The DREAM Endstation at the Linac Coherent Light Source

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Abstract: Free-electron lasers (FEL), with their ultrashort pulses, ultrahigh intensities, and high repetition rates at short wavelength, have provided new approaches to Atomic and Molecular Optical Science. One such approach is following the birth of a photo electron to observe ion dynamics on an ultrafast timescale. Such an approach presents the opportunity to decipher the photon-initiated structural dynamics of an isolated atomic and molecular species. It is a fundamental step towards understanding single- and non-linear multi-photon processes and coherent electron dynamics in atoms and molecules, ultimately leading to coherent control following FEL research breakthroughs in pulse shaping and polarization control. A key aspect for exploring photoinduced quantum phenomena is visualizing the collective motion of electrons and nuclei in a single reaction process, as dynamics in atoms/ions proceed at femtosecond ($10^{-15}$ s) timescales while electronic dynamics take place in the attosecond timescale ($10^{-18}$ s). Here, we report on the design of a Dynamic Reaction Microscope (DREAM) endstation located at the second interaction point of the Time-Resolved Molecular and Optical (TMO) instrument at the Linac Coherent Light Source (LCLS) capable of following photon–matter interactions by detecting ions and electrons in coincidence. The DREAM endstation takes advantage of the pulse properties and high repetition rate of LCLS-II to perform gas-phase soft X-ray experiments in a wide spectrum of scientific domains. With its design ability to detect multi-ions and electrons in coincidence while operating in step with the high repetition rate of LCLS-II, the DREAM endstation takes advantage of the inherent momentum conservation of reaction product ions with participating electrons to reconstruct the original X-ray photon–matter interactions. In this report, we outline in detail the design of the DREAM endstation and its functionality, with scientific opportunities enabled by this state-of-the-art instrument.

Keywords: COLTRIMS; REMI; gas phase; CEI; TMO; AMO; LCLS II; FEL

1. Introduction

Ion and electron time-of-flight spectroscopy has enabled photoionization physics and the chemical dynamics of quantum systems by evolving to provide capabilities such as high energy resolution, high angular resolution of charged particles, and order-of-magnitude higher collection efficiency in the past few decades. Furthermore, advanced analysis through multi-particle covariance and coincidence methods of experimental data has provided insights into light–matter interactions across a broad excitation wavelength ranging from the infrared to X-ray photoionization regime. One such particular technique for coincidence measurements is the so-called cold-target recoil ion momentum spectroscopy (COLTRIMS) [1,2]. COLTRIMS is a state-of-the-art technique that is capable of reconstructing the momentum representation of the ions, electrons, atoms and molecules undergoing photon impact reactions. Thus, this is a powerful tool for following photoinduced reaction dynamics in the time domain for various photon-interacting systems. Another experimental technique based on coincidence measurement is Coulomb Explosion Imaging (CEI) [3].
Initially inspired by high-energy and collision physics, CEI enabled the study of molecular fragmentation pathways through further development of this experimental technique over time [4–6]. Recently, CEI and coincidence techniques found widespread application in the atomic physics community through the development of reaction microscopy technique enabled by COLTRIMS. One of the most significant advantages of a reaction microscope is the possibility of capturing all constituent ions in a photofragmentation process, including hydrogen atoms/ions. This is crucial for investigations of ultrafast photochemical dynamics involving multiple channels with various structural changes. Many pump-probe spectroscopic methods have investigated ultrafast photochemical reactions in the gas phase. Most of these spectroscopic methods are significantly more sensitive to ultrafast changes in a molecule’s electronic structure than its structural dynamics. However, the correlated nature of electronic and nuclear motion during non-adiabatic dynamics requires an equally detailed investigation of the structural dynamics. Novel femtosecond time-resolved imaging techniques enabled by ultrashort X-ray free-electron laser (XFEL) [7,8] and electron sources [9,10] have demonstrated the substantial benefit of direct measurement of the nuclear geometry evolution during non-adiabatic processes.

