Research Project Descriptions at Colorado State University, University of California, Berkeley, and Lawrence Berkeley National Laboratory

The 10-week summer program will be centered on engaging students in exciting and authentic research projects. Interns will be assigned specific research projects based on interest, level of prior preparation and challenge, and fit with faculty and graduate mentor research. The projects will allow interns to engage in advanced research that build on concepts they may be familiar with, such as microscopes, interferometers, or Fourier Transformations, and will be designed to familiarize the interns with new concepts in optics, lasers, advanced light sources, and EUV technology. The projects reflect the varied and complementary expertise of the EUV ERC faculty and examples are listed below by core capabilities of partnering research institution. The REU intern's research activities will be aligned with the primary research thrusts of the Center.

Projects at Colorado State University

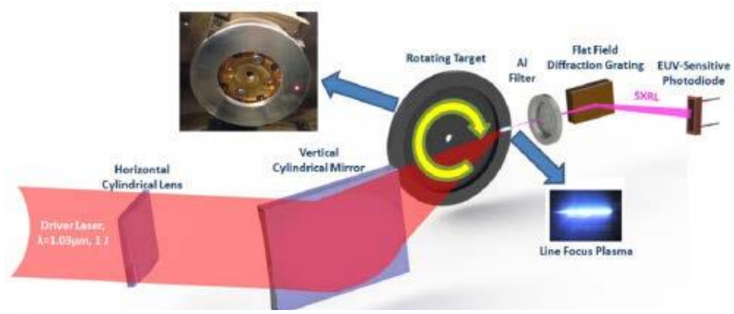

Figure 1: Schematic of table-top EUV laser. An optical laser beam is focused into a selected solid rotating target to generate a line focus plasma from which an EUV laser beam is generated.

<u>Development of EUV lasers to study dynamics in magnetic materials</u> (CSU Prof. Jorge Rocca & Prof. Mario Marconi). We will build a compact diode-pumped/solid state laser that will pump a EUV laser source (Figure 1) to be dedicated to magnetic imaging. This laser will be based on EUV light generation in inverted atomic transitions in a laser-created plasma, a topic in which CSU is a world leader. Strategies to obtain wavelengths in the 20-30 nm spectral region from atomic transitions will be implemented to probe magnetic materials containing Co and Fe. This EUV laser will be used in holographic imaging described below to image magnetic materials in collaboration with Profs. K. Buchanan and M. Marconi. A spatial resolution of ~25 nm and picosecond temporal resolution are expected. The high energy per pulse of the CSU EUV laser source, orders of magnitude higher than what is available at a synchrotron, will enable single-shot dynamic imaging of magnetic processes. An REU student participating in this project will work on the design and implementation of the optical system necessary to create the plasmas in which the EUV light will be amplified. S/He will use programs such as Zemax ®, and subsequently will diagnose the laser-created plasmas learning how to use an EUV spectrometer and to analyze atomic spectra. In a follow-on project, a second REU will work to optimize these plasmas to obtain EUV laser action at different wavelengths of interest, using the same instrumentation and EUV photodiodes. The students will become familiar with EUV laser design and diagnostics.

<u>Nanoscale Resolution Mass Microscope by Extreme Ultraviolet Laser Ablation Mass Spectrometry</u> (CSU Prof. Carmen Menoni). Mass Spectrometry Imaging (MSI) is a powerful analytical tool to assess the chemical composition of solids. Typically, a visible/ultraviolet laser is used to ablate the sample and create ions that when mass-analyzed make it possible to identify molecules and atoms within the probed volume. We have demonstrated a MSI system that uses a focus laser beam of 46.9 nm wavelength, 10x shorter than blue light, in the implementation of a laser ablation mass spectrometry

nanoprobe that can assess chemical composition from sample regions of a few atto-liters volume and with high sensitivity (Figure 2a). We have used this powerful analytical instrument to map composition of a single micro-organism[6] and the isotopic content in micron-size uranium oxide particles (Figure 2b) in collaboration with micro-biologists at CSU, and chemists at Pacific Northwest National Laboratory respectively. We are now focused on improving mass resolution and sensitivity. REU students working on this project will contribute to: develop an automated method to optimize laser ablation using LabVIEW® or MATLAB ®; design and fabricate EUV photodiodes to monitor the laser energy from pulse to pulse; and implement code for data analysis. This is a highly inter-disciplinary project that will expose students to diverse areas of science and engineering, including optics, lasers, materials chemistry, biology, control and image processing.

