Project Summary

Continued and significant progress in the generation and use of novel ultrashort X-ray pulses at the LCLS X-ray Free Electron Laser (XFEL) ensures that LCLS-II will maintain its international leadership. We are sure to continually develop even more novel modes based on the new variable gap undulators, split and Delta undulator configurations that allow multiple polarizations, "fresh slice" lasing for dual pulses, and even attosecond pulse generation. Multi-color, multi-polarization and multiple x-ray pulses with exquisite temporal control are all featured priority areas for LCLS-II which cites as a principle need being attosecond time resolved photo and Auger electron emission in order to directly capture detailed dynamics of correlated electron motion. We will no longer be making molecular movies, but rather movies of the electrons moving around and through those molecules. To do this, however, one requires a diagnostic that can recover the exact temporal profile for each Self-Amplification of Spontaneous Emission (SASE) pulse. Such a single shot diagnostic must recover potentially complicated pulse shapes for on-the-fly data sorting and veto.

So called "angular streaking" was recently identified as the likely method to deliver the needed pulse characterization. In actuality, it provides the foundation for a comprehensive attosecond experimental paradigm. Familiar in the high-harmonic generation laser community, angular streaking uses a long-wavelength streaking laser to provide a "clock" against which attosecond electron dynamics can be measured. Given the similar requirements, we propose to address the pulse diagnostic needs as well as provide this basis for attosecond resolved electron spectroscopy at LCLS-II.

We propose a new generation of attosecond diagnostic capability, one that is tailored to the unique XFEL pulses existing and expected, having learned from our recently demonstrated reconstruction of attosecond scale pulse structures of LCLS. We found that the synchrotron-optimized detector array that was used for that initial demonstration suffers multiple shortcomings for FEL use; limitations that have proven the most challenging impediments to accurate pulse reconstruction.

The scope of this proposed project is therefore the Research and Development required to develop an XFEL optimized angular array of electron Time-of-Flight (eTOF) spectrometers that will meet the stringent needs of LCLS-II. The new detector array will be optimized specifically for single-shot angular streaking measurements at the FEL while minimizing the inter-detector cross-talk experienced in the previous design. We target a spectral resolution of 0.25 eV by improving the sensor electronics and by integrating on-board signal processing that is specifically matched to the LCLS-II data reduction pipeline. Furthermore, we will design for a new feature whereby the eTOFs are capable of analyzing multiple spectral windows, each of high energy resolution, to accommodate two-color double pulses from widely detuned variable gap undulators. This multiple window feature is a key development for the sake of element-specific tracking of electron transfer and charge migration, unlocking the core element of the LCLS-II attosecond science program. Furthermore, we will design to accommodate also the split undulator method in combination with Delta undulator production of variably polarized pulses, ensuring sub-spike polarization analysis.

The output from this project will not only provide the design basis for attosecond resolving single-shot x-ray pulse reconstruction but also an advanced instrumentation concept as a platform for core and future LCLS-II science.

Enabling long wavelength streaking for attosecond x-ray science.

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Project narrative

Introduction

The advent of x-ray free electron lasers (xFELs) brought the ability to resolve ultrafast processes in molecular and material systems on their natural vibrational time and length scales [2–5]. With the coming of the next genreation xFEL, the Linac Coherent Light Source II (LCLS-II) the electronic horizon is upon us: we will use attosecond x-ray pulses to control and interrogate the correlated electronic motion as we delve into the age of molecular electronic movies. Given the importance of understanding electronic flow in photo-excited systems (Fig. 1), there is a strong desire to drive electrons into concerted coherent motion [6–8] and then probe the molecular environment with time-resolved atomic site-specific x-ray sepctroscopies. Opening the field of attoscience to the xFEL machine, inspires one to imagine attosecond resolved extension to two-dimensional resonant Auger electron spectrosopies of Ref. [9]. This inspiration has led to direct funding of a major research effort focused specifically on attacking this regime with FEL sources [10].

Table 1: Soft x-ray conditions for LCLS-I and the high-repetition rate LCLS-II. [1]

Parameter	LCLS-I	LCLS-II
Max rep. rate	120 Hz	930 kHz
Average power	0.5 W	200–900 W
Pulse energy	4 mJ	0.1–5* mJ
Photon energy	0.25–2 keV	0.2–5 keV
Arrival stability	100 fs rms	20 fs rms

* $\geq$ 200 μJ typically at reduced repetition rates. Lower charge modes for short pulse operation can conserve peak power while allowing full repetition rate.

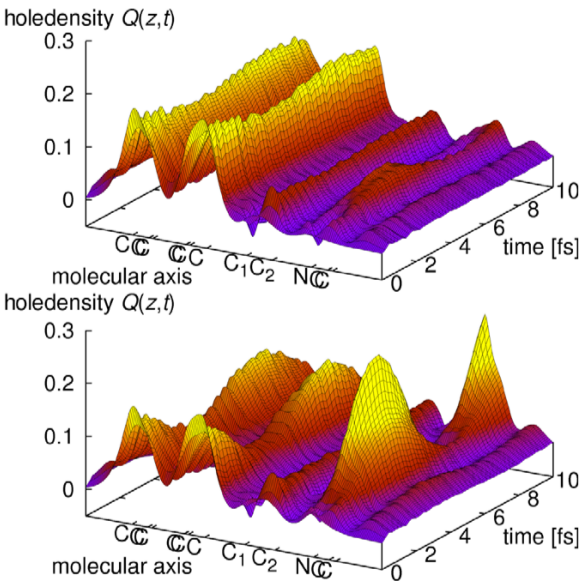

Figure 1: Hole migration in PENNA molecule following photoionization in the ground neutral molecular configuration (top) versus the C_2-C_2 20pm stretched configuration reproduced from Ref. [6].

One of the biggest challenges for the traditionally laser-based attosecond science community is the difficulty in producing significant pulse energy in the 200 eV – 2 keV regime using high harmonic generation (HHG) [11, 12]. These sources may be encroaching on this x-ray photon energy range [13, 14] but the intensities and repetition rates available at FEL sources like the LCLS-II (Table 1) are driving the community to look toward all x-ray pump-probe. There have been numerous schemes proposed for developing the attosecond capability of x-ray FEL facilities [15, 16] with a particular push funded by the Office of Basic Energy Science aimed at LCLS-II implementation [17, 18].

The development of temporally shaped x-ray FEL pulses would not only facilitate attosecond pulse generation but also a number of multi-pulse non-linear techniques [19]. A continued progress on this front [17, 20–27] will require full spectral phase, amplitude and polarization characterization as recently demonstrated in Ref. [28]. We therefore propose a single-shot diagnostic that reports the full temporal intensity, wavelength, and polarization distributions with ∼150 attoseconds resolution at the highest repetition rates,

limited only by the optical laser repetition rate that is used for the streaking drive laser.

We identify two fundamental but parallel objectives:

1. *Detector:* We will perform the necessary reasearch and development required to design an optimized angular array of electron Time-of-Flight spectrometers.

2. *Analysis:* We will integratively develop the requisite algorithms and machine learning compliments together to optimally match the real-time analysis routines with detector electronics and computing hardware that pushes the tenchnology envelope.

