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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DOE National Laboratory Announcement Number: **FWP#100498**

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Project Narrative

Introduction

The advent of x-ray free electron lasers (xFELs) has brought the ability to resolve ultrafast processes in molecular and material systems on their vibrational time and length scales [?, ?, ?, ?]. The next 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 the molecular electronic movie. Given the importance of understanding such electronic flow in photo-excited systems (Fig. 1), there is a strong desire to drive electrons into concerted coherent motion [?, ?, ?] and then probe the local electronic environment with time-resolved x-ray sepctroscopies. This has even led to direct funding of a major research effort focused specifically on attacking this regime with FEL sources [?]. Opening the field of attoscience to the xFEL machine, one could even imagine an attosecond resolved extension to the two-dimensional resonant Auger electron spectrosopies of Ref. [?].

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

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.

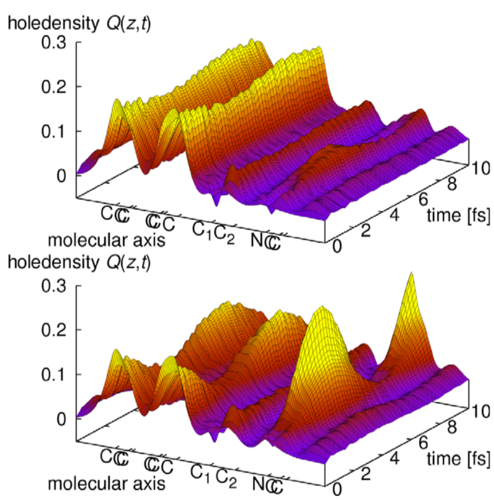

Figure 1: Hole migration in PENNA molecule following photoionization in the ground neutral molecular configuration (top) versus the C_2-C_2 20pm stretched configuration reproduced from Ref. [?].

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) [?, ?]. These sources are encroaching on this range [?, ?], however 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 [?, ?] with a particular push funded directly by the Office of Basic Energy Science aimed at LCLS-II implementation [?, ?].

Achieving the full capability of attosecond x-ray laser science will require the diagnosis and control of the x-ray spectral phase. 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. A continued progress on this front [?, ?, ?, ?, ?, ?, ?] will require full spectral phase, amplitude and polarization characterization as recently demonstrated in Ref. [?]. 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 restrict ourselves to two fundamental objectives:

1. We will perform the necessary reasearch and development required to eventually deliver an optimized angular array of electron Time-of-Flight spectrometers.

2. We will co-develop the requisite algorithms and machine learning compliments together with the detector electronics such in odred to optimally match the real-time analysis routines with detector electronics and computing hardware that push the tenchnology envelope.

Streaking for Xray Pulse Reconstruction

There is increasing momentum in the development of spectro-temporally shaped x-ray FEL pulses [?, ?, ?, ?, ?, ?, ?, ?, ?, ?] in response to the rising tide of demand [?, ?, ?, ?, ?, ?, ?]. The predominant method to characterize such novel temporal profiles is based on an x-band transverse accelerating cavity (XTCAV) [?] whereby the spent electron bunch is deflected horizontally, streaked in time by the phase of the transverse accelerating field. 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. This technique has been a critical tool for developing the recent x-ray FEL pulse shaping methods [?, ?, ?, ?]. Unfortunately, it indirectly measures the x-ray temporal profile by 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.

Since the inception of streaking as a pulse reconstruction method [?, ?] there have been great recent gains in diagnosing laser-based pulses that encroach on the soft x-ray regime [?, ?, ?]. Photo-electron streaking, a direct interaction, has the capability to measure the instantaneous temporal structure of x-ray pulses [?].

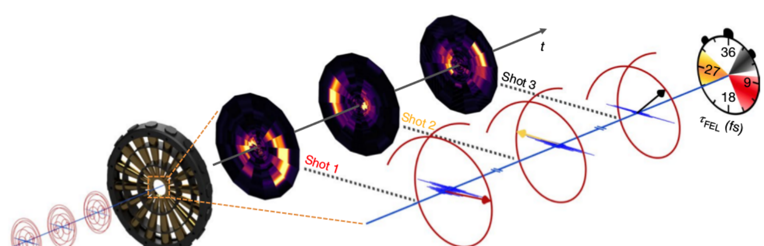

Figure 2: Schematic of angular photo-electron streaking. [?]

