# **PROGRESS REPORT ON POPULATION INVERSION-BASED X-RAY LASER OSCILLATOR**

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### Abstract

The population inversion X-ray Laser Oscillator (XLO) is a fully coherent, transform limited hard X-ray source. It operates by repetitively pumping inner-shell atomic transitions with an XFEL, in a closed Bragg cavity. XLO will produce very bright monochromatic X-ray pulses for applications in quantum optics, X-ray interferometry and metrology. We report the progress to build the first XLO operating at the copper alpha line, using LCLS 9 keV SASE X-ray pulses as a pump.

### **INTRODUCTION**

X-ray free electron lasers (XFELs) offer tremendous improvement in peak brightness, compared to other X-ray sources. Yet, the temporal and spectral coherence of hard Xray XFEL pulses remains poor. The next generation XFELs is posed to tackle this problem, e.g. by building a cavitybased XFEL. Recently, an alternative approach, similar to a classical laser, was proposed [1]. In this approach, an XFEL pulse (5-10 keV) serves as a pump, creating population inversion and resulting in highly coherent amplified spontaneous emission (ASE) radiation [2-5]. This radiation can be further filtered and returned with an X-ray cavity for subsequent seeding, similar to a classical laser oscillator; see Fig. 1. Our initial studies based on simulations suggest that total of four pump pulses can result in a fully coherent, transform limited pulse [1].

We have recently started a program to develop a population inversion X-ray laser, called XLO, at SLAC, using LCLS to provide the XFEL pulses. For this program we have selected the copper K $\alpha_1$  (8048 eV) fluorescence for the first round of experiments. As our gain medium we chose a copper nitrate solution delivered as a high-speed liquid jet. Creating ASE processes in the hard X-ray range requires an X-ray pump with high peak power. This is achieved at the CXI nanofocus station at LCLS, using KB mirrors that provide a beam size of less than 200 nm. The goal of the first experiment in this program, shown schematically in Fig. 2, is to a) observe ASE from our copper nitrate gain medium, and b) determine the minimum time separation between the X-ray pump pulses (7 - 35 ns) that is required for the gain medium to recover from the impact of the firs pulse. This minimum time separation depends how fast the

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Figure 1: X-ray laser oscillator (XLO) schematics. Please refer to [1] for the detailed XLO concept description.



CC BY 3.0 licence (© 2021). Any distribution of this work must maintain attribution to the author(s), title of the work, publisher, and DOI Figure 2: Layout of the first experiment: 9 keV XFEL radiation is focused onto a liquid jet of cupric nitrate. The resulting radiation is dispersed with a Si (220) crystal and registered at the spectrometer.

gain medium can be replaced, and we were able to change this parameter by varying the jet velocity between 10 and 100 m/s. (Higher velocities will be achievable in the next experiment.) The diagnostic for the experiment uses three diodes capable of time resolving the pump pulses. Diode 1 the was used in forward scattering geometry and diodes 2 and 3 in the direct beam geometry.

## LCLS-HXR PERFORMANCE

be used under For the first round of experiments, we have utilized beamtilt compensation scheme [6], to obtain 5 kA, 20 fs long electron bunches generating up to 2 mJ, or 100 GW peak power, X-ray pulses at 9 keV photon energy. This marks the highest power to date with the new LCLS HXR undulator.

Two XFEL pulses, separated by 7, 14, 28 and 35 ns, have been generated. Laser heating of both electron bunches has been used to obtain two XFEL pulses with similar pulse Content energy. Two fast diodes were used to measure the pulse

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energies of the two pulses. Diode 1 was placed sideways of the beam in a scattering geometry to measure the pulse energies of the incident pulses. Diode 2 was placed in the forward direction downstream of the sample jet to measure the pulse energy transmitted through the gain medium (see Fig. 2). The raw wave forms from Diode 1 were used to tune the two-bunch performance of LCLS, by adjusting the timings and phases of the individual linac RF stations. (See [7-11] for more details on the LCLS multi-bunch mode of operation.) Figures 3 and 4 illustrate the XFEL performance of the two electron bunches in the undulator. One can see the anti-correlation in XFEL intensity in the X direction, which is difficult to correct without a bunch-by-bunch trajectory control system. Such a system is currently under development at LCLS [11].



Figure 3: X and Y orbit displacements in the undulator line for the first and second electron bunches, 35 ns apart, as a function of their XFEL performance.



Figure 4: LCLS-HXR double bunch time-resolved profile registered with a fast rise-time diode.

#### **OBSERVATION OF ASE**

Accomplishing goal a) of this experiment, we were able to generate copper  $K\alpha_1$  ASE radiation with 20 fs XFEL pulses using a 200 µm diameter jet at CXI; see Fig. 5 We believe that this is the first copper  $K\alpha_1$  ASE observed from a liquid solution. The gain was not as strong as previously observed for manganese and increasing the XFEL pulse duration to 40 fs did not produce ASE radiation. We are working to further increase the peak power of the XFEL pump pulses. Our previous studies on manganese suggest that a factor of two increase will enhance the copper  $K\alpha_1$  ASE signal by several orders of magnitude.

Another important consideration is the number of oxygen atoms in the solution. There are 25 Oxygen atoms and 20 Oxygen atoms for 3 and 4 molar cupric nitrate concentrations respectively. Approximately 20 Oxygen atoms absorb as much as one copper atom. Thus increasing the concentration and therefore reducing the number of oxygen atoms is important. We found that in practice we couldn't use jet with higher than 5 molar concentration, due to spontaneous crystallization issue. It indicates, that in order to increase the number of atoms per unit volume, one must consider a solid target.



Figure 5: Multi-shot average of the registered Cu ASE radiation at 8048 eV in a 4 molar of cupric nitrate solution.

#### JET PERFORMANCE

A solution of cupric nitrate was prepared in 3 and 4 molar concentrations (1.656 and 2.112 Cu atoms per nm<sup>3</sup> respectively) and delivered to the interaction point at low and high speeds via 160 µm and 200 µm diameter nozzles. Detailed numerical simulations of the XLO jet are discussed in Ref. [12]. The jet was imaged with a 640 nm pulsed laser ( $\tau = 6$  ns). Jet images (see Fig. 6) reveal the ionization channel created by the XFEL pulse. The ionization channel photons are short-lived, few fs, and overlap on the camera image, which has along exposition time. As expected, the jet is disrupted by the impinging XFEL pump pulse forming a gap [13–15]. We observed that the gap center is moving at approximately the jet velocity.

We attempted to produce jets with up to 100 m/s velocity. This velocity was limited by the viscosity of the nitrate (up to 8 mPa s for 4 molar solution) and the pump flow rate, and will be increased in the future. We found that high speed jets pose challenges in operation. First, the drain flowrate must match the nozzle flow-rate. This was achieved by applying negative pressure at the drain. Second challenge is the jet spray, which is produced due to high Reynolds number values and the resulting turbulence. Spray contributes to the background noise on the diode detectors and must be mitigated as much as possible. We are currently analyzing the data on jet recovery with double pulse pumping.

#### SUMMARY

In summary, we have performed the first experiment towards XLO development. We observed copper  $K\alpha_1$  ASE

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Figure 6: Liquid jet imaged in the transmission 640 nm light. The ionization channel and XFEL-induced gap are shown at times of 20  $\mu$ s apart. Jet velocity is about 10 m/s.

by creating a population inversion in a jet of cupric nitrate. Our next experiments will be dedicated to establish the gain medium recovery and characterize the diamond Bragg crystals of the cavity, in particular the required cooling for incoupling of the XFEL pump pulses at the first crystal (see top left in Fig. 1).

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