</td>
</tr>
<tr>
<td></td>
<td></td>
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<td></td>
<td>100</td>
<td>3.50</td>
<td>6.9</td>
<td>50.5</td>
</tr>
<tr>
<td>280</td>
<td>Ar 2p</td>
<td>0.128</td>
<td>251</td>
<td>30</td>
<td>0.097</td>
<td>10</td>
<td>1.81</td>
<td>18.8</td>
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</tr>
<tr>
<td></td>
<td></td>
<td>0.128</td>
<td>248</td>
<td>650</td>
<td>0.454</td>
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<td>4.50</td>
<td>9.9</td>
<td>4.54</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>100</td>
<td>3.50</td>
<td>7.7</td>
<td>45.4</td>
</tr>
<tr>
<td>700</td>
<td>Xe 3d</td>
<td>0.490</td>
<td>676</td>
<td>34</td>
<td>0.104</td>
<td>10</td>
<td>2.93</td>
<td>28.2</td>
<td>1.04</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.510</td>
<td>689</td>
<td>1084</td>
<td>0.586</td>
<td>100</td>
<td>2.60</td>
<td>25.0</td>
<td>10.4</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td>100</td>
<td>5.30</td>
<td>9.0</td>
<td>58.6</td>
</tr>
<tr>
<td>1750</td>
<td>Ne 1s</td>
<td>0.24</td>
<td>870</td>
<td>30</td>
<td>0.097</td>
<td>10</td>
<td>4.50</td>
<td>43.4</td>
<td>0.97</td>
</tr>
<tr>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>100</td>
<td>5.30</td>
<td>10.0</td>
<td>52.8</td>
</tr>
<tr>
<td>3250</td>
<td>Kr 2p</td>
<td>1.31</td>
<td>1678</td>
<td>72</td>
<td>0.151</td>
<td>10</td>
<td>6.65</td>
<td>44.0</td>
<td>1.51</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.17</td>
<td>1730</td>
<td>1572</td>
<td>0.705</td>
<td>100</td>
<td>5.30</td>
<td>35.1</td>
<td>15.1</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>100</td>
<td>7.95</td>
<td>11.3</td>
<td>70.5</td>
</tr>
</tbody>
</table>
Taking the recommended energy shells in a different energy region, the streaking speed $S$, required eTOF spectrometer absolute energy resolution $\delta_{e\text{TOF}}$, and relative energy resolution $\delta_{e\text{TOF}}/W_0$ were calculated for X-ray pulse lengths of 10 and 100 fs. For example, a spectrometer with an absolute energy resolution of 7.05 eV and a relative energy resolution $\delta_{e\text{TOF}}/W_0$ of $4.50\times10^{-3}$ can achieve ~ 10 fs temporal resolution with a ~ 3 keV X-ray pulse. However, as mentioned above, the temporal resolution not only depends on the energy resolution of the eTOF spectrometer but on the natural bandwidth of the chosen target energy shell and the X-ray bandwidth. One can find the resolution limit introduced by the X-ray bandwidth and natural linewidth in Table 7.

Apart from direct X-ray photoemission lines, one can streak Auger electrons that are emitted in addition to an inner-shell photoelectron [27]. An Auger electron is generated from re-filling the core hole by a bound electron with a lower binding energy, and the Auger electron carries the result-in energy release. Since the linewidth of Auger electrons is only determined by the lifetime of the inner-shell vacancy and is independent from the X-ray photon energy and energy jitter, it is promising to improve the temporal resolution of THz streaking by streaking Auger electrons. Actually, laser-assisted Auger decay for the temporal characterization of X-ray pulses was already demonstrated at LCLS [36]. Selected Auger lines within the SASE3 photon energy range are listed in Table 8. Auger electron streaking curves should exhibit a similar streaking structure compared to that of the photoelectron, and the maximum energy shift and intensity are determined by the initial Auger electron energy and the probability of Auger emission [35-37].

Note that narrow Auger lines give rise to a higher temporal resolution, while on the other hand they bring about uncertainties due to the Auger decay with a typically few fs lifetime (Table 8). The measured X-ray temporal profile is no longer the actual temporal profile but the convolution of the temporal profile with the Auger decay time, which would bring additional ambiguity to the analysis of few fs (~ 5 fs) X-ray pulse. The high average brilliance of the European XFEL makes single-shot measurements possible even though the cross sections of Auger lines are much smaller than those of the
photoelectron lines. However, one has to be aware that the Auger lines are quite often a very complicated and complex “group of lines” and have certain overlap with the photoionization lines, making the analysis of any streaking spectrum with additional shift and broadening quite difficult to unravel. On the other hand, streaking Auger electrons do not allow for a determination of the X-ray chirp, since it is in general not sensitive to the X-ray photon energy and the corresponding temporal changes in energy, i.e. the chirp.