Some phase retrieval analysis require interferences with reference oscillators as in Refs. [?, ?], we favor a more flexible scheme that can also accommodate *in situ* x-ray experiments as well as the pulse retrieval diagnostic. We will therefore 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. [?]. In x-ray photo-electron streaking, a noble gas like neon is dressed by a strong long wavelegnth infrared or THz field [?, ?, ?]. The streaking field vector potential 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 [?, ?, ?, ?], the intensity profile of the photo-electrons versus their shifted energies can be mapped to the time-variation of the vector potential in a familiar "streak camera" interpretation, of course given 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 sensative to the exact focussing conditions of the streaking laser. However, in angular streaking, as depicted in Fig. 2, that vector potential is made to rotate by a using 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 [?]

We will leverage our long history with photo-electron streaking at the LCLS [?, ?, ?] by extending the attosecond angular streaking method of Refs. [?, ?] to the x-ray regime. Pulse-to-pulse variations at an FEL require single-shot measurements much like the velocity map imaging (VMI) [?] extension of attosecond angular streaking [?]. Although we have explored such a single-shot VMI solution for 120Hz operation [?], the requirement of a two-dimensional detector precludes high repetition rates. 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.

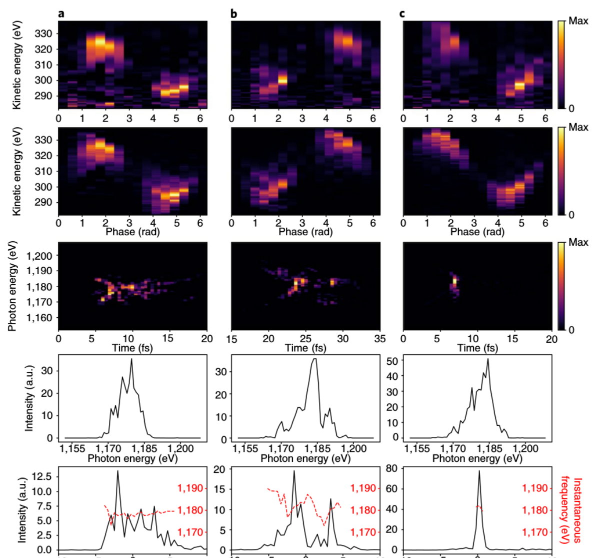

Figure 3: X-ray pulse shape retrievals reproduced from Ref. [?].

As an alternative, we repurposed what was originally considered for x-ray FEL polarimeter [?, ?, ?, ?] 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 [?]. The result was an x-ray pulse temporal reconstruction with 500 attoseconds resolution owing to the poor 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 also target its use for a full spectral and polarization reconstruction with better than 250 attosecond temporal resolution and 0.25 eV energy resolution.

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 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.

Attosecond pump-probe experiments

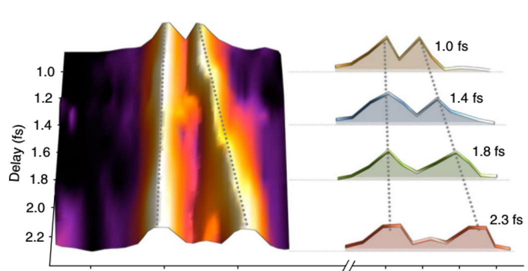

Figure 4: Reproduced from Ref. [?].

Many attosecond scale experiments are currently only enabled by high-harmonic generation (HHG) [?, ?, ?, ?, ?, ?, ?, ?]. These experiments could greatly benefit from the much higher brightness of an attosecond xFEL beamline [?, ?, ?]. They would effectively exchange the flux challenge of the HHG sources for the synchronization challenge of xFEL pulses. With the ability for x-ray pulse characterization at the attosecond level, one removes the synchronization challenge clearly 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 [?]. Roughly 1% of the SASE pulses measured consisted of only two spikes when running in low charge mode with emittance shaping. Such pulses can then be sorted, allowing a "measure-and-sort" x-ray pump/x-ray probe experimental paradigm.

In fact, if one prefers a weaker isolated attosecond pulse for a probe pulse, one could use a more traditional laser-based HHG source. Such pulses in the euv could be used to pump inner valence transitions in molecular systems, setting up rather pronounced coherent electronic motions, or simply used as broad-band transient absorption probes [?]. Then the attosecond x-ray pulses from the LCLS could be used to time resolve the valence occupation via time-resolved photo and Auger electron spectroscopy. In this case, 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.