Angular streaking

There is increasing momentum for the development of spectro-temporally shaped x-ray FEL pulses [17, 20–22, 24, 26, 27, 29–31] in response to the rising tide of demand [6–8, 10, 32–34]. So far, the predominant method to characterize such novel temporal profiles is based on an x-band transverse accelerating cavity (XTCAV) [35] whereby the spent electron bunch is deflected horizontally, streaked in time by the phase of the transverse accelerating field. This technique has been a critical tool for developing the recent x-ray FEL pulse shaping methods [17, 23, 24, 27, 31]. A bending magnet then deflects this time-streaked beam vertically proportional to the energy. Imaging the result, one records the time-energy distribution of the spent bunch, indirectly identifying the imprint of lasing on the electron bunch. Furthermore, barring a superconducting upgrade to the x-band cavity, the XTCAV can only run at 120 Hz, providing only intermittent information at best.

We seek a direct method based on the concept of photoelectron streaking, a direct interaction and one that has the capability to measure the temporal structure of x-ray pulses [36]. Since its inception as a pulse reconstruction method [37, 38] there have been great recent gains in diagnosing laser-based pulses that encroach on the soft x-ray regime [39–41]. Although some exquisite phase retrieval analyses require interference with reference oscillators [42, 43], we favor a simpler and more flexible scheme that can also accommodate *in situ* x-ray experiments as well as the pulse retrieval diagnostic. We focus on the angular array of electron spectrometers that are more compatible with the x-ray regime 0.2keV and up, as used for our recend proof of concept in Ref. [28].

In x-ray photo-electron streaking, a noble gas like neon is dressed by a strong long wavelegnth infrared or THz field [19]. The streaking field shifts the outgoing photo-electron energy depending on the phase of the field at the time of photoionization. In the more common linear polarized streaking [44–48], the intensity profile of the photo-electrons versus their shifted energies can be mapped to

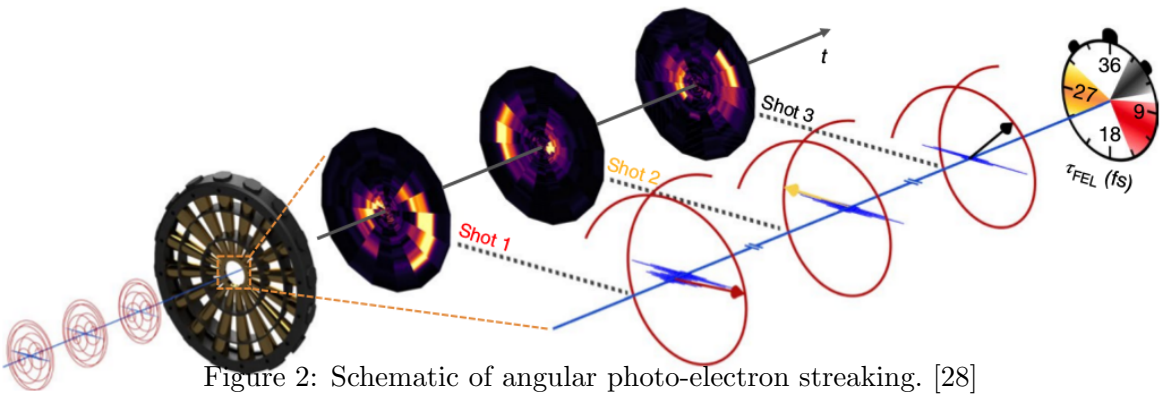

Figure 2: Schematic of angular photo-electron streaking. [28]

the time-variation of the vector potential in a familiar "streak camera" interpretation. This requires, of course, that the x-ray pulse arrives near the zero-crossing of streaking vector potential. There have been recent developments by the Cavalieri group to arrange two photo-electron spectrometers to sample the focal volume of the streaking field at two different places across the Gouy phase of the focus. This in principle relieves the need for a zero-crossing carrier field, it is particularly sensitive to the exact focussing conditions of the streaking laser. However, in angular streaking as depicted in Fig. 2, the vector potential is made to rotate as a circular polarized long-wavelength field. We thus intend to use the angular streaking laser field as a "clock" that imprints time into the electron spectra and thus allows for a time-resolved interpretation of electron emission [49], leveraging our long history with photo-electron streaking at the LCLS [44,45,48,50,51] to extend the attosecond angular streaking method of Refs. [38, 52] to the x-ray regime.

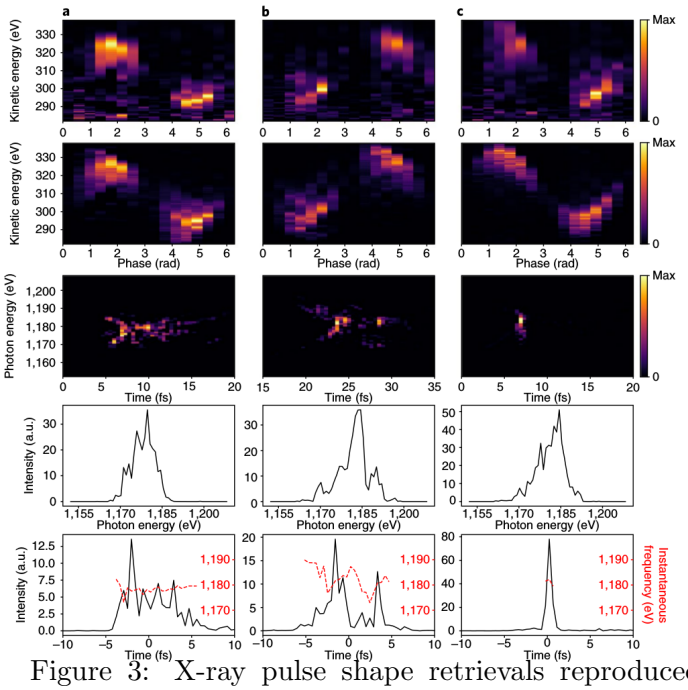

Figure 3: X-ray pulse shape retrievals reproduced from Ref. [28].

As represented in Fig. 2, we recently repurposed what was originally considered for x-ray FEL polarimeter [22, 27, 53, 54] to measure the angularly streaked photo-electron spectra with an angular array of 16 electron Time-of-Flight (eTOF) detectors as depicted in Fig. 2 [28]. Normally, the x-ray pulse produces undressed neon photo-electrons that distribute into a dipole probability distribution for linearly polarized x-rays, and a circular pattern for random or circular x-ray polarization with a common kinetic energy regardless of emission angle. When dressed with the circularly polarized laser field (Fig. 2) those electrons receive a momentum kick toward the instantaneous direction of the vector potential, away from center in Fig. 2, in a reference frame that spirals relative to the lab frame at the carrier cycle frequency. In this way, one detector will measure electrons with an excess of energy, the opposite detector with less energy, and the two orthogonal detectors will measure the photo-electrons as the projected vector-potential sweeps through a zero-crossing. The additional detectors further constrain the pulse shape retrieval shown in Fig. 3. Depicted in Fig. 2 and judging from the displacement of the electron emission pattern from center, x-ray shot 1 arrives when the vector potential is pointing toward 3 o'clock, shot 2 at 9 o'clock, and shot 3 at about 2 o'clock. The result was an x-ray pulse temporal reconstruction with 500 attoseconds resolution, limited primarily by the compromised single-shot energy resolution and conservatively long 10 μm wavelength for the streaking field. Given that the array of eTOFs was originally used at LCLS as a polarization diagnostic, we target here a full spectral and polarization reconstruction with 150 attosecond temporal resolution and 0.25 eV energy resolution.