Table 8: Auger lines within the SASE3 photo energy range suggested for streaking. The data of the Auger lines are taken from [35-37].

<table>
<thead>
<tr>
<th>Atomic level</th>
<th>Interpretation</th>
<th>Energy (eV)</th>
<th>Natural width (eV)</th>
<th>Lifetime (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kr 3d</td>
<td>M2N1N23</td>
<td>37.67</td>
<td>0.101</td>
<td>5.47</td>
</tr>
<tr>
<td>Kr 3d</td>
<td>M2N1N23</td>
<td>38.71</td>
<td>0.101</td>
<td>5.47</td>
</tr>
<tr>
<td>Kr 3d</td>
<td>M3N23N23</td>
<td>51.15</td>
<td>0.101</td>
<td>5.47</td>
</tr>
<tr>
<td>Ar 2p</td>
<td>L3M23M23</td>
<td>201.09</td>
<td>0.128</td>
<td>4.31</td>
</tr>
<tr>
<td>Ar 2p</td>
<td>L3M23M23</td>
<td>203.47</td>
<td>0.128</td>
<td>4.31</td>
</tr>
<tr>
<td>Ne 1s</td>
<td>K1L1L23</td>
<td>804.0</td>
<td>0.24</td>
<td>2.30</td>
</tr>
<tr>
<td>Kr 2p</td>
<td>L3M45M45</td>
<td>1513.2</td>
<td>1.31</td>
<td>0.42</td>
</tr>
</tbody>
</table>

4.3 Gouy phase shift–induced ambiguity

As pointed out above, tight focus can be applied to generate high electric field THz and to improve the temporal resolution. However, a focused Gaussian beam acquires a phase shift that differs from that for a plane wave with the same frequency, i.e. the so-called Gouy phase shift and is given by [38]

$$\phi(z) = \arctan \left( \frac{z}{z_R} \right)$$

(16)

where \(z_R\) is the Rayleigh length and \(z = 0\) is the position of beam waist. The overall Gouy phase shift of a Gaussian beam before and after the focus is \(\pi\). Spatial intensity distribution of a focused Gaussian beam and Gouy phase shift are shown in Figure 14, where 0.5 THz, a beam diameter of 20 mm, and a focal length of 100 mm are assumed for the calculation.
Figure 14: Calculated focused THz intensity distribution, beam diameter (white-solid curves), and the additional Gouy phase cross the THz focus (red-dashed curve)

As shown in Figure 14, a tight focus of the THz beam results in short Rayleigh lengths of few mm, and the photoelectrons generated in different interaction regions experience different THz phase owing to the contribution of Gouy phase. As a result, the streaked photoelectron spectra show an additional broadening $\delta_{\text{Gouy}}$ from the phase difference $\Delta \phi$, which is mainly determined by the size $\Delta s$ of the gas target, shown in Eq. 17,

$$
\tau_{\text{Gouy}} = \sqrt{\tau_{\text{FEL}}^2 + \delta_{\text{Gouy}}^2} \quad (17)
$$

$$
\delta_{\text{Gouy}} = \frac{\Delta \phi}{\omega} = \frac{\lambda}{2\pi c} + a \tan\left(\frac{\Delta s \cdot \lambda}{\pi \omega_0^2}\right) \cdot \frac{\lambda}{2\pi c}
$$

where $\omega$ and $\lambda$ are the THz frequency and wavelength, respectively, and $\omega_0$ is the THz beam waist.

As one can see from Eq. 17, when $\Delta s$ is small, $\delta_{\text{Gouy}}$ increases linearly with the gas target size, while it increases quadratically with the wavelength. The effect of the Gouy phase shift on the pulse length measurement are calculated and listed in Table 9, where three THz streaking fields with different frequencies and three interaction lengths are used for calculation.
Table 9: Calculated Gouy phase shift–induced additional broadening on the pulse length measurement. A THz steaking field with different peak frequency and three different interaction lengths are assumed for the calculation.

