LCLS | Linac Coherent Light Source

In House Proposal – Run 20

LCLS Department: SXD

Title:
Multi-photon x-ray spectroscopy via stochastic strong fields

LCLS Lead/Leads *(department or scientist)*:
Ryan Coffee, Razib Obaid, Andrei Kamalov

LCLS Collaborators:

Ruraidh Forbes, James Cryan, Taran Driver, Siqi Li, Dan Ratner, Alberto Lutman, Erik Hemsing

Non-LCLS/External Collaborators *(names, affiliations, and* ***description of roles****)*:

Jan Michael Rost (MPG-Dresden) (Theory), Ulf Saalmann (MPG-Dresden) (Theory), Sajal Giri (formerly MPG-Dresden) (Theory)

Proposal Criteria
(select all criteria that are relevant for this proposal)

☐	Proposed activities **are outside** the scope or guidelines for a regular PRP proposal
X	**Commissioning & Technical Development**
☐	**Career Development Opportunity Proposal:** Developing LCLS staff scientists and expertise to remain at the forefront of their fields. Describe in detail how the proposed activity will advance the scientific career of the proposer.
X	**Developing or demonstrating new experimental methods, technical capabilities, or instrumentation** with clear potential to enhance the science impact, and exploit the unique capabilities of LCLS. (Links to the LCLS strategic development plan, existing programs, initiatives, or development efforts e.g. R&D, L2S-I, XIP, AIP, OIP etc. should be clearly articulated, where appropriate.)
X	**Demonstration experiments** to develop a **new user community** and/or **establish a new area of science** where LCLS will have a significant impact.
☐	**Enhancing the operational effectiveness of LCLS**

Justification for criteria selection

This proposal will demonstrate the use of statistical methods on SASE pulses (e.g. machine learning) for reconstructing nonlinear spectra from so-called "noisy" sources. This idea was recently shown in simulation by Giri, Saalmann, and Rost [1-2]. The experimental demonstration requires well defined resonant absorption in the x-ray regime which in fact the CookieBox group has recently shown in Neon for autoionizing states. Therefore we have preliminary evidence that the Rydberg levels in atomic Neon would serve as narrow resonance features for demonstrating this stochastic pulse version of nonlinear spectroscopy, a new method that uniquely leverages the eventual MHz operation of LCLS-II. The broader field of multi-photon physics will thus be welcomed given the innovation of the "spectral purification" method.

Nonlinear spectroscopies often circumvent the occlusion of features that commonly occurs when background signals overwhelm the desired weak features for conventional methods. This inspirational demonstration would trigger a new methodology that capitalizes on the naturally stochastic structure of FEL pulses.

Additionally, this program will further commission the fully outfitted CookieBox angular array of spectrometers as will be needed before its introduction to the general user community at high repetition rates.

LCLS | Linac Coherent Light Source

In House Proposal – Run 20

Technical and Experimental Summary [1]

Instrument	CookieBox
Number of Shifts	5
Detailed shifts justification	1. Calibrate the full 20 ToF CookieBox, Recover Neon single photon resonances 2. High statistics low-energy electrons 3. High statistics high-energy electrons 4. High statistics interleaved low-high correlations 5. w-3w pathway interference effects
Std. configuration	Y/N : Probably not (if Yes specify :)
Scheduling constraints (end of the run, near a user experiment, etc.)	End of run 20 would be helpful for time constraints of receiving the 2nd set of 10 ToFs.
Multiplexing	Y/N : (if Yes specify multiplexing scheme :) Will be changing photon energy and running "Alberto mode" in data taking shifts, but Calibration shifts (1-2/3) are OK with multiplexing.
Photon energy (range)	867 +/- 25, 433 +/- 25, 290 +/- 25
Pink/ Mono/CCM	pink or mono : pink
Operation mode (SASE, seeded, other)	SASE using Alberto's scheme for chicane delay to limit the lasing to small portion of electron bunch as in June 2021, few longitudinal mode conditions
Lasers parameters	None
Resources required (M&S, equipment, manpower, etc.)	8-10 more ToFs for the CookieBox as per the XIP project.
Additional information	

[1] IH proposals considered for beamtime must submit the same technical questionnaire required for standard user experiments