As mentioned, COLTRIMS and CEI are both equally capable of detecting light and heavy atoms involved in photoionization processes, providing information in momentum space in a molecular frame of reference. This differentiates COLTRIMS from other gas-phase X-ray or electron diffraction techniques in terms of spatial sensitivity to target size. Therefore, sensitive methods such as coincidence measurements can be crucial for investigations of ultrafast photochemical dynamics involving multiple channels with diverse structural changes [11–16]. To acquire sufficient statistics, coincidence methods require the detection of a large number of events. This requirement is increasingly met by high-repetition-rate optical laser systems and the next generation of XFELs, such as LCLS-II. Here, we present the design of the Dynamic Reaction Microscope (DREAM) endstation at the TMO instrument [17] located at LCLS-II, the 1 MHz repetition rate superconducting FEL at SLAC.

2. Hardware

In this section, we will describe the DREAM design parameters and the hardware layout of TMO’s flagship endstation. We will describe how we addressed the problems coming from a highly demanding experimental technique combined with the technical challenges posed at a high-repetition-rate FEL. The main idea of a coincidence experiment is to collect all fragments (molecules, atoms, and electrons) coming from the sample target, with only one target at the interaction volume per X-ray pulse. This drives most of the design specifications for such an endstation, such as vacuum conditions, sample delivery properties, and focii spot size for X-rays and optical laser. The following section will focus on the applied solutions for the DREAM endstation.

2.1. The TMO Instrument

The state-of-the-art TMO instrument is located at hutch 1.1 of the Near Experimental Hall (NEH) at LCLS. This instrument is designed to operate using two Kirkpatrick–Baez (KB) mirror systems [18] in an in-line refocusing configuration. Each of the two KB mirror systems—KB1 and KB2—is designed specifically to meet the key performance parameters of the corresponding focus spot, also known as interaction points (IP), for light–matter interactions. The DREAM endstation is located at the second IP, with a dedicated KB mirror (KB2) system located downstream of the first KB system. For DREAM, the incident angle of the KB2 system is 21 mrad with respect to the KB1 mirror surface. This incident angle allows for an X-ray energy range of 250–1400 eV. The TMO beamline operates in this soft X-ray regime without a monochromator. The active water-cooled KB mirror systems are designed to take in excess of 100 W average beam power. A detailed description of the TMO instrument can be found in [17], while a description of TMO’s KB system can be found in [19]. Figure 1 shows an overview of the TMO instrument layout.
2.2. The Main Chamber

The vacuum chamber of the DREAM endstation is designed to be modular, with one core chamber and a extension chamber, to be flexible for future upgrades. The main chamber is cylindrical in shape and is mounted horizontally, with its rotation axis being perpendicular to the incoming X-ray beam. The chamber has an inner diameter of 324 mm and it is made out of aluminum to reduce outgassing contamination [20]. This size of the inner diameter allows one to place all needed hardware and still be close enough to the KB system to allow for an nm-size X-ray focus spot. It features ports and feedthroughs for the X-rays, electronics, gauges, and vacuum pumps. Vertically attached to it on the opposing side from the optical laser input is the extension chamber, with the same inner diameter and chamber material. This extension chamber can be used as an adaptable housing for different lengths of spectrometers. It also consists of more pumping ports and feedthrough possibilities. The spectrometer and the detectors are connected to the main chamber via the side flanges, and the spectrometer slides in and out with a tailored installation mechanism.

The chamber itself is mounted to a height-adjustable stand, and since the optical laser mounts are mounted on the granite bellow and decoupled from the main chamber via bellows, the height adjustment only moves the spectrometer and the detectors for possible jet velocity adjustments. With this solution, the X-ray and optical laser alignment does not change, while the main chamber and spectrometer can move freely in height. Another advantage of decoupling the laser focusing optics and X-ray cleanup slits from the chamber is the removal of parasitic motion due to vacuum-applied stress on the chamber. Figure 2 shows the main DREAM chamber with all the above-described features.
Figure 2. Overview of the DREAM instrument, shown as endstation mounted on the support granite (a), and the chamber assembly without support infrastructure (b). Indicated are the CW supersonic jet (1), the three-axis jet manipulator (2), the first skimmer stage pumping (3), the holding frame for the skimmer section, which can be moved in tip and tilt (4), the Helmholtz coil pair (5), the long-range microscope (6), the main chamber (7), the close optical laser in-coupling next to the out-coupling (8), the two-stage catcher section with RGA (9), the three-axis coil mover (10), the support granite (11), and the extension chamber (12). Figure and caption adapted from [17].