Attosecond pump-probe

Many attosecond scale experiments that are currently only enabled by high-harmonic generation (HHG) [11–14, 36, 39, 40, 55, 56] could greatly benefit from the much higher brightness of an attosec-

ond xFEL beamline [15, 16, 18]. They would effectively exchange their current flux challenge for the synchronization challenge of xFEL pulses. However, the ability for x-ray pulse characterization at the attosecond level removes the synchronization challenge for x-ray pump/x-ray probe experiments. Figure 4 indeed shows that angular streaking can not only identify double pulses, but also sort such pulses into relative delay [28]. In that demonstration roughly 1% of the SASE pulses measured consisted of only two spikes when running in low charge mode with emittance shaping [23,57]. Such pulses can then be sorted, allowing a familiar "measure-and-sort" x-ray pump/x-ray probe experimental paradigm.

At the FEL, we are free to vary the x-ray photon energy of the pulse pairs independently [20, 31] and also thier polarization states [27,58]. This is a key feature for studying the kind of chirality dynamics that was listed as a major scientific thrust area for LCLS-II [1]. One could imagine using one color of x-ray pulses to pump a chiral molecule such as trifluoromethyloxirane [59] at a fluorine or cabon atom and then probe the evolution of the valence electronic response via XAFS at the oxygen edge. This requires different polarizations [27,58] where also one pulse is tuned to the oxygen edge and the other is tuned to the carbon or fluorine edge. Inspired by Fig. **??**, such a capability is enabled by the combination of Delta undulator for polarization control together with the variable gap soft x-ray undulator of LCLS-II in split mode [20,31]. Captializing on this novel x-ray shaping mode sets the requirement that our attosecond pulse diagnostic allow for also polarization determination for pulse pairs separated by well over to 200 eV.

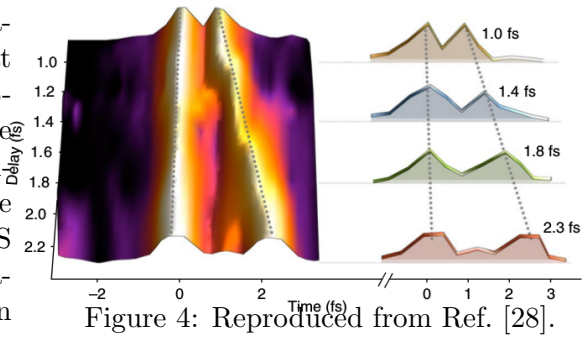

Figure 4: Reproduced from Ref. [28].

We also note that, if instead, one prefers a weaker isolated attosecond probe pulse [11,12,39,40], one could use a more traditional laser-based HHG source and still adress the HHG/xFEL synchronization challenge. Here, the euv light is sufficient to produce attosecond bursts of electrons from a helium buffer gas while the x-ray pulses would equally well produce high energy photoelectrons also from helium. In this way, angular streaking could simultaneously recover, on a single shot basis, the relative delay between x-ray/x-ray and even euv/x-ray attosecond pulse pairs. Attosocond pulses generated by the high-harmonic plateau region [56,60] could be used to induce inner valence transitions in molecular systems, setting up rather pronounced coherent electronic motions. Then the attosecond x-ray pulses from the LCLS could be used to interrogate the valence occupation via time-resolved photo and Auger electron spectroscopy. Alternatively, one could use the weaker HHG isolated attosecond pulses [39] as a supercontinuum probe up to the carbon K-edge. The LCLS-II would provide the much higher power attosecond pump pulses for $1s \rightarrow$ valence resonant transitions. This would allow for strong pumping of valence electronic correlations from a chosen atomic site in the molecule well above nitrogen and

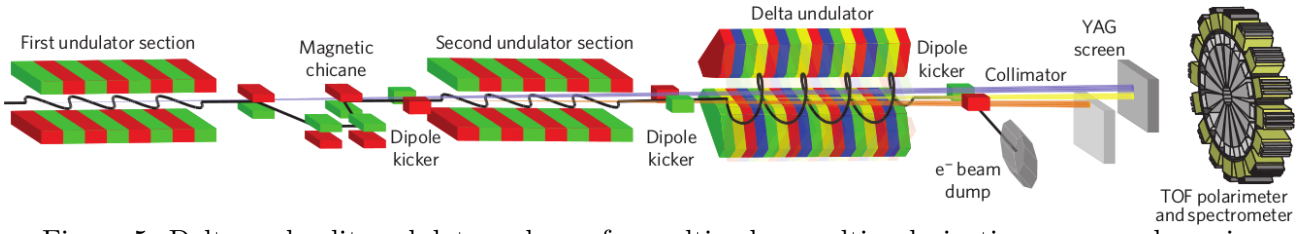

Figure 5: Delta and split undulator scheme for multi-color, multi-polarization, x-ray pulse pair generation reproduced from Ref. [27].

oxygen, even fluorine and transition metal L-edges.

Objectives

Optimized Detector Array

The technical requirements that enable pulse retrievals for accurate reconstruction are not trivial to achieve. Pulse-to-pulse variations at an FEL require single-shot measurements much like the velocity map imaging (VMI) [62] extension of attosecond angular streaking [63]. Although we have explored such a single-shot VMI solution for 120 Hz operation [64], its requirement of a two-dimensional area detector precludes its use for the high repetition rates expected. Furthermore, the single resolution window of the VMI precludes its use for the kinds of novel multi-color pulses we have come to expect from LCLS [20,21,24,27,31]. We are therefore designing toward a single-shot diagnostic that reports the full temporal intensity, wavelength, and polarization distribution also with attosecond scale resolution and at the highest repetition rates up to 1 MHz. We further require a two-fold over sampling in the angular dimension in order to fully characterize even multi-pulses (Fig. 7) [27, 31]. Modeled after the original so called "Cookie Box" design of Jens Viefhaus, we propose a main chamber that accepts micro-channel plate based electron detectors in a 16-fold symmetric angular array.

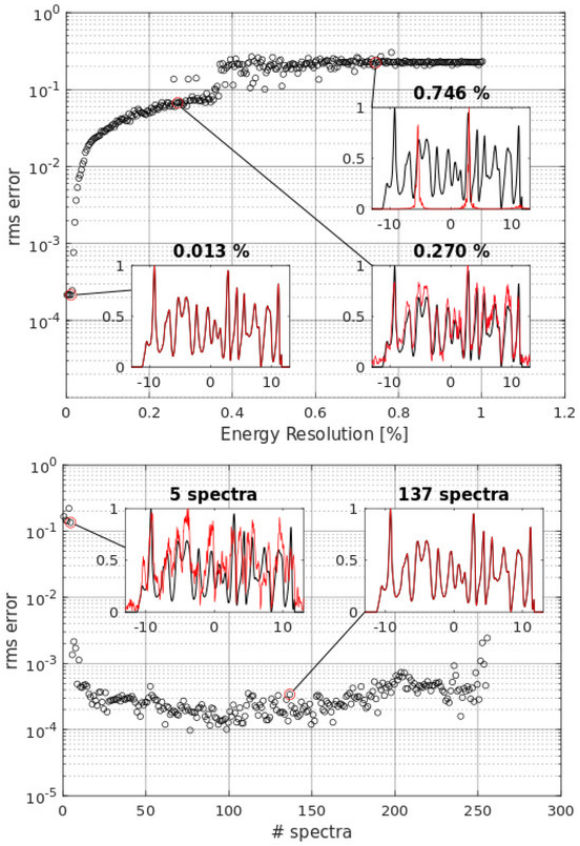

Figure 6: Reproduced from Ref. [61]. (upper) X-ray pulse retrieval error versus % energy resolution. (lower) Retrieval error versus angular sampling.