Narrative: two-page limit, excluding references

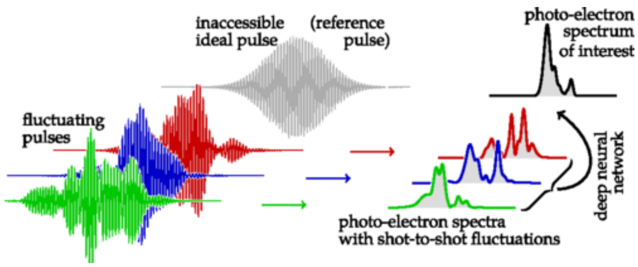


Figure 1. Reproduced from Ref. [1]. Noisy incident spectra cause fluctuations in the measured signal. Notably the fluctuating relative contributions for the partial wave patterns allow the deep neural network to lock onto correlation and thus “predict” what the purified nonlinear spectrum would be.

We will demonstrate how stochastic field fluctuations, that are natively produced at SASE FELs, can be used in combination with machine learning methods to uncover nonlinear multiphoton resonant effects in atomic systems. We will test the hypothesis that so-called “spectral purification” [1-2] can reveal nonlinear resonant absorption. This experiment will reveal the extent to which the CookieBox end station, with a fully outfitted array of 20 Time-of-Flight spectrometers, can capture nonlinear atomic and molecular spectroscopic methods.

The biggest unknown to be addressed is that simple experimental methods for creating moderately short, e.g. few to ten femtosecond, x-ray pulses can nevertheless compete with highly specialized FEL modes for attosecond pump-probe style experiments. The currently proposed scheme, based on the *in silico* demonstration of Refs [1-2], would uniquely leverage the high repetition rate of LCLS-II together with the data intensive machine learning paradigm to reveal signals that are otherwise too weak to detect conventionally. It is often the case that minor excitation channels evade conventional linear spectroscopies. There have been a number of attempts to show multi-photon absorption in the x-ray regime to marginal success, but one more recent (as yet unpublished) method required a heroic effort and truly impressive x-ray pulse energies. Here we plan instead to follow the logic that resonance, rather than bound--free transitions, will concentrate the oscillator strength for the target bound--bound transition. In this case, if a pulse consists of a single SASE spike, approximated by a flat spectral phase of ~1-2 eV width, then narrow transitions gain from all available spectral contributions across the full bandwidth. Since the number of single SASE spike shots is exceedingly infrequent, we will use a deep neural network, trained on sufficiently similar but computationally simple example simulations, in the way of Giri, Saalmann, and Rost [1-2] to reveal the two- and three-photon x-ray absorption.

Our detection channel will be the low energy autoionization features that follow initial spectator Auger-Meitner decay of the Neon 1s--np resonant excitation followed by inner-valence relaxation such that the Rydberg electron finds itself free with 10-30 eV of energy above the barrier. These features are in fact visible for the single photon case at right in Fig. 2.

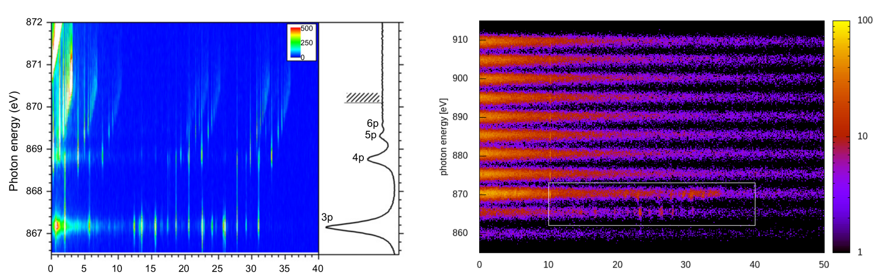
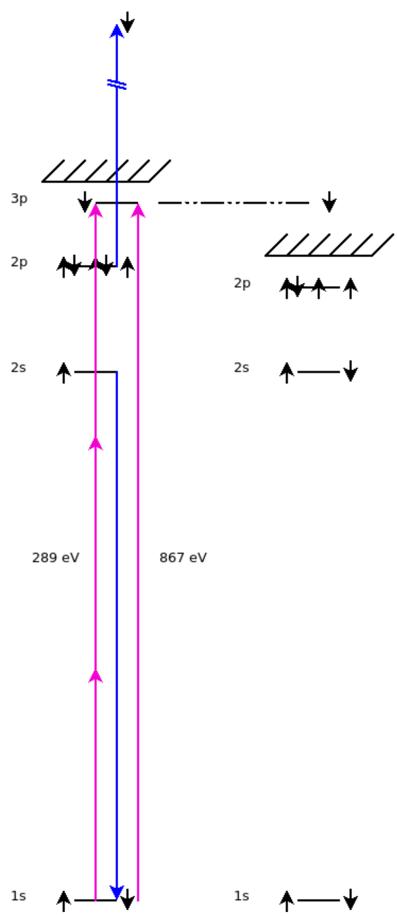


Figure 2 (left) Reproduced from Ref. [5]. (right) Preliminary results from June 2021 indicating 1s-3p excitation and subsequent auto-ionization bin the 30-40eV range.