Objectives

Optimized Detector Array

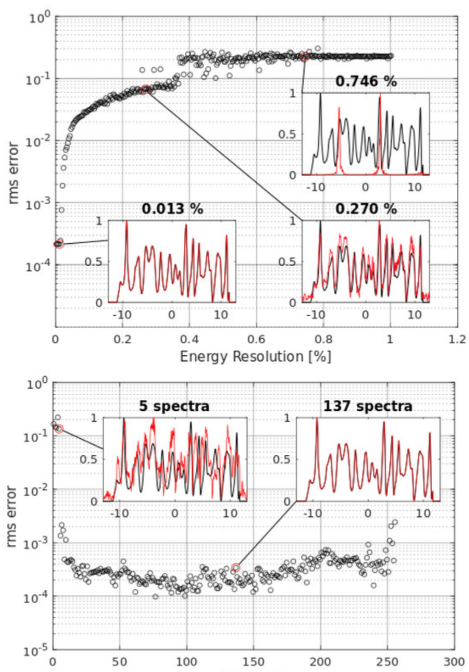

Figure 5: Reproduced from Ref. [?]. (upper) Xray pulse retrieval error increases dramatically for resolutions poorer than about 0.4%. (lower) The retrieval also fails when the angular sampling falls below 8 detectors.

The technical requirements in order to achieve the pulse retrievals as needed for accuarate reconstruction are not trivial to achieve. We are designing toward a single-shot diagnostic that reports the full temporal intensity, wavelength, and polarization distribution also with attoseconds resolution and at the highest repetition rate, up to 1MHz. Modeled after the original Jens Viefhaus, so called "Cookie Box" design, we propose a universal main chamber that accepts micro-channel plate based electron detectors in a 16-fold array.

Preliminary results of Ref. [?] were based on the original 16-fold detector array afforded 1 eV eTOF spectrometer resoltuion when working in the required single-shot current mode, not the synchrotron style high resolution counting mode. From Fig. 5(upper) we can see that the energy resolution for the streaked photoelectrons should ideally be in the sub 0.25% range. We therefore are targeting detectors that will have an energy resolution of 0.25 eV at up to 100 eV electron kinetic energy above the retardation voltage. The retardation voltage will allow us to view even high energy electrons, including Auger electrons, with the same high resolution. We note that the fine tuning of the sorts of novel FEL modes that allow for attosecond FEL experiments typically require a diagnostic of such 0.25 eV resolution of the spectral sub-spikes [?].

The resolution in pulse reconstruction depends also on the number of angular sample points per optical cycle. In Fig. 5(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 which is expected to provide a two-fold over-sampling of the angular dimension. Furthermore, by shifting the dressing laser frequency toward the near-infrared we can further improve the temporal resolution. Based on Refs. [?, ?, ?, ?] 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 to both improve the fractional bandwidth for a robust carrier shape and also to improve the temporal resolution while still preserving an appropriate window for pulse shape retrieval. This change 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 6 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 resoltuions competative with laser-based HHG state-of-the-art [?, ?].

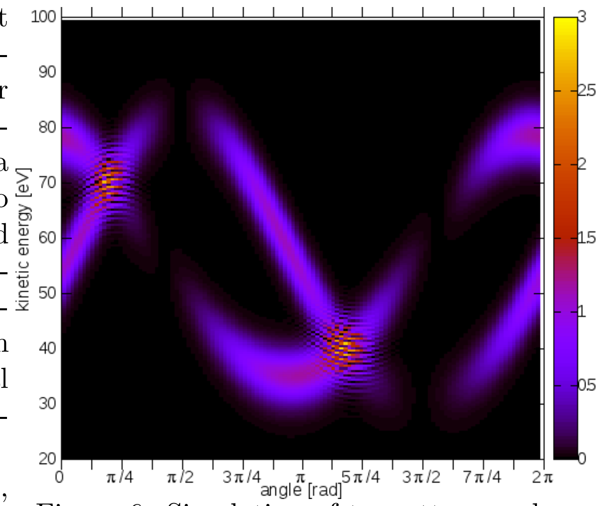

Figure 6: Simulation of two attosecond x-ray pulses separated by 4 fs dressed by a 4 μm streaking field, courtesy J. Cryan.