(a) (b) (c)

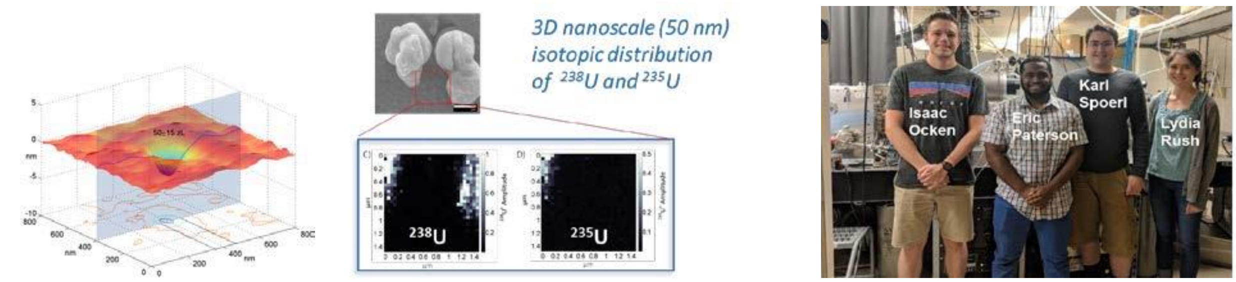

Figure 2: (a) 50 zepto-liter volume crater in resist showing the high resolution possible with EUV MSI. The intact protonated analyte ion is detected from this ablated volume . (b) Nanoscale 3D imaging of isotopic content in Uranium oxide nanoparticles . (c) 2018 REUs Isaac Ocken (CSU), Eric Paterson (Morehouse College) and Karl Spoerl (CSU) work with mentor graduate student Lydia Rush. Their projects include data acquisition and analysis, and design of a new chamber for the instrument to couple to a multi-collector magnetic sector instrument to improve sensitivity and mass resolution.

<u>Design, Fabrication and Diagnostics of interference coatings for EUV laser drivers</u> (CSU Prof. Carmen Menoni). This project relates to the growth and characterization of interference coatings for near infrared ultra-high intensity lasers that are used as drivers to pump EUV lasers. These advanced thin film structures consists of thin layers of transparent amorphous oxides that are deposited by ion beam sputtering. The REU student will participate in using optical metrology to characterize amorphous thin for their absorption loss at near infrared wavelengths and for their stress. The REU student will also be involved in design and characterization of multilayer coatings that are used in the laser amplifiers.

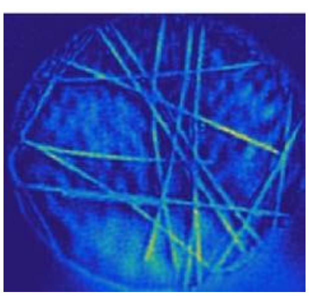

Figure 3: Single shot FTH image of 200 nm Ag nanowires obtained with 13.9nm EUV laser illumination. The spatial resolution is 2 nm and the temporal resolution is 5 ps.

<u>Nanoscale Resolution Extreme Ultraviolet Fourier Holography</u> (CSU Prof. Mario Marconi). Recent advances in nanotechnology and nanoscience have created a need for new compact imaging systems capable to resolve nanometer scale features. Optical imaging systems are unsurpassed in versatility, although they are limited in their ability to resolve objects at the nanoscale by the wavelength of the illumination. Using EUV lasers illumination we have developed a microscope based on Fourier Transform Holography (FTH) which can capture single shot holograms that are processed in real time to produce three dimensional images of nanostructures (Figure 3). In FTH, the hologram is formed by the interference of a spherical wave front reference beam and the object beam. Their interference is recorded on an array detector and the images are obtained by a straightforward reconstruction algorithm that renders almost “live” images of the object, with the

additional advantages that a large field of view and nanoscale spatial resolution are simultaneously attainable. This is how the image of 200 nm Ag nanowires of Figure 3 was obtained. We plan to use FTH for the imaging of magnetic materials in collaboration with Prof. K. Buchanan. REU students engaged in this project will contribute to develop hardware to adapt the FTH imaging system to achieve improved spatial resolution, will collect and analyze data, and will contribute to optimize the reconstruction code. This is a highly inter-disciplinary project that will expose students to diverse areas of science and engineering, including optics, lasers, nanomaterials, and image processing.