Preliminary results of Ref. [28] were based on 1 eV eTOF spectrometer resolution when working in the required single-shot current mode (Viefhaus points in Fig. 8. We note that used in counting mode as at a synchrotron, higher resolution can be achieved by electron event edge detection, but the FEL use case requires a direct spectral readout with the MCP electronics run in "current" mode with the voltage waveform being digitized as a single shot spectrum. This sets the resolution not by how well one can locate the center of the impulse response waveform, but rather the actual width of that impulse response waveform. This motivates us to design for the sharpest achievable response from the MCP assembly and that we digitize that waveform with appropriately high sampling over a sufficiently long temporal span. The current target of 400 ps FWHM for the MCP response then pushes us to design for at least 5 GSps digitizer sampling over at least a 200 ns long window.

Although the final decision on retrieval algorithm is a subject of active research, we use so called "Attoclock Ptychography" [61] to inform our specifications for spectral and angular resolution. From Fig. 6(upper) we can see that the energy resolution for the streaked photoelectrons should ideally be in the sub 0.25% range. The fine tuning of the sorts of novel FEL modes that allow for attosecond [18] and two-color [31] FEL experiments typically also require 0.25 eV resolution [65]. We are therefore targeting 0.5 m long eTOF spectrometers which we expect to give an energy resolution of 0.25 eV at

up to 100 eV electron kinetic energy above the retardation voltage, based on an estimated comparison of existing detectors (points) to the simulations (line segment) shown in Fig. 8.

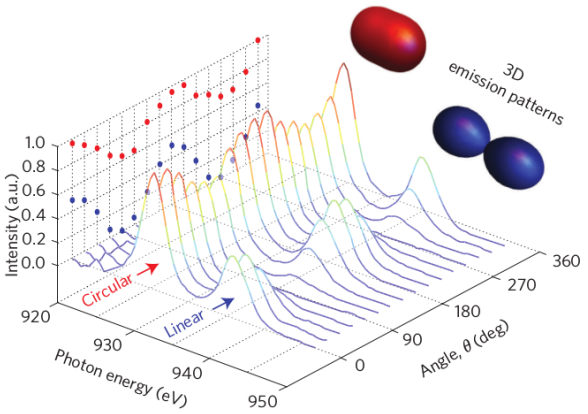

Figure 7: Two-polarization, two color, pulse pair demonstration using the original CookieBox, reproduced from Ref. [27]

The resolution in pulse reconstruction depends also on the number of angular sample points per optical cycle. In Fig. 6(lower) we see a clear prescription that the angular streaking pattern should be sampled at least along 6–8 angular sample points. The diminishing returns for adding more detector assemblies drives the design to 16 angles, providing a two-fold over-sampling of the angular dimension for the sake of our two-color aspirations. Furthermore, by shifting the dressing laser frequency toward the near-infrared we can further improve the temporal resolution. Based on Refs. [1, 6–8] we expect that much of the x-ray pulse characterization needs will lie in the sub-10 fs regime. We propose therefore to shift to a 2-3 μm wavelength for the streaking laser to improve the temporal resolution while still preserving an appropriate window for pulse shape retrieval. The shorter wavelengths also typically improve the fractional bandwidth for a more robust carrier shape, though stable and ultrashort THz [66–68] and mid infrared [69,70] are active research topics at SLAC and indeed will aid the retrieval of long duration shaped x-ray pulses. Nevertheless, a change to short wavelength should take our initial demonstration of 500 attosecond resolution for a 10 μm, 33 fs optical cycle, angular streaking field to an expected 150 attoseconds resolution for a 3 μm field — 11 fs optical cycle. Figure 9 shows a 4 μm wavelength simulation of two xFEL pulses that are separated by only 4 fs. By pushing further down to a 1 μm streaking field, we could expect to achieve temporal resolutions competitive with laser-based HHG state-of-the-art [14, 41].

When diagnosing closely separated double pulses, the angular acceptance of the electron spectrometers are a principle concern [71]. We plan to optimize this angular acceptance such that we relax slightly the collection efficiency of each detector for the sake of preserving the energy resolution. We were able to reduce the sample density by 10-fold in Ref. [28] to avoid the onset of space-charge blurring of the streaking resolution. We expect that we can sacrifice a factor of 6 in signal in order to relax the angular collection of the eTOFs also by partially compensating for this loss by using new funnel-pore microchannel plates [72].

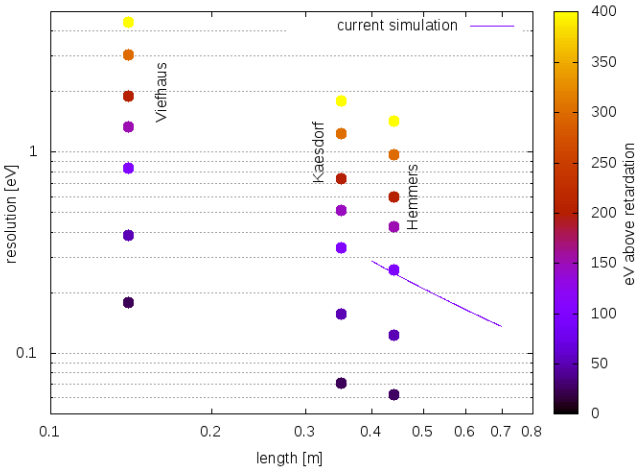

Figure 8: Detector resolution versus length comparison.

We are designing for a novel configuration whereby individual detectors in the eTOF array can have vastly different retardation voltages. Motivated by the great progress in multi-color FEL modes [17,20–22,24,26,27,31], we will capitalize on the two-fold over sampling in the angular dimension to accommodate extreme color separations between multi-pulses. Figure 10 shows that the soft x-ray undulator (SXU) for LCLS-II will be capable of K values from 2 to 5 that could provide two-color

pulses with one pulse below the carbon edge and the other above the oxygen edge. We plan for tests of such a novel mode by demonstrating interleaved retardations used to measure the carbon and the oxygen Auger electron spectra simultaneously with high resolution as early as spring 2020.

Real time analysis

Given the MHz scale repetition rate of Table 1 for LCLS-II, a measure-and-sort method that required every XFEL shot be recorded would increase the data load from the 10 TB/day of today to 100 PB/day. Such a load would require an enormous cost for developing the ultra-high duty-cycle area detectors and the corresponding network and storage infrastructure. If instead one could use real-time information about the x-ray pulse shape, then one could veto events where the pulse characteristics were not amenable to the physics being measured. Furthermore, one could allow for on-board histogram updates in a memory buffer that could collate the real-time sorted results prior to network transfer. Such a vision fuels our strong motivation to not only provide high repetition rate veto, but also event rate, low-latency sorting triggers.