When diagnosing closely separated double pulses, the angular acceptance of the electron spectrometers are a principle concern [?]. We plan to optimize this angular acceptance such that we relax slightly the collection for the sake of preserving the resolution. We were able to reduce the sample density by 10-fold in Ref. [?] 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. This reduced collection will be partially compensated by a new chamfered-pore design for the microchannel plates that improves collection efficiency.

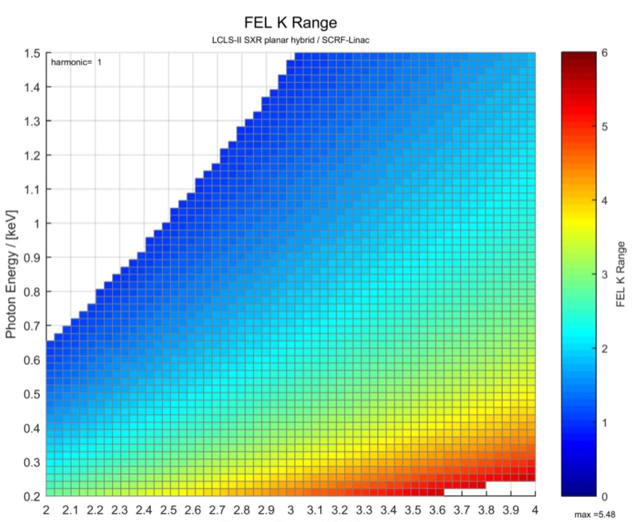

Figure 7: Soft x-ray undulator tuning range. [?]

Motivated by the great progress in multi-color FEL modes [?,?,?,?,?,?,?,?], we are capitalize on the two-fold over sampling in the angular dimension. The color separation between multi-pulses using the new soft x-ray undulator of LCLS-II could be made to interrogate different atomic species with absorption resonances separated by hundreds of eV. Our two-fold over sampling in the angular dimension allows us to fully retrieve the temporal, spectral, and polarization characterization of shaped multi-pulses [?, ?]. Figure 7 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 are designing for a novel configuration whereby individual detectors in the eTOF array can have vastly different retardation voltages. 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.

Furthermore, as laser based HHG isolated attosecond pulses come available as in Refs. [?, ?, ?, ?] one could use a helium buffer gas to enable angular streaking simultaneously of HHG-generated EUV attosecond pulses with x-ray attosecond pulses in the identical scheme as the x-ray/x-ray pairs discussed above. One could use the weaker HHG isolated attosecond pulses [?] as a supercontinuum probe up to the carbon K-edge. The LCLS-II would provide the much higher power and energy attosecond pump pulses for $1s \rightarrow$ valence resonant pump transitions. This would allow for strong pumping of valence electronic correlations from a chosen atomic site in the molecule well above nitrogen and oxygen, even fluorine and transition metal L-edges.

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 coallate the real-time sorted results prior to network transfer. Such a vision fuels out strong motivation to not only provide high repetition rate veto, but also event rate, low-latency sorting triggers.

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 200–500 ns of record length with a sample frequency of ideally about 10GS/s, one waveform for each of the 16 detectors. All together this would be comparable to analyzing one 256×256 image every microsecond. Detailed in Ref. [?], 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 method of projections [?] are computationally expensive and would not likely allow for MHz or even 100 kHz repetition rate. We therefore plan to reserve these and other retrieval methods [?, ?] for the generation of so-called "ground truth" for a sub-set of full fidelity recorded x-ray pulse shapes.

We plan to use the full fidelity 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.

Schedule

Organization of Major Activities

Period 0.0: 7/1/2018 – 12/31/2018
Test 2μm streaking with attosecond pulses from LCLS-I using Axial VMI [?].
Publication(s): "Attoclock Ptychography" – Tobias Schweizer *et al.* [?]

Table 2: Timeline of the R&D schedule. Project Years (PY) are sub-divided into 6 month blocks.

PY0.0	PY0.5	PY1.0	PY1.5
somethign	else	and	this
somethign	else	and	this

Period 0.5: 1/1/2019 – 6/31/2019
Design detectors for array, use existing data to develop algorithms for streaming analysis.
Implement on-board FPGA processing and demonstrate/simulate calibration learning, detail implementation plan for LCLS-II.
Publication(s): "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.*

Period 1.0: 7/1/2019 – 12/31/2019
Construct detector array prototype for Building 40 benchmarks and LCLS-II beneficial occupancy.
Publication(s): "Benchmarking/Novel algorithm for real-time x-ray pulse characterization at the LCLS-II."