<table>
<thead>
<tr>
<th>THz frequency (THz)</th>
<th>Wavelength (μm)</th>
<th>$z_R$ (mm)</th>
<th>Sample size (μm)</th>
<th>$\delta_{\text{Gouy}}$ (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>600</td>
<td>8.85</td>
<td>150</td>
<td>5.40</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>250</td>
<td>8.99</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>500</td>
<td>17.96</td>
</tr>
<tr>
<td>0.6</td>
<td>500</td>
<td>10.62</td>
<td>150</td>
<td>3.75</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>250</td>
<td>6.24</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>500</td>
<td>12.48</td>
</tr>
<tr>
<td>1.0</td>
<td>300</td>
<td>17.70</td>
<td>150</td>
<td>1.35</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>250</td>
<td>2.25</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>500</td>
<td>4.49</td>
</tr>
</tbody>
</table>

Gouy phase–induced broadening is typically on the order of a few fs or few tens of fs. To reduce the spectral broadening that is caused by the Gouy phase shift, it is desirable to minimize the interaction length. This could be achieved with a narrow high-density gas target and a restricted eTOF acceptance angle. Besides, high frequency THz radiation is highly demanding for measuring short X-ray pulses with a few fs pulse length.
5 Requirements

In general, the THz streaking timing tool should contain the following parts, as shown in Figure 15:

1. Optical laser split and delay
2. THz generation setup
3. THz beam transport
4. THz EO sampling
5. Sample delivery
6. Data acquisition (DAQ) and data analysis (DAN)
Figure 15: Possible design that fits the requirement of the SQS instrumentation. The whole setup contains six parts: optical laser beam transport, split and delay, THz generation, THz beam transport, THz pulse characterization, sample delivery, and data acquisition and analysis.
5.1 Electron time of flight spectrometer (eTOF)

The eTOF spectrometer is a standard tool in atomic and molecular physics to study and probe the electronic structure of a target. In the case of THz streaking, it is used to measure the kinetic energies of the photoelectrons generated by the X-ray pulse and accelerated or decelerated by the THz field. Typically, electrostatic lenses and retardation potentials are used to increase the transmission and temporal resolution, since the energy resolution of the eTOF is inversely related to the flight time. After propagation through a flight tube, the electrons hit an MCP detector, which is connected to a capacitor, and the produced charge cloud can be recorded by an oscilloscope or other digitization device [39].

The requirements for the eTOF in Ref. [3] are also applicable here:

1. Non-invasive operation
2. Suitability for temporal structure of the European XFEL’s radiation
3. Universal applicability for all SASE3 photon energies (0.25–3.0 keV)
4. High energy resolution $\Delta E/W_0 \sim 5 \times 10^{-4}$
5. Shot-to-shot capability
6. Low response time
7. High-speed data acquisition and processing

Candidate eTOF spectrometer:

1. ETF20 high-energy eTOF spectrometer
2. eTOF spectrometer from X-ray FEL photoelectron spectrometer (PES, Cookie box) [3]
3. eTOF spectrometer from AQS chamber of SQS instrument
5.2 Sample delivery

As one can directly see from Table 6, the X-ray photoionization cross sections decrease as the X-ray photon energy increases. For example, the cross section of Kr 3d under photon energies of 250 is 20 times higher than that of 900 eV. Thus a special method of compensation at higher photon energies should be considered. Piezo-drive valves [40] used in SQS/PSI are proven to be an effective solution that increases the sample density in the interaction region, while at the same time decreasing the gas load in the streaking chamber. The pulsed valve can be operated at 10 Hz with an opening time of at least 600 μs. Such a pulsed valve should be mounted with a xyz translation stage (xyz adjustable, ~ 25 μm spatial resolution) for optimization of photoionization yield with feedback from the eTOF spectrometer. As indicated in Table 9, the size of the gas target inside the interaction region should be as small as possible to reduce the uncertainty induced by the Gouy phase.

5.3 Optical laser and THz delivery

THz streaking occurs downstream of the KB focusing mirror and NQS experiment chamber of SQS. The KB mirror delivers focused X-ray beam to the experiment chamber, i.e. NQS and AQS [5]. This is why one has to refocus the X-ray beam to the THz streaking chamber with a refocusing mirror. As seen from Figure 15, one can put the THz generation and THz EO sampling on a customized optical breadboard that is adjacent to the vacuum chamber and transport the THz beam into the vacuum chamber. The THz beam is focused by the off-axis parabolic mirror to the interaction region. The refocused X-ray beam propagates collinearly with the focused THz streaking beam, schematically shown in Figure 16. One has to note that the presence of water vapour is a problem for THz streaking because it changes the THz waveform and spectrum. THz generation and transport are planned to be put in a sealed box and purged with dry nitrogen when finishing optimization to reduce water vapour. Nitrogen is chosen as the purge gas since it does not absorb THz in the THz region generated here.
5.3.1 THz polarization

