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XFELO Scientific Opportunities Retreat

Participants of the XFELO Science Workshop at SLAC National Accelerator Laboratory

The first "XFELO Science Workshop" (see https://portal.slac.stanford.edu/sites/conf_public/xfelo2016 for full list of participants and scientific program) was held at SLAC National Accelerator Laboratory, Menlo Park, CA, from June 29 to July 1, 2016. The main purpose was the discussion of scientific opportunities an XFELO extension of the LCLS-II complex in the future would enable. Chaired by Jerry Hastings (SLAC), it brought together more than 40 researchers from institutes around the world.

In modern experimental physics, high-gain X-ray free-electron lasers (XFELs), such as the Linac Coherent Light Source (LCLS) at SLAC National Accelerator Laboratory, SACLA in Harima, Japan and several others in various stages of completion have proven to be extremely powerful scientific tools for understanding light-matter interactions and a broad range of structural studies spanning the materials, chemical, biological and high energy density sciences. High-gain FELs are not the only approach to achieve intense x-ray laser beams. One alternative to produce coherent x-rays with narrow bandwidth was the XFEL oscillator (XFELO). This low-gain device uses multi-GeV electrons radiating in an undulator to produce X-ray photons that are then closed in a low-loss resonant cavity built from Bragg reflecting mirrors. In each pass, the photon pulse recirculates through the undulator to act as the seed for the next “fresh” electron bunch for additional gain. The repeated broadband FEL gain combined with spectral filtering provided by the Bragg reflectors leads to fully coherent, intense X-ray pulses with meV-scale bandwidth at MHz repetition rate in the 5 – 25 keV energy range. This idea is achievable with current technology when driven by a superconducting, MHz-rate linac such as that under construction for the LCLS-II at SLAC.

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The Workshop began with, Kwang-Je Kim (University of Chicago & ANL) giving an introductory presentation "XFELO Review" in which he described the concept of the XFELO.. Theoretical and experimental efforts conducted in collaborating groups in the U.S., Germany, China and the Russian Federation are leading the way to bring the XFELO to life. The idea of the "ultimate FEL facility" was presented, in which the concept of the XFELO works in tandem with a high-gain XFEL with the XFELO serving either as the seeding source for a high-gain XFEL, or both devices work as separate but complementary facilities. The potential to utilize the X-ray oscillator as a seed to a second-stage, high-gain XFEL in a master oscillator power amplifier or regeneratively amplified FEL configuration was also presented as an avenue toward generating fs-class pulses.

The workshop then began in earnest. The first topic discussed was non-resonant and resonant inelastic x-ray scattering (NR-IXS and RIXS) where methodological improvements for extremely high-resolution non-resonant IXS, high-resolution resonant IXS (HR-RIXS), electronic excitation studies through NR-IXS, and nuclear IXS were considered significant. Applications discussed included studies of deep earth core material evolution, inter-system coupling in, e.g., high-T_c superconductors, and atomic dynamics in disordered materials (e.g.–liquids, glasses). In materials with strong electronic correlations, very high spectral resolution (0.1 meV) would enable the retrieval of detailed information about electron-phonon coupling. One major advantage of an XFELO over current and planned IXS experiments is orders of magnitude higher spectral flux (photons/s/meV). This would allow for improved signal rate while simultaneously reducing radiation damage per scattering event. Since the spectral flux is higher, whereas integrated flux is proportionally lower sample lifetime is increased. Additionally the spectral range of XFELOs extending to ~25 keV while maintaining narrow (meV) resolution is ideal for NR-IXS. This increased spectral range is also critical for RIXS giving access to a broader range of nuclear and electronic resonances.

Next, the workshop focused on coherent diffractive imaging and x-ray photon correlation spectroscopy (XPCS) both methods that make use the coherent flux from the source. The XFELO has clear advantage with an unprecedented degree of coherence in the hard x-ray regime. With higher spatial and temporal resolution there would be the possibility to do ultrafast time-resolved imaging of nanoscale disorder in strongly correlated electron systems (XPCS films with microsecond resolution). The most interesting discussion focused on the potential for Fourier Transform X-ray Spectroscopy as is already done in the optical wavelength range. If realized, this gives direct access to the field autocorrelation function, G1, in the time domain.