Period 1.5: 1/1/2020 – 6/31/2020, instrument commissioning
Install array on LCLS-II and benchmark resolution with XAFS using 3 μm laser dressing of N_2O. **Publication(s):** "Streaming attosecond resolution 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."

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 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 the RA to help guide the work.

The RA will focus his or her on the design and integration of the chamber with the detector systems and will take responsibility for the detector construction. He or she will organize and lead benchmark tests of attosecond angular streaking at other facilities such as EuroXFEL and in the Cryan lab in building 40.

We intend to recruit two graduate students from Stanford University to participate in the project. We will seek funding through student research fellowships and LCLS-sponsored student outreach programs. The PI has identified one such funded post-doctoral fellow, Audrey Therrien, who has needed expertise in charged particle simulations with GEANT4 as well as integrated circuit design.

Related research by the PI has attracted numerous externally funded post-doctoral fellows and visiting scientists, most notably Wofram Helml (Marie Curie Foundation), Anton Lidahl (Wallenberg

Foundation), and Markus Ilchen (Volkswagen Foundation). We expect these collaborative relationships to continue. In particular, the close collaboration with the Kienberger Group of TU Munich/MPQ Garching Germany is often financially supported through the Bavaria California Technology Center (BaCaTeC) program [?].

Appendix 1: Biographical Sketch

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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

01/2014–present Staff Scientist, PULSE Institute
04/2009–present Staff Scientist, LCLS Laser Division, 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. *Optical Shaping of X-Ray Free-Electron Lasers* A Marinelli, **R Coffee**, *et al.* Physical Review Letters, **116**, 254801 (2016)
2. *Polarization control in an X-ray free-electron laser* AA Lutman . . . **R Coffee**, *et al.* Nature Photonics, **10**, 468 (2016)
3. *Generating femtosecond X-ray pulses using an emittance-spoiling foil in free-electron lasers* Y Ding, C Behrens, **R Coffee**, *et al.* Applied Physics Letters **107**, 191104 (2015)
4. *High-intensity double-pulse X-ray free-electron laser* A Marinelli, . . . **R Coffee**, *et al.* Nature Communications **6**, 6369 (2015)
5. *Measuring the temporal structure of few-femtosecond FEL X-ray pulses directly in the time domain* W Helml, . . . **R Coffee**, *et al.* Nature Photonics, **8**, 950 (2014)

6. *Sub-femtosecond precision measurement of relative X-ray arrival time for free-electron lasers* N Hartmann,, *et al.* **RN Coffee** Nature Photonics **8**, 706 (2014)
7. *Spectral encoding method for measuring the relative arrival time between x-ray/optical pulses* M Bionta, *et al.* **R Coffee** Review of Scientific Instruments, **85**, 083116 (2014)
8. *Multicolor Operation and Spectral Control in a Gain-Modulated X-Ray Free-Electron Laser* A Marinelli,, *et al.* ...**RN Coffee**, and C Pellegrini Physical Review Letters **111**, 134801 (2013)
9. *Experimental demonstration of femtosecond two-color x-ray free-electron lasers* AA Lutman, R Coffee, *et al.* Physical Review Letters **110**, 134801 (2013)
10. *Spectral encoding of x-ray/optical relative delay* Mina R. Bionta,, *et al.* **R. N. Coffee** Optics Express, **19**, 21855 (2011)

Synergistic Activities

Ultrafast electron diffraction (UED) The PI has recently become intimately involved with the use of ultrafast electron diffraction (UED) in order to merge spectroscopic studies at the LCLS with the structural sensitivity of electron diffraction [**?**,**?**]. The merger of the two experimental paradigms is enabled by induced time-domain coherent molecular motions that in-turn produce concerted fluctuations in both x-ray spectra and in electron diffraction features. By pattern recognizing these common-mode fluctuations, one can merge experimental results as if the two experiments were performed simultaneously. The PI is therefore keenly attuned to need for exquisite synchronization at high energy ultrafast electron diffraction sources. The successful outcome of Objective **??** would be immediately applicable to high energy electron diffraction facilities.

Deep ultra-violet During period two, the LCLS facility will be down. Here we will draw on our existing research program investigating routes to broad-band tunable deep uv. This collaborative work together with the Keinberger group of TU Munich produced a Marie-Curie Fellowship project of Wolfram Helml to develop ultra-broadband, sub-10 fs duration deep ultraviolet pulses. This project later led to a Masters student research project of Patrick Rupprecht to develop the required pulse characterization in the deep uv regime. This deep ultraviolet will be used as a surrogate for the x-ray pulse during the LCLS down time from August 2018 – August 2019. This existing setup will continue to serve as a surrogate source for testing and debugging new concepts and materials.