Maximum streaking happens only when the THz field and the X-rays have the same polarization, and the pulse duration can be derived when comparing the photoelectron spectra with and without the THz field. However, the THz pulse, generated by the setup shown in Figure 7, is p-polarized, while the X-ray pulse is s-polarized and one has to rotate the THz polarization by 90 degrees. A periscope with the geometry shown in Figure 17 has been designed and built.
5.3.2 THz focus

The off-axis parabolic mirror should be mounted on a multi-dimensional adjustable mount for fine-tuning the spatial overlap between X-ray, THz, and EO sampling probe beam. The spatial overlap should be optimized to have the maximum THz electric field and the highest temporal resolution.
5.3.3 THz focusing lens

The most commonly used THz lens is made from polytetrafluoroethylene (PTFE, Teflon), which is a white solid at room temperature. The divergent THz radiation is collimated by a Teflon lens and coupled into the vacuum chamber. The refractive index of the Teflon material $n$ is important for designing the Teflon lens, as indicated in Eq. 18. Teflon lenses with a different focus length of 250, 300, 325, and 350 mm are prepared.

\[
\frac{1}{f} = (n-1) \left( \frac{1}{R_1} - \frac{1}{R_2} \right)
\]

\[
f = \frac{R_1}{n-1} \quad (R_2 \rightarrow \infty)
\]

Apart from the Teflon lens, a lens made from polymethylpentene (TPX) and cyclic olefin polymers (COP) can be used for THz focusing as well. The latter two exhibit higher THz transmittances, while, on the other hand, they are transparent to the optical excitation pulse, and a proper filter has to be used to filter out the excitation pulse.
5.3.4 THz EO-sampling inside vacuum

The EO sampling probe beam should be delivered to the interaction region via a preset optical window. EOS can give the spatial and temporal overlap of the THz and probe beam and will provide the precise position of THz focus. A 500 μm thick ZnTe and a 200 μm thick GaP EO crystal are mounted on a sample holder, together with a YAG screen and a fast diode for the purpose of spatial and temporal overlap, as shown in Figure 18. The EO sampling probe beam will transport out of the vacuum chamber via an optical window and detect near the setup as shown in Figure 19.

*Figure 19: EO sampling setup near the setup used for measuring the THz waveform and peak field strength and optimizing the streaking THz field.*

5.3.5 THz window

Optically, quartz is commonly used in the IR and mm wave regions of the spectrum as a vacuum window on instrumentation. One of the best materials for THz region is a z-cut quartz crystal. Since the z-cut quartz window is
transparent in the visible range, it is easy to adjust the whole system by using a visible laser, and the z-cut quartz does not change the polarization of the incident light pulse. Thus, the z-cut quartz window with ~80% transmittance is planned to be used as a THz window in the THz streaking chamber.

5.4 Control system and interfaces

Most of the items have to be motor-controlled since such devices are located in the experiment hutch and no access is allowed during experiments.

The following features are to be remotely accessible:

1. TPLM vacuum chamber positioning
2. eTOF spectrometer: voltages of MCP, electronic focusing, and retardation
3. Pulsed gas valve: position adjustment, opening time tuning, and gas samples switch
4. Coarse time photodiode readout
5. Spatial overlap image of the X-ray FEL and EOS on the scintillator
6. THz focusing off-axis parabolic mirror and the EO crystal holder
7. Interlocks to protect the eTOF spectrometer in emergency cases like vacuum leakage and overflow of the gas target

Hardware and software control can be adapted from existing devices in WP74 and SQS, such as the PES spectrometer, imager, and filter chamber. All electronic components that require control through the European XFEL DAQ and control system “Karabo” [41], or components that deliver data to this system, have to be validated by the DAQ and Control group of the European XFEL.
5.5 Data acquisition and analysis

TPLM involves different techniques of THz generation, characterization, and photoelectron streaking. Functioned as a temporal diagnostic tool, real-time data acquisition, analysis, and display, feedback of the pulse duration to the FEL machine and the calibration, display, and recording of THz peak electric field strength on a shot-to-shot basis have to be developed. We have already developed several customized analysis program for pulse length reconstruction, and, in the end, all these program have to be coupled into the European XFEL’s Karabo software. Meanwhile, SQS will be operated at the baseline repetition rate of 10 Hz with the flexibility to the burst mode operation of the highest repetition rate up to 4.5 MHz. One thus anticipates a high-speed data acquisition, storage, and system recovery cycle that enables single-shot measurement within the European XFEL intra-bunch pulse train.
A References


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