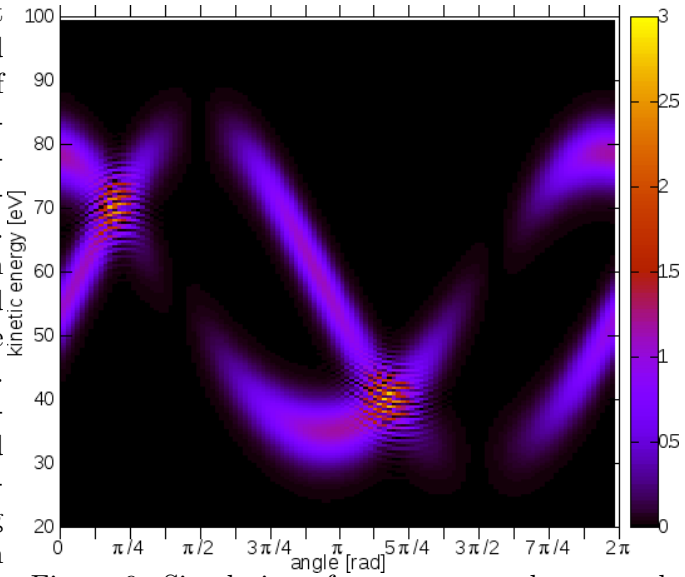

Figure 9: Simulation of two attosecond x-ray pulses separated by 4 fs dressed by a 4 μm streaking field, courtesy J. Cryan.

The on-board analysis of the data is a challenging bottleneck in the angular streaking scheme. The raw data in angular streaking is the digitized waveform spanning about 500 ns of record length with a sample frequency of ideally about 10GS/s, one waveform for each of the 16 detectors. Transferring and writing this data would require nearly 160 GBps steady-state continuous feed. We therefore require that the analysis occur ideally directly in combination with the digitization. All together though, this analysis would be comparable to analyzing one 256×256 10 bit deep image every microsecond.

Detailed in Ref. [28], we iteratively account for intensity in the polar representation of the angular photo-electron spectrum. This so called "PacMan" routine as well as a similar methods [64, 71, 73] and certainly "Attoclock Ptychography" [61] are computationally expensive and are not likely to allow MHz or even 100 kHz data throughput. They certainly will not achieve the desired microsecond scale latency for providing sort and veto triggers. We therefore plan to reserve such computationally expensive methods for the generation of so-called "ground truth" x-ray pulse shapes for a sub-set of full fidelity raw data.

We plan to use the ground truth set for training and validating a low-latency inference matrix solution that could be implemented as a series of FPGA-based on-board matrix multiplications. Only very small data of the retrieved pulse would be transferred to remote data recording nodes along with intermittent high fidelity shots for continually populating the validation and re-training sets. We will use simulations of angular streaking from expected FEL pulses, together with the detector array simulations used for the design modelling, to iterate on the detector hardware configurations and the analysis pipeline from electronics to inference output. We will use existing data to build a repository of example waveforms with realistic expectations for the eTOF point-spread functions.

The inference model will be trained based on a transfer learning paradigm whereby the central hidden layers, e.g. the simulation trained layers, will be optimized to recover x-ray pulse shapes given simulated angular streaking results. The next step of the model training then adds a thin surface set of layers at the input side that is connected to the raw data of the detector. We expect that such an architecture will preserve the physics of angular streaking as contained in the simulations, while also incorporating the raw sensor calibrations as well as adjusting to any remaining systematic errors in the retrieval interpretation. By performing different algorithms on the same data, we can benchmark the pre-analysis that will ultimately be used to enhance resolution in the retrieval while maintaining the very high throughput needed for real-time analysis.

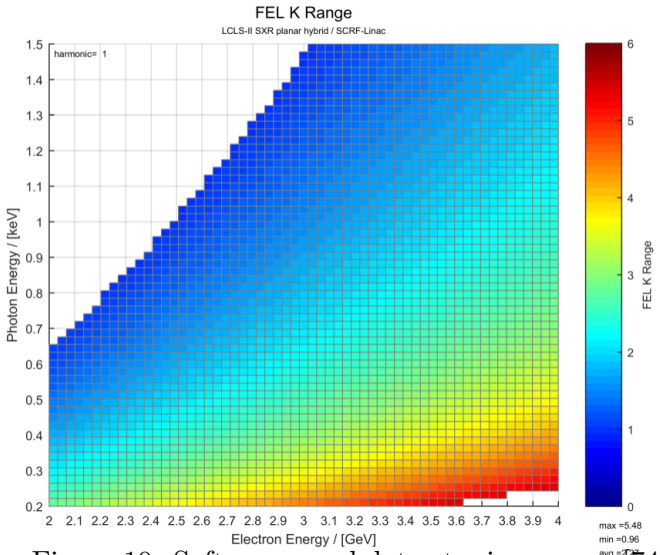

Figure 10: Soft x-ray undulator tuning range. [74]

This chain of analysis will be compiled for FPGA deployment. By deploying to FPGA, we will only need to transfer the reconstructed x-ray pulse description rather than each of the individual raw waveforms. We will leverage our close collaboration with the Stanford CS/EE group of Kunle Olukotun, in particular though a shared Graduate Student Fellow, to deploy the analysis chain to FPGA as a very high throughput (10^6 Fps) and low latency (~ 10 μs) streaming inference engine.

Organization of Major Activities

Scope

The scope of this project is the Research and Development required for the design of an electron spectrometer system that is capable of reconstructing the x-ray pulse shapes with attosecond time resolution, 0.25 eV spectral resolution, and sub-pulse polarization resolution. It should function as a single-shot diagnostic for multi-pulse xFEL configurations that span hundreds of eV photon energy difference. Motivated by the recent experimental success of Ref. [28] and further by the pulse reconstruction efforts of Refs. [61,64], we target an angular array of newly designed electron Time-of-Flight spectrometers. The project will deliver a design that meets specifications for each eTOF spectrometer, including mutual interaction considerations in the angular array. These specifications are set by the needs for reliable reconstruction of both SASE and x-ray pulse pairs of complicated polarization, multi-color spectrum, and temporal structure; more succinctly, the polarization resolving attosecond Time-Energy distribution.

The project will also deliver an integrated data pipeline plan. We will use the simulations that result from the designing the optimised detectors to also develop in parallel the optimized analysis stream. From simulations we will benchmark the retrieval algorithms of Refs. [28, 61, 64] against the new specifications of higher energy resolution over wider energy windows. We will match not only the electronics to the expected signals, but also the on-board pre-analysis in the overall data pipeline. Since we target at least 100 kfps analysis throughput, ideally with microsecond level latency, we expect the need to use high-speed machine learning inference. Therefore we will integrate the inferencing with

the early stages of the digitization and FPGA-based signal processing. The output of the inference engine is expected to inform user detectors how to route buffered data, either into sorted order for histogram updates or for intelligent veto.

Schedule

Project Year 1		Project Year 2	
eTOF spectrometer design	Simulate final design	Test inital MCPs	Test Final system
Initial MCP electronics	Install initial MCPs	Complete MCP set	Final install
Chamber Design	Purchase Chamber	Install inital spectrometers	Complete Chamber
Initial Digitizers	Simulate waveform sampling	Complete Digitizer set	Minimize cross-talk
Purchase SystemOnChip		Purchase FPGA	
Develop algorithms	Train ML on sims	Transfer Learning	Benchmark latency

Project Year 1 In the beginning of Project Year 1, two prototype detector electronics, the MCP assemblies, will be ordered. The bench testing of these two detectors will test for cross-talk interactions and ringing. We will also purchase LCLS-II compatible digitizer electronics that represent an upgrade to the current expectations. These digitizers will be chosen based on the simulated waveforms and the measured impulse response of the MCP assemblies. The signal testing will also provide further insight into the actual signal processing that will be needed in the final build out. The vacuum chamber chamber represented by the preliminary sketch shown in Fig. 11 will be optimized with a final design readied for purchasing. The detectors will be installed into the vacuum chamber. Since fringe electric fields may compromise the final electron energy resolution, along with stray magnetic fields, we must test the prototype detector pair for cross-talk and ringing in the final chamber environment.