2.3. The Spectrometer and Detectors

The spectrometer has an inner diameter of 120 mm and can be adjusted in length by adjusting the amount of spectrometer plates. On both ends of the spectrometer, there are two detector assemblies consisting of state-of-the-art 120 mm delay line detectors combined with 120 mm active area multichannel plates (MCP). The detector assemblies are commercially available products from RoentDek (delay lines, Type HEX120XL) and Photonis (MCP, type MCP 120/32/25/19). To accommodate the access of the sample and timing paddle, as well as an unobstructed entrance of the highly divergent optical laser beam and an unblocked field of view of the interaction region, the spectrometer has several cut-outs around the interaction point. Figure 3 shows the spectrometer setup and the detector assembly.
Figure 3. The DREAM spectrometer; indicated are the optical laser out-coupling (1), the detector HV feedthroughs (2), the optical laser in-coupling (3), the 120 mm delay line detector (4), the spectrometer blades with an inner diameter opening of 120 mm (5), the opening for the X-rays and optical laser (6), the spectrometer rail and support frame (7), the second 120 mm delay line detector (8), the second spectrometer flange (9), and the installation support rail guiding (10).

2.4. The Gas Jet and Skimmer Section

The main sample delivery scheme of the DREAM endstation is a CW supersonic gas jet. The key performance parameters for the standard configuration gas jet are given in Table 1. One of the main jet features is a two-nozzle setup. The basic idea is to have one line for the main sample, and the second gas line is for a calibration gas. This way, there is one line for a well-understood gas, which is used to align, calibrate, and find timing for any pump and probe experiments. This means that one has only to translate the sample gas manipulator in one direction from one line to the other to check the calibration and timing for experiments. In case of a clogged line, the second line also can be used for sample gas, without having the whole jet removed to fix the clogged line. However, the removal of the jet source is necessary to separate the first skimmer section from the rest of the gas delivery line and the main chamber with one gate valve (see 6 in Figure 4). To avoid pumping the volume between the first skimmer and the gate valve, a bypass with a second gate valve is in place. Another major feature of the sample line is the capability to tip and tilt the whole gas jet assembly (see 4 in Figure 2). This can be used to align the jet to the X-ray beam if necessary. The catcher is a two-stage setup with a residual gas analyzer (RGA) in the second section for analysis and alignment of the sample jet. The first section is equipped with a leak valve in case the main chamber needs to be backfilled. For this reason, all pumps in the catcher section can be slowed down. The supersonic gas jet with two nozzles is only one possibility of a sample source. Future capabilities will include but are not limited to sample sources for aerosols [21], pyrolysis [22], and effusive oven sources [23,24]. Different sample delivery schemes are feasible upon reasonable request.

Table 1. Main gas jet parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operation Temperature [K]</td>
<td>5 to 700</td>
</tr>
<tr>
<td>Gas Nozzles</td>
<td>2</td>
</tr>
<tr>
<td>Gas Nozzle Aperture [µm]</td>
<td>10 to 500</td>
</tr>
<tr>
<td>Operation Mode</td>
<td>CW</td>
</tr>
<tr>
<td>Operating Pressure [hPa]</td>
<td>100–6 × 10^4</td>
</tr>
<tr>
<td>Distance to First Skimmer [mm]</td>
<td>2 to 100</td>
</tr>
</tbody>
</table>
2.5. The Optical Laser Setup