The first day concluded with non-linear optics (NLO) in the X-ray regime. Both electronic and nuclear non-linear effects were discussed. Just as the optical lasers revolutionized atomic physics, XFELOs have the potential to revolutionize nuclear physics. For example, a fully inverted nuclear state may be achieved and pump-probe experiments in nuclear physics could be performed. In condensed matter physics, probing the momentum density-density correlation functions via XFELO would also be possible. Nonlinear quantum optics with an XFELO would further allow the study of squeezed and/or entangled states in the X-ray regime. Ghost imaging is proposed as a method for damage-free exploration of atomic scale structure.

While the complete and stable 3D coherence of XFELO pulses are an enabling feature for NLO techniques, it was noted that ultra-fast extensions of the XFELO would add much more to the scientific value. As discussed in Kwang-Je Kim's introduction, the X-ray oscillator could be a seed to a second-stage, high-gain XFEL in a master oscillator power amplifier or used in a regeneratively amplified FEL configuration as an avenue toward generating fs-class pulses.

The second day of the workshop began with the nuclear resonant scattering (NRS) and Mössbauer spectroscopy session. NRS is an element-selective technique that can give information about the partial density of states of a given Mössbauer resonant element. The partial density of states in turn yields information about specific heat, heat conductivity, sound velocities and so on. NRS in the forward direction gives information about hyperfine interactions. Nuclear resonant techniques require very good monochromatization of the source beam, and the excellent spectral flux of an XFELO will open the opportunity to study very subtle properties of matter with enhanced spatial and energy resolution. This includes μeV-resolved spectroscopy for dynamics in mesoscopic and artificially structured materials, pump-probe NRS for non-equlibrium dynamics, NRS ptychography for high resolution spectroscopy of hyperfine interactions, and probing of very narrow nuclear transitions. In biological and medical tissues, XFELO-based NRS can serve as a tool to study the oxidation states and chemical environment to better understand the evolution of diseases.

The next topic discussed was extending frequency combs into the x-ray spectral regime. An XFELO will produce x-ray pulses with a tremendous degree of spatial and temporal coherence. Extension to an x-ray comb additionally requires exceptional temporal stabilization. Approaches to improve temporal coherence from picoseconds to microseconds by phase locking successive output pulses were proposed, using narrow nuclear resonances using, e.g., ^{57}Fe as a reference. By counting the number of standing-wave maxima formed by the nuclear resonance-stabilized XFELO over a known optical length, the wavelength of the resonance can be precisely measured. This technique could be used as a length/time (frequency) standard at x-ray wavelengths. At low energies, laser radiation can be used as a stabilizing tool for the coherence. In this regime, comb output could be used to test the quantum theory for analytically calculable atoms in atomic physics as well as a test for quantum electrodynamics at low energies. Finally, a dual-energy XFELO comb cavity was proposed to enable multi-color phase-coherent spectroscopy.

Required x-ray optics were the topic for the final workshop discussion. Current state of the art components required to build the real cavity were presented. Synthetic, large area, defect-free diamond single crystals were shown exhibiting close to 99% Bragg reflectivity with very narrow bandwidth for the reflected x-rays, as required for the low-loss cavity. Current studies also show diamond to be resilient to the irradiation in the x-ray flux densities expected in an XFELO. Focusing elements are required to maintain beam stabilization within the cavity. Two solutions are being considered: ellipsoidal mirrors (recently developed at Spring-8 with 99% reflectivity and less than 0.1 μrad figure error) as well as beryllium compound refractive lenses which exhibit high transparency to X-rays, and moreover, as tests at APS have shown, are compatible with the expected high power density x-ray flux. Ultra-thin single crystals as out-coupling elements of the cavity were also shown. Technologies to maintain angular stability of the crystals in the cavity at the nanoradian level were also shown to be achievable.

The last day of the workshop was dedicated to final discussions and closing remarks. The overall impression among the participants was that the Workshop was very stimulating. All agreed that the next step would be realized through focused workshops on the topics identified. This will provide the opportunity for the respective communities to focus on the science and requirements for the first XFELO.

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