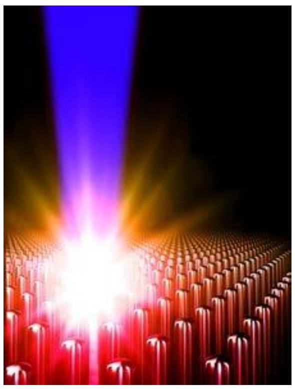

Figure 4. The interaction of femtosecond laser pulses of relativistic intensity with arrays of aligned nanostructures offers an opportunity to create extraordinarily bright, compact x-ray sources.

<u>Generation of intense ultrafast x-ray flashes from nanostructures irradiated with ultra-intense laser pulses</u> (CSU Prof. Jorge Rocca). The interaction of femtosecond laser pulses of relativistic intensity with arrays of aligned nanostructures offers an opportunity to create extraordinarily bright, compact x-ray sources. The REU student will have the opportunity to work growing arrays of nanowires and characterizing them by electron microscopy. The REU student will participate in experiment in which these nanostructures are irradiated with one of the world's most powerful lasers. A recent experiment has shown converting a record ~ 20% of the laser energy into > 1keV photons. Previously, the conversion efficiency of optical laser light into picosecond x-ray pulses was limited to less than 1%, owing to the rapid hydrodynamic expansion of the thin, hot plasmas created near the surface. The measured increase in x-ray yield is made possible by volumetrically heating arrays of vertically aligned gold nanowires with relativistic laser pulses (5×1019 W cm-2)**.** The laser energy is deposited deep into the nanowire array, where is practically totally absorbed to form a hot, near solid density plasma several micron in depth. This leads to a condition where the plasma energy is dissipated primarily by x-ray radiation greatly increasing the x-ray flux.

Projects at University of California, Berkeley

<u>Transient Absorption in Solids Utilizing High Harmonic EUV Light</u> (UCB Prof. Stephen Leone). REU students will participate in transient absorption and transient reflectivity experiments utilizing high harmonic and attosecond EUV radiation. A project that is well suited to the summer REU program involves the measurement of dynamics in metal oxide materials or metal dichalcogenides to observe charge state dynamics. In MoTe2, for example, holes and electrons are observed after excitation across the band gap. In addition, coherent phonon motion is readily detected, and carrier cooling is measured. In Fe2O3 hematite, the initial photoexcitation with visible light results in the transfer of charge from an oxygen atom to an iron atom, changing the oxidation state of the iron from 3+ to 2+. This change is then accompanied by the formation of a polaron, where the vibrational motion of the lattice (phonons) adjusts to trap a charge on the iron atom. By probing these dynamics in materials that are tailored to alter the propensity for polaron formation, it is possible to characterize materials that have better or worse properties for charge separation (the formation of polarons reduces the possibility for charge separation.) New measurements are being initiated with four wave mixing of EUV light in solids. In these experiments, students will learn about charge carrier dynamics in materials that can provide for solar utilization and energy conversion from sunlight to electricity. In addition, students will learn modeling of EUV spectra to interpret their meaning, effects such as hot carrier relaxation and

polaron formation, as well as deposition of energy into phonon modes. Concepts of electronic charge migration and recombination are also acquired.

Molecular Photophysics at the Carbon K Edge[5] (UCB Prof. Stephen Leone). The photochemistry of organic molecules is a highly investigated field of study, in which processes such as ring opening, singlet-to-triplet transfer, and radical formation are ubiquitous. In this platform, small molecules are excited with ultraviolet pulses and 300 eV photons are used to probe the changes in orbital structure around carbon atoms in the molecule. An example is ring opening (Figure 5), in which is found that ring shaped molecules containing heteroatoms such as oxygen have dramatically different spectra at the carbon K edge when one atom of carbon becomes free from its initial bond to the heteroatom. The timescales for the dynamics are directly obtained in the ultrafast femtosecond time domain. Students will learn about EUV spectroscopy, bond breaking, simulation of spectra, differential absorption, and molecular photophysics. Combining data analysis with simulation and global fitting provide important mathematical concepts for future careers.