At TMO, the optical excitation of gas-phase samples in the target chamber is established by a synchronized, femtosecond pulsed laser source situated in the NEH laser hall. These pulses are generated from an Optical Parametric Chirped Pulse Amplification (OPCPA) system, which is developed in-house and is capable of providing 800 nm, 100 kHz, 35 W, <20 fs pulses [25]. Ultimately, the goal is to reach a laser pulse repetition rate of 1MHz for complete synchronization with the 1MHz repetition rate of LCLS II. The optical laser hall is upstairs of TMO and the 800 nm fundamental is transported via vacuum tubes towards the hutch. To achieve a sub-10 µm optical laser spot size at 800 nm, the in-coupling of the laser is located as close to the interaction point as possible and finally focused with an in-vacuum off-axis parabolic (OAP) mirror mounted on a piezo motor driven by a hexapod—a motorized stage with 6 degrees of freedom. The designed focal length of the OAP at 800 nm is 110 mm. For beam diagnostic reasons, the optical laser beam is collimated directly behind the interaction point and out-coupled directly behind the spectrometer, as shown in Figure 5, and sent to the laser diagnostic mounted on the granite stand.
Figure 5. The DREAM optical laser setup; indicated are the sample and focus diagnostic paddle (1), the long-range microscope (2), the main chamber body (3), the off-axis focus mirror for the optical laser (4), the X-ray out towards the diagnostic (5), the X-ray scatter slits (6), the optical laser out towards the optical laser diagnostic (7), the in-vacuum hexapod for the alignment of the optical laser focus (8), the optical laser in towards the focus spot (9), the X-ray incoming from the FEL (10), the vacuum extension of the main chamber for the laser posts, which are decoupled from the vacuum chamber (11), the optical laser infrastructure post (12), the support granite (13), and the out-of-vacuum laser support and alignment structure (14).

The TIXEL Detector

To meet the detection challenge posed by high-power, high-repetition-rate FEL, SLAC is actively developing a novel front-end ASIC detector [26] based on silicon technology, known as TIXEL. Unlike conventional TOF detectors, this detector is designed to have high spatial and time resolution with a low damage threshold, without needing a charge amplifier MCP. The ability to detect the Time over Threshold (TOT) and Time of Arrival (TOA) allows this detector to detect charged particles with spatial, energy, and arrival time information. When fully commissioned, this pixelated charged particle detector can accommodate >300 hits at a repetition rate of up to 1 MHz. Thus, it is possible to perform, for the first time, massive multi-particle coincidence experiments. Since the number of combinations grows exponentially with the number of hits, detecting and analyzing large densities of multi-particle hits at a high repetition rate will soon be possible.

The readout rate of this detector is variable and scalable since only an active or triggered pixel is read out with the maximum readout rate of 5 kHz for the full array. This capability is ideal for an equally spaced bunch pattern at a high repetition rate. All the described parameters make the TIXEL detector superior to state-of-the-art detection schemes and the ideal detector for COLTRIMS types of experiments.

Table 2 shows the main parameters for the TIXEL detector, and Table 3 shows the performance parameters of the new DREAM endstation.
Table 2. TIXEL key performance parameters.

<table>
<thead>
<tr>
<th>Mode of operation</th>
<th>TOT and TOA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time resolution</td>
<td>&lt;100 ps</td>
</tr>
<tr>
<td>Pixel size</td>
<td>100 × 100 µm</td>
</tr>
<tr>
<td>Energy range</td>
<td>1–10 keV</td>
</tr>
<tr>
<td>Array</td>
<td>176 × 192</td>
</tr>
<tr>
<td>Si technology</td>
<td>0.13 µm</td>
</tr>
<tr>
<td>Full matrix readout</td>
<td>5 kfps</td>
</tr>
</tbody>
</table>

Table 3. Parameters and capabilities of the DREAM endstation.

<table>
<thead>
<tr>
<th>DREAM Parameter</th>
<th>IP 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy range (eV)</td>
<td>250–1400</td>
</tr>
<tr>
<td>Mirror incidence angle (mrad)</td>
<td>21</td>
</tr>
<tr>
<td>Mirror coating</td>
<td>Si or B₄C</td>
</tr>
<tr>
<td>Smallest focus (µm)</td>
<td>&lt;0.3</td>
</tr>
<tr>
<td>Flux, up to (photons/cm²)</td>
<td>10²²</td>
</tr>
<tr>
<td>Mirror transmission (%)</td>
<td>60</td>
</tr>
<tr>
<td>Pulse duration (fs)</td>
<td>0.3–100</td>
</tr>
<tr>
<td>Repetition rate X-rays (kHz)</td>
<td>up to 929</td>
</tr>
<tr>
<td>Polarization control</td>
<td>H, V, and elliptical/circular R, L</td>
</tr>
<tr>
<td>Target sample</td>
<td>gas jet, aerosol, nanoparticle</td>
</tr>
<tr>
<td>Gas sample delivery</td>
<td>CW and pulsed valves</td>
</tr>
<tr>
<td>Gas sample delivery temperature (K)</td>
<td>10–650</td>
</tr>
<tr>
<td>Optical laser source</td>
<td>OPCPA</td>
</tr>
<tr>
<td>Laser peak intensity on sample (W/cm²)</td>
<td>10¹⁴</td>
</tr>
<tr>
<td>Repetition rate OPCPA (kHz)</td>
<td>33</td>
</tr>
<tr>
<td>Optical laser wavelength (µm)</td>
<td>0.2–5</td>
</tr>
<tr>
<td>Optical laser focus spot size (µm)</td>
<td>&lt;10</td>
</tr>
</tbody>
</table>

¹ at 100 kHz and 800 nm. ² later upgrade to MHz. ³ at 800 nm.