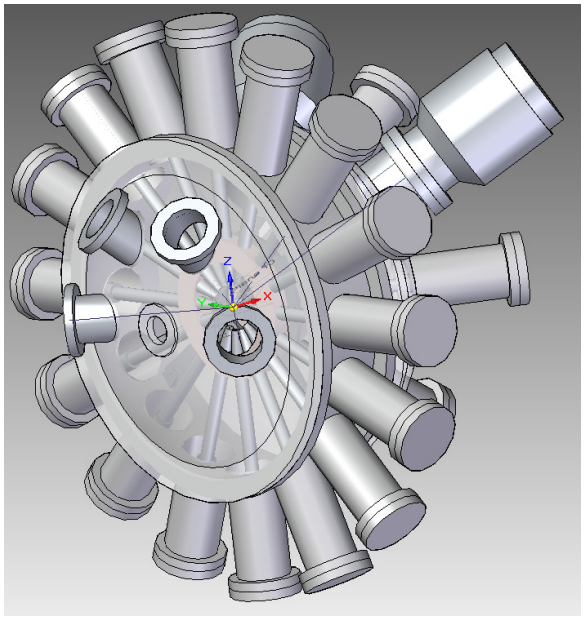

Figure 11: Initial design sketch of the proposed 16-fold symmetric vacuum chamber. Image courtesy Jean-Charles Castagna and Michael Holms.

- Test 2μm streaking with attosecond pulses from LCLS-I using Axial VMI [64].
- Design detectors for array, use existing data to develop algorithms for streaming analysis.
- Purchase all electronics supplies and chamber.
- Design transfer learning implementation plan for LCLS-II.
- Test waveform sampling versus pulse reconstruction accuracy.

Project Year 2 In the beginning of Project Year 2, we will partially complete the detector array with an additional 4 detectors assemblies with corresponding digitizers. With a total of 6 modules, we will be able to test the multi-retardation window scheme and its impact on also measuring a third, Auger channel. This will allow a demonstration of the design performance for simultaneous reconstruction of two-color x-ray pulses along with time-resolved Auger electron spectroscopy. We will

use the 120 Hz LCLS-I machine running into the soft x-ray undulators to demonstrate the spectral range of the detectors and the resolution. We will also develop the calibration protocol which will then be automated as the system becomes integrated into the LCLS-II controls system and DAQ. We note that expected configuration of an ML-enabled data reduction node is currently being designed with this detector as one of the use-cases for on-board streaming analysis. We will therefor interface directly with the group of Jana Thayer to ensure full compatibility with the LCLS-II Data Reduction Pipeline (DRP). By August of 2020, we expect the system would be ready for full deployment, just in time to receive LCLS-II, high repetition rate pulses.

- Implement on-board FPGA processing and demonstrate calibration learning.
- Optimize FPGA usage and benchmark SoC solution compared to larger ML-server solution.
- Construct detector array prototype for Building 40 benchmarks and LCLS-II beneficial occupancy.
- Install array on LCLS-II and benchmark resolution with XAFS using 3 μm laser dressing of N_2O.

Responsibilities of key project personnel The PI will be responsible for leading the design effort to ensure a fundamentally integrated system, from chamber design and detector hardware to signal readout and waveform analysis. He will also be principally concerned with the pulse reconstruction algorithm, inference engine, and data analysis pipeline.

Co-investigator Peter Walter is a Staff Scientist in LCLS Science Research and Development and will principally lead the detector simulations and construction. Peter will also primarily lead the installation for early testing of the final concepts. Co-investigator James Cryan is a Staff Scientist in LCLS Science Research and Development and PULSE and he will host much of the detector euv-based commissioning in his lab. James will also continue to lead the VMI-based approach as well as many of the experimental simulations. Both James and Peter will have regular interactions with both the PI and the RA to help inform and guide the work.

The Research Associate (RA) will spend 100% of his or her time over the 2-year proposal period. In Year 1, he or she will use existing computational infrastructure to simulate the electron Time-of-Flight detector array, iterating the design in order to minimize the cross-talk between eTOF spectrometers in the array. Furthermore, he or she will identify and order the electron multiplier assemblies and digitizer electronics that work together to optimize energy resolution and spectral window. Upon receipt, the RA will complete the initial 2 prototype eTOF detectors and begin the test/iterate cycle. In Year 2, the RA will focus on completing construction of the prototype detector arrays and use existing laser facilities to demonstrate the new capabilities. He or she will work with the LCLS Staff to secure time during early testing of the new soft x-ray undulators to demonstrate the improved window and resolution capabilities.

In addition to the budgeted RA, the PI has identified a Banting post-doctoral fellow, Audrey Therrien, who has needed expertise in charged particle simulations with GEANT4 as well as integrated circuit design and FPGA expertise. The PI has also identified a Stanford graduate research fellow in Electrical Engineering, Matt Feldman, who holds expertise in digital signal processing in FPGA as well as deploying machine learning models (inference engines) to FPGA.

The PI will continue to leverage his group alumni fellows, most notably Wofram Helml (Univ. Dortmund), Anton Lidahl (Quamcom Inc.), and Markus Ilchen (Euro XFEL/Univ. Kassel) who, along with then graduate student Nick Hartmann (Coherent Inc.), formed the original attosecond angular streaking team within the PI's research group. The PI expects these collaborative relationships to continue,

having grown to include also a long standing collaboration with Andreas Galler who is currently organizing the EuroXFEL angular streaking effort as well as with Reinhard Keinberger of TU Munich/MPQ Garching Germany who is a familiar name in attosecond laser science. Furthermore, the ongoing close relationship with Thomas Feurer of University of Bern Switzerland will ensure continued development of the attosecond angular streaking retrieval algorithms.

Expected publications

- "Attoclock Ptychography" – Tobias Schweizer *et al.* [61],
- "Online, single-shot characterization of few-femtosecond X-ray temporal pulse substructures at free-electron lasers via angular streaking" – Rupert Heider *et al.*,
- "Machine Learning enabled x-ray pulse reconstruction" – Gregor Hartmann *et al.*,
- "Machine Learning for streaming x-ray pulse characterization: spectrum, polarization, and time,"
- "Shaped attosecond x-ray FEL pulses for nonlinear x-ray science,"
- "Direct observation of laser-mixing of valence electronic symmetries."

Appendix 1: Biographical Sketch

Ryan Coffee

Senior Staff Scientist
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Education and Training

Research Associate		SLAC National Accelerator Laboratory	06/2006–04/2009
Ph.D.	Physics	University of Connecticut	06/2006
M.S.	Physics	University of Connecticut	12/2001
B.S	Physics	University of Arkansas	06/1999
B.A.	Philosophy	University of Arkansas	06/1999

Research and Professional Experience

11/2017–present Senior Staff Scientist, LCLS Science Research and Development & The PULSE Institute
04/2009–10/2017 Staff Scientist, LCLS Laser Division & The PULSE Institute, SLAC
Spectral and spectrogram encoding of relative x-ray arrival time, sub-10 fs pulse generation for FEL multiplicative seeding and for time resolved photo-chemistry, optical and THz laser streaking techniques at the LCLS, angle-resolved double- and single-core hole spectroscopy of impulsively-aligned molecules, x-ray pump/x-ray probe experiments at LCLS, x-ray pulse shaping for multi-dimensional x-ray spectroscopy, gas phase ultrafast electron diffraction, LCLS experimental laser facility installation and commissioning
06/2006–04/2009 Research Associate, PULSE Institute
Coherent control of rotational wave-packet motion in ambient nitrogen and iodine.
01/2006–06/2006 Research Associate, University of Michigan
Participation in two of the final SPPS experiments
09/1999–06/2006 Research Assistant, Department of Physics, University of Connecticut
Two-color pump-probe optical experiments with nitrogen, molecular vibrational wave-packet motion on laser induced potential energy surfaces, ion time-of-flight spectroscopy, vuv-fluorescence spectroscopy of selective high-order multi-photon absorption in N_2, transient absorption spectroscopy.