Why is this important or urgent? Why now in Run 20?

This experiment is timely since it will form the basis for high repetition rate, data intensive methods for the imminent LCLS-II turn on. This method is an x-ray only method and therefore is free to run at the highest achievable repetition rate of the FEL. Since the stochasticity is ideally restricted to the few femtoseconds regime, we expect that very high peak fields can nevertheless be achieved for the highest repetition rates. We will only enable partial lasing of the electron bunch as done for the June 2021 CookieBox beamtime. Furthermore, this method will demonstrate the unique ability of CookieBox to disentangle linear and nonlinear ionization processes from a shot-to-shot basis of a high repetition rate FEL.

The June 2021 beamtime served as the initial demonstration of spectral resolution and resonant feature identification. Figure 2 (right) shows a progression of narrow electron spectral features in the range of 30-40eV that appear only for 875-880eV x-ray excitation energies. These features come about by autoionization of the Rydberg excitation induced by spectator decay of the threshold k-shell photoionization of Neon. These excitations are of 1s→np character as per the single photon excitation pathways shown in Figure 2 (right).



Detailed Experimental Description

We will scan the incoming photon energy through the Rydberg resonances 1s--np to recover the resonant features found in June 2021. Upon identification of such, and careful calibration of the incoming FEL energy, we will shift the photon energy to ½ the resonance and look for indication of the 1s--ns(nd) Rydberg series. In this case, we will vary the gas attenuator, preferentially attenuating the fundamental, in order to confirm that signal is due to the nonlinear interaction rather than the second harmonic of the FEL. We will then shift the photon energy to ⅓ of the resonance and look for the three-photon signal. If time allows in the final shift, we will attempt to enhance 3rd harmonic lasing from the FEL and vary slightly the undulator with the hope of measuring an excitation pathway interference for the 1s--np transition via one or three photons as per the seminal Ref. [4] indicated at left.

Impact

The impact will be to introduce nonlinear methods that have been employed in the optical regime [3] to the x-ray regime. This will also demonstrate a comprehensive and robust machine learning method to filter out various photoionization physics from such noisy spectra expected to be a bottleneck for high repetition rate FEL. Aside from attosecond angular streaking, the optimized mu-metal CookieBox as designed is ideally suited to such methods bringing stochastic pulse statistical analyses together with angle resolved partial wave spectral decomposition. This experiment will showcase its use for femtosecond scale nonlinear x-ray spectroscopy.

References

[1] Sajal Kumar Giri, Ulf Saalmann and Jan Michael Rost, “Purifying electron spectra from noisy pulses with machine learning using synthetic Hamilton matrices,” Phys. Rev. Lett. **124**, 113201 (2020)

[2] Sajal Kumar Giri, Lazaro Alonso, Ulf Saalmann and Jan Michael Rost, “Perspectives for analyzing non-linear photo-ionization spectra with deep neural networks trained with synthetic Hamilton matrices,” Faraday discuss. **228**, 502 (2021)

[3] Xiaoji G. Xu, Stanislav O. Konorov, John W. Hepburn, and Valery Milner, “Noise autocorrelation spectroscopy with coherent Raman scattering,” Nature Physics **4**, 125–129 (2008)

[4] Ce Chen, Yi-Yian Yin, and D. S. Elliott, “Interference between optical transitions,” Phys. Rev. Lett. **64**, 507 (1990)

[5] Yusuke Tamenori and Isao H Suzuki, “Multiplet exchange Auger transitions following resonant Auger decays in Ne 1s photoexcitation,“ J. Phys. B: At. Mol. Opt. Phys. **47** 145001 (2014)

[6] S. Stock, R. Beerwerth, and S. Fritzsche, “Auger cascades in resonantly excited neon,“ Phys. Rev. A **95**, 053407 (2017)