3. Performance and Simulations

The great flexibility of DREAM, which makes it such a versatile tool for AMO physics and gas-phase chemistry at high-repetition-rate FEL, also makes it increasingly difficult to explain all the possible features and geometries in limited space. Therefore, we have limited the following section to one fixed spectrometer geometry and are only showcasing the possibilities with the current configuration by varying only the E and B field. Different spectrometer geometries are feasible, which include different detector to interaction length dimensions and/or inner spectrometer diameters. Given the tremendous workload and timescales to change a spectrometer setup, this can only be done in close cooperation with the LCLS TMO instrument team, and 3D models and simulations of different spectrometer geometries are available upon reasonable request.

It is anticipated that the majority of the experiments to be performed with the DREAM endstation can be divided into two categories. One is the Coulomb explosion imaging experiments, where the measured coincident ion momenta can be used to retrieve the molecular structural dynamics. The other is the electron–ion coincidence experiments, where the molecular-frame momenta of fragment electrons and ions can be reconstructed in order to reveal the ultrafast electronic and nuclear processes in molecules. In this section, we present the simulation results for these two types of experiments. Exemplary spectrometer field and charged particle trajectories are simulated with the SIMION software [27], by using the spectrometer model shown in Figure 6, which was converted by the SIMION SL tools from the 3D CAD drawing discussed in Section 2.3.
3.1. Simulation for Coulomb Explosion Imaging Experiments

Coulomb explosion imaging experiments performed at X-ray free-electron (XFEL) laser facilities demand special spectrometer considerations compared with those with table-top lasers or synchrotron light sources. With the number of photons typically on the order of $10^{12}$, a femtosecond XFEL pulse with ~1µm focal spot size can drive a molecule into extremely high charge states and induces the molecule into an almost instantaneous explosion. Therefore, the kinetic energy of the resulting ion fragments is typically much larger than in the experiments with conventional light sources. The spectrometer is required to collect such highly energetic particles over a $4\pi$ solid angle, while simultaneously preserving the mass and momentum resolution adequate for the imaging purposes.

Carbon and oxygen ions $C^i$ and $O^i$ with $i = 1, 2, 3$ are chosen for simulating the spectrometer performance in CEI experiment as shown in Figure 7. The kinetic energies of $C^i$/$O^i$ ions are set to be $i \times 100$ eV, based on data from a previous CEI experiment performed at the European X-ray Free-Electron Laser Facility. In the simulation under each setting, the spectrometer voltage is set such that it is sufficient to detect the charged particles over a $4\pi$ solid angle.

A typical charged particle “time of flight (TOF) versus detector hit position (R)” plot from the simulation is shown in Figure 8a. A “TOF vs. R” plot is often used for identifying and isolating different ion species. Thus, for a cleaner separation, decreasing the overlap between different species in such plots, which is equivalent to increasing the mass resolving power, is needed. As shown in Figure 8a, we define $\sum_{i=1}^{3} \frac{1}{T_i}$ as the metric for the mass resolving power, with the vector $\frac{1}{T_i}$ connecting the point of the largest $C^i$ TOF to the point of the smallest $O^i$ TOF. Figure 8b displays $\sum_{i=1}^{3} \frac{1}{T_i}$ as a function of the acceleration length $L_a$ and acceleration-plus-drift length $L_{ad}$. At fixed $L_a$, the mass resolving power increases with increasing $L_{ad}$, and at fixed $L_{ad}$, the mass resolving power increases with decreasing $L_a$. This dependence is better visualized in Figure 9. With $L_{ad}$ fixed at 270 mm and as $L_a$ is decreased from 270 mm to 10 mm, the distribution of each ion species is more compressed in the TOF direction, making adjacent distributions better separated from each other. Figure 8c displays the required electric field strength as a function of the acceleration length $L_a$ and acceleration-plus-drift length $L_{ad}$. At fixed $L_a$, the field strength increases with increasing $L_{ad}$, and at fixed $L_{ad}$, the field strength increases with decreasing $L_a$. As
L_a is decreased from 270 mm to 10 mm, the electric field is required to be increased from 35 V/mm to 256 V/mm.