Stanford Medical The PI has recently opened a new collaboration with the group of Dr. Craig Levin in the Division of Nuclear Medicine at Stanford. Through this collaboration we expect a mutually beneficial investigation into novel scintillation materials for improving the cascade time and the signal per dose ratio. Such a time resolution would greatly improve the spatial resolution of the PET scan per unit time spent undergoing the scan. Ideally one could imagine then making such scans in short time-series if each scan only takes a few seconds. Then one can observe the dynamic processes in e.g. brain activity.

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 [**?**,**?**,**?**,**?**,**?**,**?**]. From performing the first double-slotted foil experiment for two x-ray pulses [**?**] to demonstrating optical carving of the electron bunch [**?**] and helping demonstrate multi-polarization multi-color operation [**?**], 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. This close overlap with the LCLS Accelerator R&D group is in fact what allows for the shared resources such as the two Project Scientist/RA positions to be shared so intimately. These positions will be fundamentally interwoven between the development of novel FEL methods and the attosecond diagnostics required to interrogate those methods.

Machine learning Such so-called "deep learning" is used to interrogate the output of the artificial neural networks to check the physical interpretation of how the output is determined [**?**]. Our preliminary test, shown in Fig. **??**, uses the neural network classifier to sort LCLS shots based on a CNN predicted two-color spectral distribution. The sorted shots are shown at right and an example of an XTCAV image is shown in the top left of Fig. **??**. Guided back propagation is then used to interrogate the neural network about which regions of that image influenced the sorting most strongly, and this is shown in the lower left. These two regions indeed indicate the loss of electron energy due to the lasing process, thus boosting our confidence that the CNN is sorting based on valid physical properties of the x-ray pulse, photon energy separation in this case.

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

Collaborators:

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Nora Berrah	Univ. of Connecticut
Martin Beye	HZB Berlin
Christoph Bostedt	ANL
Marco Cammarata	Univ. of Rennes, France
Adrian Cavalieri	CFEL Hamburg
Martin Centurion	U. Nebraska,
Tilo Doeppner	LLNL
Gilles Doumy	ANL
Stefan Düsterer	FLASH DESY Hamburg
Raimund Feifel	Univ. of Gothenburg, Sweden
Thomas Fennel	Univ. Rostock, Germany Thomas
Feurer	Univ. of Bern, Switzerland
Thornton Glover	Gordon & Betty Moore Found.
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Marion Harmand	IMPMC-UPMC, Paris, France
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Reinhard Kienberger	TU Munich
Jochen Küpper	CFEL, Hamburg
Jerry LaRue	Chapman, Irvine CA
Jon Marangos	Imperial College, UK
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Jens Viefhaus	DESY

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

Appendix 3: Bibliography and References Cited

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 Photon Science Laboratory Building (PSLB).
- *in situ* testing and experiments will be carried in Hutch 1.1 of the Near Experimental Hall and in the Photon Science Laboratory Building (PSLB).

Analysis resources exist from both the SLAC-unix farm and the LCLS-unix farms. 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 is 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 one graduate student, recruited from Stanford University, who will work on this project. This student will participate via the student outreach programs of LCLS at SLAC.

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 (15.8)
3. 1x 22k Digitizer + PCIe carrier, = 2x6GSps
4. 4U computer (6K)
5. Xilinx Zynq UltraScale+ RFSoC ZCU111 Evaluation Kit (9)

year 2

1. AIR-TEC electronics technician 120Hrs
2. 2x 22k Digitizer+PCIe carrier, gives a total of (3 digitizers total, each running 2x6GSps for total of 6 channels)
3. 4x MCP electronics (31.5)
4. FPGA for streaming analysis Virtex at 7k
5. Miscellaneous vacuum and electronics M&S (14k)

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, externally via coordinated data formatting and access FTP.

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 laser lab based data will be housed in the SLAC Central Computing. The PI currently maintains a 1 TB/year subscription and that will be incrementally increased up to a 5 TB/year storage, expanded when needed. The data will be made broadly available by SLAC unix account access and externally by contacting the PI and coordinating an FTP service of the data.