Selected publications

1. *Attoclock Ptychography* Schweizer, Tobias and Brügmann, Michael H. and Helml, Wolfram and Hartmann, Nick and **Coffee, Ryan** and Feurer, Thomas Applied Sciences, **8** 1039 (2018)
2. *Attosecond timeenergy structure of X-ray free-electron laser pulses* N. Hartmann and G. Hartmann and R. Heider and M. S. Wagner and M. Ilchen and J. Buck and A. Lindahl and C. Benko and C. Bostedt and J. Gruenert and J. Krzywinski and J. Liu and A. Lutman and A. Marinelli and T. Maxwell and A. Miahnahri and S. Moeller and M. Planas and J. Robinson and J. Viefhaus and T. Feurer and R. Kienberger and **R. N. Coffee** and W. Helml Nature Photonics, **12** 215 (2018)

3. *Characterizing isolated attosecond pulses with angular streaking* Siqi Li and Zhaoheng Guo and **Ryan N. Coffee** and Kareem Hegazy and Zhirong Huang and Adi Natan and Timur Osipov and Dipanwita Ray and Agostino Marinelli and James P. Cryan Optics Express, **26** 4531 (2018)
4. *Ultrashort Free-Electron Laser X-ray Pulses* Helml, Wolfram and Grgura, Ivanka and Jurani, Pavle N. and Dsterer, Stefan and Mazza, Tommaso and Maier, Andreas R. and Hartmann, Nick and Ilchen, Markus and Hartmann, Gregor and Patthey, Luc and Callegari, Carlo and Costello, John T. and Meyer, Michael and **Coffee, Ryan N.** and Cavalieri, Adrian L. and Kienberger, Reinhard Applied Sciences, **7** 915 (2017)
5. *Optical Shaping of X-Ray Free-Electron Lasers* Marinelli, A. and **Coffee, R.** and Vetter, S. and Hering, P. and West, G. N. and Gilevich, S. and Lutman, A. A. and Li, S. and Maxwell, T. and Galayda, J. and Fry, A. and Huang, Z. Physical Review Letters, **116**, 254801 (2016)
6. *Circular dichroism measurements at an x-ray free-electron laser with polarization control* Hartmann, G. and Lindahl, A. O. and Knie, A. and Hartmann, N. and Lutman, A. A. and MacArthur, J. P. and Shevchuk, I. and Buck, J. and Galler, A. and Glownia, J. M. and Helml, W. and Huang, Z. and Kabachnik, N. M. and Kazansky, A. K. and Liu, J. and Marinelli, A. and Mazza, T. and Nuhn, H.-D. and Walter, P. and Viefhaus, J. and Meyer, M. and Moeller, S. and **Coffee, R. N.** and Ilchen, M. Review of Scientific Instruments **87** 083113 (2016)
7. *Polarization control in an X-ray free-electron laser* Lutman, Alberto A. and MacArthur, James P. and Ilchen, Markus and Lindahl, Anton O. and Buck, Jens and **Coffee, Ryan N.** and Dakovski, Georgi L. and Dammann, Lars and Ding, Yuantao and Drr, Hermann A. and Glaser, Leif and Grnert, Jan and Hartmann, Gregor and Hartmann, Nick and Higley, Daniel and Hirsch, Konstantin and Levashov, Yurii I. and Marinelli, Agostino and Maxwell, Tim and Mitra, Ankush and Moeller, Stefan and Osipov, Timur and Peters, Franz and Planas, Marc and Shevchuk, Ivan and Schlotter, William F. and Scholz, Frank and Seltmann, Jrn and Viefhaus, Jens and Walter, Peter and Wolf, Zachary R. and Huang, Zhirong and Nuhn, Heinz-Dieter Nature Photonics, **10**, 468 (2016)
8. *Generating femtosecond X-ray pulses using an emittance-spoiling foil in free-electron lasers* Ding, Y. and Behrens, C. and **Coffee, R.** and Decker, F.-J. and Emma, P. and Field, C. and Helml, W. and Huang, Z. and Krejcik, P. and Krzywinski, J. and Loos, H. and Lutman, A. and Marinelli, A. and Maxwell, T. J. and Turner, J. Applied Physics Letters **107**, 191104 (2015)
9. *High-intensity double-pulse X-ray free-electron laser* Marinelli, A. and Ratner, D. and Lutman, A.A. Turner, J. and Welch, J. and Decker, F.-J. and Loos, H. and Behrens, C. and Gilevich, S. and Miahnahri, A.A. and Vetter, S. and Maxwell, T.J. and Ding, Y. and **Coffee, R.** and Wakatsuki, S. and Huang, Z. Nature Communications **6**, 6369 (2015)
10. *Measuring the temporal structure of few-femtosecond FEL X-ray pulses directly in the time domain* Helml W. and Maier, A. R. and Schweinberger, W. and Grguraš, I. and Radcliffe, P. and Doumy, G. and Roedig, C. and Gagnon, J. and Messerschmidt, M. and Schorb, S. and Bostedt, C. and Grüner, F. and DiMauro, L. F. and Cubaynes, D. and Bozek, J. D. and Tschentscher, Th. and Costello, J. T. and Meyer, M. and **Coffee, R.** and Düsterer, S. and Cavalieri, A. L. and Kienberger, R. Nature Photonics, **8**, 950 (2014)
11. *Multicolor Operation and Spectral Control in a Gain-Modulated X-Ray Free-Electron Laser* A. Marinelli and Lutman, A. A. and Wu, J. and Ding, Y. and Krzywinski, J. and Nuhn, H.-D. and Feng, Y. and **Coffee, R. N.** and Pellegrini, C. Physical Review Letters **111**, 134801 (2013)
12. *Experimental demonstration of femtosecond two-color x-ray free-electron lasers* Lutman, A. A. and **Coffee, R.** and Ding, Y. and Huang, Z. and Krzywinski, J. and Maxwell, T. and Messerschmidt, M. and Nuhn, H.-D. Physical Review Letters **110**, 134801 (2013)

Synergistic Activities

Machine learning The PI is also PI of a Laboratory Directed Research and Development (LDRD) project entitled "Machine Learning for Data Reduction at LCLS-II: a path toward 1 MHz detection." That FY18-FY19 project uses the LCLS-II Arrival Time Monitor (ATM) as a surrogate for demonstration of analysis at the detector. That project is successfully informing LCLS and SLAC more broadly regarding the computing hardware and software configurations that will enable inference engines to rapidly digest high throughput data pipelines before the need to transfer via network to remote storage farms. Much of the algorithm development under that project can be leveraged to guide the design of the angular streaking inference engine design choices here.