Figure 7. Ion trajectory simulation for CEI-type experiments. The electric filed strength is 460 V/cm. The trajectories of C\textsuperscript{2+} with kinetic energy of 150 eV and C\textsuperscript{3+} with kinetic energy of 250 eV are shown in orange and blue, respectively. The ejection angles relative to the spectrometer axis range from 0 to 360 degrees. (a) Side display of spectrometer equipotential lines (red) and particle trajectories. (b) Spectrometer equipotential lines (red) and particle trajectories along the potential energy surface (green).

Figure 8. Mass resolving power and electric field dependence on the acceleration length L_a and acceleration-plus-drift length L_{ad}. (a) Simulated “TOF vs. R” plot of C\textsuperscript{q+} and O\textsuperscript{q+} ions detected with a certain spectrometer configuration, with q = 1, 2 and 3. The vector $\mathbf{T}_i$ connects the point of the largest C\textsuperscript{i+} TOF to the point of the smallest O\textsuperscript{i+} TOF. $\sum_{i=1}^{3} T_i$ is used as the metric for the mass resolving power. (b) The dependence of the mass resolving power $\sum_{i=1}^{3} T_i$ (ns) on the acceleration length L_a and acceleration-plus-drift length L_{ad}. (c) The dependence of the required electric field strength (V/mm) on the acceleration length L_a and acceleration-plus-drift length L_{ad}. 
A similar dependence trend emerges in resolving the ionic mass for different electric fields, as shown in Figure 8b,c, as is expected. When the kinetic energy acquired by the charged particle from the spectrometer field is much larger than that gained from the interaction, which is often the case in CEI experiments, the TOF spread of each ion species with a fixed momentum distribution can be approximated to be inversely proportional to the electric field strength. Since the larger the TOF spread is, the smaller is $\sum_{i=1}^{3} \tilde{T}_i$, the mass resolving power should follow the same dependence trend as the electric field strength. Thus, increasing the electric field strength can be an effective way to increase the separations between ion species in the “TOF vs. R” plot. However, it can also degrade the momentum resolution, as we will illustrate with the simulation results using the dimensions of the DREAM spectrometer.

Figure 10 shows the simulated “TOF vs. R” ((a) and (c)) and ion momentum/energy resolution ((b) and (d)) plots for the DREAM spectrometer, with $L_{ad} = 275$ mm and $L_a = 99$ mm. A position resolution of $250 \mu$m and time resolution of 0.25 ns were assumed in the simulation. Figure 10a,b are obtained with the spectrometer voltage only high enough to detect the charged particles over $4\pi$ solid angle. There are overlaps between different ion distributions in the “TOF vs. R” plot, which typically would require the additional step of applying coincidence or momentum-conservation conditions to fully disentangle distinct ion species. The momentum resolution for $C_{aq}^+$ and $O_{aq}^+$ ions ranges from sub-one to a few atomic units, and the kinetic energy resolution ranges from sub-one to a few electron Volts. The simulation results after scaling the voltage up by a factor of 1.3 are shown in Figure 10c,d. Due to the electric field dependence discussed before, the separation between ion distributions grows larger with the increased voltage setting. However, the momentum and energy resolution is slightly degraded. The increased electric field compresses the spread of the charged particles on the detector in the directions along and perpendicular to the spectrometer axis, which would in turn make the smallest resolvable momenta and kinetic energy values larger. Therefore, the trade-off between mass resolving power and momentum resolution should be taken into account when choosing the electric field for the DREAM CEI experiments.