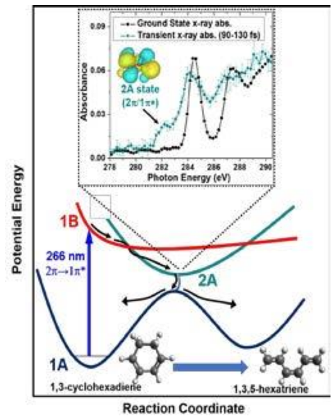

Figure 5: Direct fs identification of the transition state 2A, arrow, for cyclohexadiene ring opening.

Projects at Lawrence Berkeley National Laboratory

Ultrafast x-ray dynamics in quantum materials (LBNL Michael Zuerch). Quantum materials often host exotic, emergent properties as a result of strong correlations between electrons, for which superconductivity, magnetism, and Mott insulators provide some representative examples. Given the complex couplings between multiple degrees of freedom, so-called strongly-correlated materials often exist in a complicated phase space with multiple competing ground states. Ultrashort light pulses are the perfect tool to both probe the nature of emergent phenomena and control its properties on the natural spatiotemporal scales of charge, spin, and lattice motion. In the Zuerch lab, we use pump-probe methods to study quantum materials, with a particular interest towards mesoscale ordering phenomena and phase transitions. We are currently building a state-of-the-art beamline to perform attosecond time-resolved x-ray absorption spectroscopy and resonant x-ray diffraction in a single measurement to probe materials on ultrafast timescales with nanoscale resolution and element specificity (Fig. 6). With carrier envelope phase (CEP)-stabilized pump pulses, full intensity, and polarization control, the ultrafast dynamics that ensue following a sub-cycle optical perturbation can be followed with great detail. In the summer of 2021, we will be performing experiments on multiferroic materials that exhibit coupled ferroelectric and ferromagnetic ordering and looking towards upgrading our beamline towards higher photon energies, generating circularly polarized x-rays to study chiral systems, and implementing a THz light source to excite low-energy excitations. An NSF REU student will assist in these endeavors, learning about quantum materials, ultrafast dynamics, and x-ray attoscience in the process.

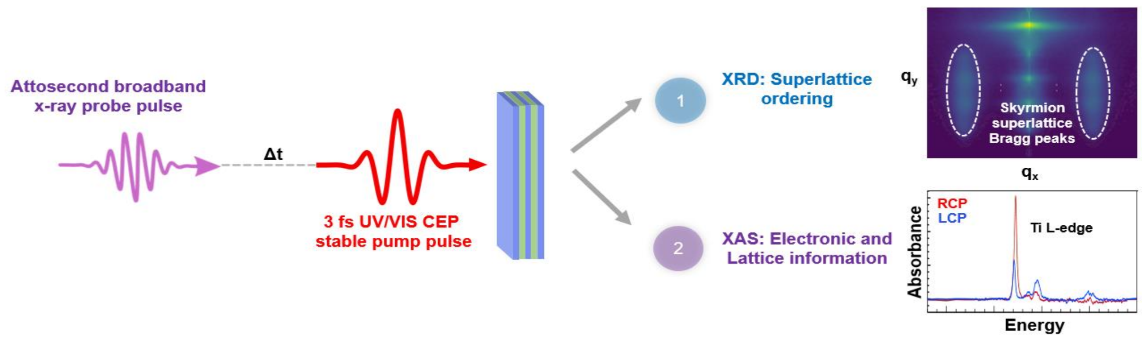

Fig 6. A few-fs UV/VIS CEP-stable pump pulse followed by an attosecond, broadband x-ray probe pulse after a variable time delay is incident on heterostructure with superlattice ordering. The diffracted beam (1) is scattered onto a camera, where the resulting Bragg peaks provide information about the superlattice periodicity. The transmitted beam (2) is simultaneously captured, yielding transient absorption spectra that report on electronic and lattice dynamics. Static diffraction and absorption spectra from ref. [1].

[1] S. Das, et al., Nature **568**, 368 (2019).