X-ray pulse shaping The PI has played a central role in motivating and helping the development of the many electron bunch based methods for x-ray pulse shaping [17,20,21,24,27,45]. From performing the first double-slotted foil experiment for two x-ray pulses [75] to demonstrating optical carving of the electron bunch [17] and helping demonstrate multi-polarization multi-color operation [27], our group has helped to pioneer many of the pulse shaping schemes that have been developed thus far at the LCLS. On the active forefront of pulse shaping, he is uniquely positioned to develop the pulse shape diagnostics hand-in-hand with the accelerator R&D for which he is already an active contributor.

Molecular frame resonant Auger spectroscopy The scientific emphasis of the PI's efforts lie in time-resolved Auger and photo-electron spectroscopy. His emphasis throughout his career for "measure-and-sort" methods for electron spectroscopy allows for the direct application of the diagnostic techniques of this project to molecular science at xFELs. His history with the molecular frame [76–80] and ultrafast x-ray communities [3,5,50,51,75,81–85] ensure that the techniques developed herein will be widely and rapidly spread to the broader community in much the same way as his developments in xFEL/laser timing [86–89] have now spread globally.

List of collaborators and co-authors: (48 months)

Collaborators:

Lorenzo Avaldi	CNR-ISM, Rome	Christoph Hauri	SwissFEL PSI, Switzerland
Nora Berrah	Univ. of Connecticut	Dan Kane	Mesa Photonics, Albequerque
Martin Beye	HZB Berlin	Reinhard Kienberger	TU Munich
Christoph Bostedt	ANL	Jochen Küpper	CFEL, Hamburg
Marco Cammarata	Univ. of Rennes, France	Jerry LaRue	Chapman, Irvine CA
Adrian Cavalieri	CFEL Hamburg	Jon Marangos	Imperial College, UK
Martin Centurion	U. Nebraska,	Marc Messerschmidt	BioXFEL, Hamburg
Tilo Doeppner	LLNL	Michael Meyer	Euro. XFEL, Hamburg
Gilles Doumy	ANL	Catalin Miron	ELI-Delivery Consortium
Stefan Düsterer	FLASH DESY Hamburg	Thomas Möller	TU Berlin
Raimund Feifel	Univ. of Gothenburg, Sweden	Serguei Molodtsov	Euro XFEL, Hamburg
Thomas Fennel	Univ. Rostock, Germany Thomas	Anders Nilsson	Uppsala University, Sweden
Feurer	Univ. of Bern, Switzerland	Steve Pratt	ANL
Thornton Glover	Gordon & Betty Moore Found.	Artem Rudenko	Kansas State University
Jan Grünert	Euro. XFEL, Hamburg	Daniel Rolles	Kansas State University
Markus Gühr	Potsdam University, Germany	Nina Rohringer	U. of Hamburg, Germany
Marion Harmand	IMPMC-UPMC, Paris, France	Arnaud Rouzee	MBI Berlin
Janos Hajdu	Uppsala Univ. Sweden	Ilme Schlichting	MPI Heidelberg

Sharon Shwartz	Bar-Ilan University, Israel	Kiyoshi Ueda	Tohoku Univ., Japan
Klaus Sokolowski-Tinten	U. of Duisburg-Essen, Essen Germany	Joachim Ullrich	PTB Germany
		Jens Viefhaus	DESY
Thomas Tschentscher	Euro. XFEL Hamburg		

Graduate and Postdoctoral Advisors

G. Gibson (University of Connecticut), P.H. Bucksbaum (PULSE/Stanford)

Appendix 2: Current and Pending Support

Both current and pending support will be predominantly covered under the U.S. Department of Energy / Stanford University Contract for Management and Operation of SLAC National Accelerator Laboratory with a small fraction under the National Institute of Health.

Current Support	LCLS-Soft X-ray Department	50%
	LCLS High Sensitivity Timing	20%
	LDRD Machine Learning for LCLS-II	20%
	NIH Time-of-Flight PET	10%
Pending support	same as current	

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Frank Scholz, Joern Seltmann, Patrick Gessler, Jan Grünert, Alberto De Fanis, Michael Meyer, André Knie, Stefan P. Moeller, Lorenzo Raimondi, Flavio Capotondi, Emanuele Pedersoli, Oksana Plekan, Miltcho B. Danailov, Alexander Demidovich, Ivaylo Nikolov, Alessandro Abrami, Julien Gautier, Jan Lüning, Philippe Zeitoun, and Luca Giannessi. Control of the polarization of a vacuum-ultraviolet, high-gain, free-electron laser. *Phys. Rev. X*, 4:041040, Dec 2014.

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Appendix 4: Facilities and Other Resources

There are two principle facilities identified for this project.

- Early commissioning and development experiments will be carried out in the PULSE Lab of James Cryan and also in the Arrillaga Science Center (ASC) as space becomes ready for occupancy.
- *In situ* testing and experiments will be carried in Hutch 1.1 of the Near Experimental Hall.

Analysis resources exist from both the SLAC-unix farm and the LCLS-unix farms including two FPGA-accelerated machine learning intensive commuting nodes that were originally purchased to support the PI's LDRD effort. Those compute nodes represent the guiding principle for how inference-based pre-analysis will unfold at LCLS-II. Our long collaboration with the data analysis and controls groups at LCLS not only allows the PI particularly early insight into the computing resources, but it also motivates his active pursuit of data compression and on-board analysis algorithms. This mutual benefit ensures the continued use and support for these computing facilities.

Office space will be available for team members with locations divided into available space in the building 901 LCLS office building with additional space for the Coffee, the RA, students and visiting scientists in the PULSE Institute of Building 40.

Additional Personnel

We expect yearly support at the roughly 120 hour level from AIR-TEC for electronics design and construction.

Appendix 5: Equipment

Required existing equipment includes: with associated laser systems:

- We will continue to work in collaboration with James Cryan for intermittent testing of the spectrometer array in the EUV PULSE Lab in Building 40 at SLAC.
- The R&D laser lab in the PSLB will be used for detector testing and construction as the space comes available in Fall 2018.

Other equipment available to the project include: Laser conversion for generation of 2 μm pulses as exists for LCLS-I.

Materials & Supplies

Year 1

1. Vacuum Chamber
2. 2x MCP electronics
3. 1x Digitizer + PCIe carrier, = 2x6GSps
4. 4U computer
5. Xilinx Zynq UltraScale+ RFSoC ZCU111 Evaluation Kit

Year 2

1. 2x Digitizer+PCIe carrier, gives a total of (3 digitizers total, each running 2x6GSps for total of 6 channels)
2. 4x MCP electronics
3. FPGA for streaming analysis Virtex
4. Miscellaneous vacuum and electronics M&S

Appendix 6: Data Management Plan

As stated in the Project Narrative, one of the central themes is to reduce the data load. The data that is accumulated as part of this project will be made broadly available both internally via SLAC/LCLS unix user account access and, by inquiry to the PI and his team, via coordinated data formatting and access based on containerized analysis algorithms and raw high-fidelity sample data along with the inference input, output, and inference model weights.

Data taken with the LCLS will be stored in accordance with LCLS policy also in SLAC Central Storage (or LCLS storage if in future LCLS moves the storage service). Access can then be granted by the PI also for any individual who obtains an LCLS user unix account.

The simulation based data will be housed in the SLAC Central Computing. The PI currently maintains a 1 TB/year subscription that will be incrementally increased up to a 5 TB/year storage, expanded when needed. Again, coordinated data formatting and access will be based on containerized (Docker or Singularity) analysis algorithms and raw high-fidelity sample data along with the inference input, output, and inference model weights.