3.2. Simulation for Electron–Ion Coincidence Experiments

In the simulations under each setting as shown in Figures 11–14, the spectrometer voltage is set such that it is only sufficient to detect the ions over $4\pi$ solid angle and retard the electrons away from the ion detector.
Figure 10. Simulated “TOF vs. R” (a,c) and ion momentum/energy resolution (b,d) plots for the DREAM spectrometer, with $L_{ad} = 275$ mm and $L_a = 99$ mm. Figures (a,b) are obtained with the spectrometer voltage setting only high enough to detect all particles over $4\pi$ solid angle. Figures (c,d) are obtained by scaling up this voltage setting with a factor of 1.3. $\delta P_\parallel$ and $\delta P_\perp$ represent the momentum resolution in the directions parallel and perpendicular to the spectrometer axis. $\delta KE$ represents the ion kinetic energy resolution.

Figure 11. Ion and electron momentum and energy resolution with the spectrometer setting as defined in Figure 12.
Figure 12. Trajectory simulation for electron–ion coincidence experiments, with electrons detected by the detector equipped to the long arm, and ions to the short arm. The electric field strength is 22 V/cm, and the magnetic flux density is 15.5 Gauss. The trajectories of electrons with kinetic energies of 50 eV and 110 eV are shown in green and brown, respectively. The trajectories of N$^2^+$ with kinetic energy of 45 eV and N$^3^+$ with kinetic energy of 60 eV are shown in orange and blue, respectively. The ejection angles relative to the spectrometer axis range from 0 to 360 degrees. (a) Side display of spectrometer equipotential lines (red) and particle trajectories. (b) Spectrometer equipotential lines (red) and particle trajectories along the potential energy surface (green).

Figure 13. Ion and electron momentum and energy resolution with the spectrometer setting as defined in Figure 14.
4. Scientific Application

With the primary scientific driver of TMO being ultrafast atomic and molecular physics in the X-ray regime, the DREAM endstation provides a powerful tool that fully utilizes the soft X-ray pulses from LCLS-II to explore challenging scientific questions. These challenges include the investigation of ultrafast charge migration, energy and charge redistribution, electron localization, as well as symmetry breaking at the molecular level and chirality, which are not well understood at the quantum level, even for simple molecular systems. These topics are central to complex molecular dynamics processes such as photosynthesis, catalysis, and bond formation and bond breaking, which play a central role in all chemical and biological reactions. The following section provides a snapshot of the scientific cases that can be probed by the DREAM endstation at TMO.

4.1. COLTRIMS

This technique developed in the late 1980s and early 1990s gives access to kinematically complete experiments on photoionization and photofragmentation processes, by enabling the coincident and momentum-resolved detection of recoiling target ions and emitted electrons [28,29]. Momentum spectroscopy is a technique extensively used in molecular (atomic) spectroscopy to detect the total multidimensional momentum of constituent particles (i.e., cations, anions, electrons, and photons) undergoing photo-reaction. The latter arises as a consequence of ionization processes such as electron–ion collision or Coulomb explosion (CE). The tools used for particle detection to resolve and reconstruct the full momentum are often called reaction microscopes (REMs) because of their source–target interaction. The DREAM endstations with the delay line anode, combined with a cold atomic and molecular target, forms an ideal workbench to study fundamental atomic physics with this technique. Previous measurements demonstrated ultrafast manipulation of the weakly bound He₂ dimer potential through an external laser field [30], with the potential to investigate the superposition of the ground state and excited Efimov state of He₃ using 4-body momentum imaging, only possible through COLTRIMS technique [30,31]. Furthermore, COLTRIMS allows the study of the multi-particle fragmentation dynamics of molecules through simple yet powerful momentum gating methods. Through measurements with high position and temporal resolution, the momentum vectors of all emitted...
electrons and ionic fragments in the same event can be measured, revealing the entangled many-particle dynamics in the molecular system. In the multistep fragmentation process, experiments [32,33] involving left and right circular polarized photons in molecules showed evidence of possible dynamically induced symmetry breaking between core-hole and Auger electrons. With LCLS-II and advanced modes of operation, such as multi-color, multi-pulse, polarization, and ultra-short sub-femtosecond duration [34–36], these symmetry breaking measurements with variable X-ray polarization would be possible with DREAM endstation.

4.2. Coulomb Explosion Imaging

To acquire sufficient statistics, coincidence methods require the detection of a large number of events. This requirement is met by LCLS III and by high-repetition-rate optical laser systems supporting DREAM. The large data sets created by coincidence experiments at repetition rates of <100 kHz will require novel data science approaches to extract the desired structural information, which is also given at TMO.

Several recent experiments have demonstrated that CEI with XFELs can image the position of each atom in a polyatomic molecule [11,13]. If an ultra short X-ray pulse is used to efficiently ionize and fragment the polyatomic molecule within a timescale in the range from femto- to atto-seconds (faster than photo-chemical reactions), the CEI method may be an ideal probe for transient molecular structures. Based on the inverse relation between the internuclear distance and the Coulomb potential, the molecular structure before the explosion is encoded in the momentum distribution of the ion fragments, which directly reflects the equilibrium geometry of the molecule. Thus, CEI is a promising imaging technique and gives the extensively used expression “molecular movie” a new and extended meaning in terms of accessible particle correlations.

4.3. Ultrafast Molecular Dynamics in Chiral Systems

With the advent of LCLS II along with novel operation modalities such as tunable polarization, attosecond X-ray pulses, multibunch modes, MHz bunch repetition rate, and the new DREAM endstation, SLAC has enabled the opportunity to gain unprecedentedly detailed insights into chirality. Finally, through the innovative FEL capabilities, it will be possible to observe and understand how chiral systems form, restructure, functionalize, and, ultimately, can be controlled. Investigating the origins of chirality with TMO instruments, the knowledge bridge between the macro-biological level, chemical dynamics, and the fundamental spin properties of matter can be made possible [8,37]. Previously, investigation of the chiral ethane derivative (CHClBrCF$_3$) demonstrated that the handedness could also be determined for a more complex species [37]. The fragmentation into constituent fragments of CH+, Cl+, Br+, and CF+, followed by momentum reconstruction, showed a clear signal for the two enantiomers from a racemic mixture of CHClBrCF$_3$. This facilitates the direct comparison of experimental measurements with high-level molecular dynamic simulations of ultrafast photochemical processes. In addition to transient diffractive imaging techniques, time-resolved momentum imaging of ionic fragments that result from molecular breakup following rapid multiple ionization provides a powerful experimental tool for mapping transient molecular configurations and reaction intermediates in light-driven processes.

4.4. Advanced Reaction Microscopy for Studying Extended Nanosystems

As the study of extended quantum systems and nanoparticles becomes more and more feasible due to the rapid development of synthesis and sample delivery systems, an instrument such as DREAM will play a crucial role in studying light–nanoparticle interactions. As such, research into the properties of various nanoscopic systems has the potential to lead to breakthroughs in both fundamental science as well as in novel applications and technologies. On the fundamental level, research into nanoparticles, clusters, and other such complex assemblies allows for the investigation of scaling laws to
understand how the physical response of a system evolves as the characteristic spatial scale changes from atomic/molecular to a macroscopic nature. Recent experiments involving the discovery of intermolecular Coulombic decay in endohedral fullerene [38], demonstrating the enhanced formation of $\text{H}_3^+$ on nanoparticles [39], multi- and single-photon processes in fullerene-like systems using hard and soft X-ray photons [24,40] were only possible through coincidence measurements of ions on a shot-by-shot basis. Such measurements will form the basis of studying gas-phase nanocatalysis and photon-induced enhancements of surface chemistry at LCLS-II using the DREAM endstation.

5. Conclusions

LCLS and LCLS II, producing high-flux femtosecond to attosecond X-ray pulses, will yield unprecedented intensities. The TMO instrument with the DREAM endstation will take advantage of the pulse properties to perform high-power soft X-ray experiments in a wide spectrum of scientific domains. The DREAM endstation provides a new COLTRIMS setup at the end of a superconducting 1 MHz FEL. With the added flexibility in terms of spectrometer setup, it allows sample delivery but also the possibility for future updates. The first user-based experiments for DREAM are planned for Fall 2023. Further updates and more details about the TMO instrument can be found at the following website: https://lcls.slac.stanford.edu/instruments/neh-1-1.

6. Facility Access

LCLS instruments are open to academia, industry, government agencies, and research institutes worldwide for scientific investigations. There are two calls for proposals per year and an external peer review committee evaluates proposals based on scientific merit and instrument suitability. Access is without charge for users who intend to publish their results. Prospective users are encouraged to contact instrument staff members to learn more about the science and capabilities of the facility, and opportunities for collaboration.

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