Using X-ray Free Electron Lasers in High Pressure Physics

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Abstract: The study of how shock waves travel through matter and the effects they induce are not well understood areas of physical sciences and material studies. Standard techniques typically used for other fronts of research are starting to be used to probe this area. X-ray free electron lasers (XFELs) have typically been used to probe biological and chemical microscopic processes on the time scale order of femtoseconds. While these lasers provide fast pulses and short wavelength probing mechanisms, they also deliver a large amount of energy that when confined can be used to send shockwaves through a material and study the effects of that shock wave. While high pressure physics is not a new area of research, innovations in the field continue to make it exciting and useful. An x-ray source has yet to be used to induce high pressures, and this study aims to do just that. By using the x-ray free electron laser at the Stanford Linear Accelerator under beamtime LP70, many targets with well-characterized phase transitions are studied to analyze the effectiveness of x-rays in creating high pressure. From meteor impact studies to quasicrystal formation, this study address important questions in fundamental physics research but also has important applications. This paper discusses the methods of preparation for the beamtime and the reasoning behind sample choice. Further papers will discus results of these studies.

I. INTRODUCTION

The study of high pressure physics has led to novel discoveries and important applications within many disciplines. It has made a large contribution to the understanding of earth and planetary sciences. The applications in this geophysical realm range from accurately predicting the speed of sound through the earth to understanding the formation of planets. Without knowing the equilibrium equation of state or how phase transitions occur, seismography interpretations would be inaccurate. In planet formation, high pressure studies have allowed astrophysicists to probe the interior atoms of gas giants and understand the He and H mixture. The novel discoveries have contributed to fundamental physics. One of the most useful discoveries has been how the electronic interactions within a material are affected by the level of pressure the material is under. When a material is not under significant pressure, its electrons will be localized, but when subjected to high pressure, the electrons can delocalize, which may change the electronic properties of the material. This explains how materials can lose or gain superconductivity or magnetism and even transition between metals and insulators. This also has applications to fusion energy.

Since high pressure studies yield significant results, it is important to understand how to reach high enough pressures experimentally to see these results. High pressure results can be reached through either static or dynamic methods. The opposed diamond anvil cell [1] is the most well known of the static methods. It is effective in that it not only reaches high pressures, but using diamond to create the high pressures allows the use of various microscopies though the diamond and if a beryllium (Be) washer is used, x-rays can also be used to characterize the sample and learn more about the effects of high pressure. Dynamically, a gas gun or electromagnetic gun may be used to shoot a target and induce shock waves within the target. Another dynamic method involves placing the sample between two pieces of metal and shining pulses of ultraviolet light onto the metal, allowing the outermost edges of the metal to ionize. As the electrons leave the metal in the outward direction, there is a recoil that sends a shock inward toward the sample, increasing the amount of pressure it is under. The current study uses an x-ray free electron laser (XFEL) to do a peening study on a variety of samples. This is analogous to the method of laser peening. Consider an aluminum (Al) sheet with a thin layer of paint on its surface. Exposing the painted surface to a laser pulse causes the painted layer to ablate with a small change in momentum and no energy transfer. However, if a confining layer of water is placed on top of the painted surface, a completely different result is achieved. This confinement acts similar to tamping in muzzleloaders so that when the laser pulse is shot at the sheet, it transfers energy to the sheet of Al and work hardens the surface. This has had an important application in the production of airplane fuselages. The excess strain in the metal from being bent into shape can be combated by work hardening the metal in this way. Using the XFEL for the peening study is important because while there are many techniques to induce high pressure in samples, an x-ray source has never been used to do so. The goal of the study was to demonstrate that XFELs could be used to induce high pressure situations. This was achieved by using the XFEL from the Linac Coher-

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ent Light Source (LCLS) at the Stanford Linear Accelerator (SLAC) under beamtime LP70 with a variety of targets ideal for studying because of well-known phase transitions and metal alloying.

II. BACKGROUND

A. Previous APS Studies

The Seidler group does ongoing research at the Advanced Photon Source (APS) at Argonne National Laboratory. The group uses the Biology Consortium for Advanced Radiation Sources (BioCARS) sector 14-1D of the synchrotron x-ray source to shoot a variety of targets to learn about sample design and x-ray induced effects on the samples. This beam gives single pulse x-rays with photon energy around 12 keV with an energy band width of approximately 500 eV. While the power of this beam is less than that at LCLS, two important concepts were established during these studies. Both of these concepts are discussed in previous publications by the Seidler group but are discussed here briefly for convenience.

1. *Hot Electron Furnace*

The concept of a hot electron furnace in Hoidn et. al. [4] has proven to be useful in determining sample design and has been verified by the Sediler group in previous APS studies. Studying the effect of x-rays on low-Z elements can be challenging because the attenuation length of x-rays in those elements is long, so the chance of the x-ray being absorbed is small. However, the attenuation length of x-rays in high-Z elements is much shorter, making it much more likely that the x-ray would be absorbed by them. Making use of this knowledge, a target can be constructed in such a way that the energy from the x-rays can be deposited into a low-Z element. By putting a high-Z element in front of the low-Z element, the x-ray has a high probability of being absorbed by the high-Z element. The excitation from the x-ray will cause hot electrons to travel from the high-Z element to the low-Z element, depositing energy into the low-Z element. This creates a hot electron furnace containing the low-Z element. The effectiveness of this type of sample design is demonstrated in **Figure 1**. These results came from an APS study involving varying thicknesses of carbon samples with 50 nm of Au on the front and back and one control sample.

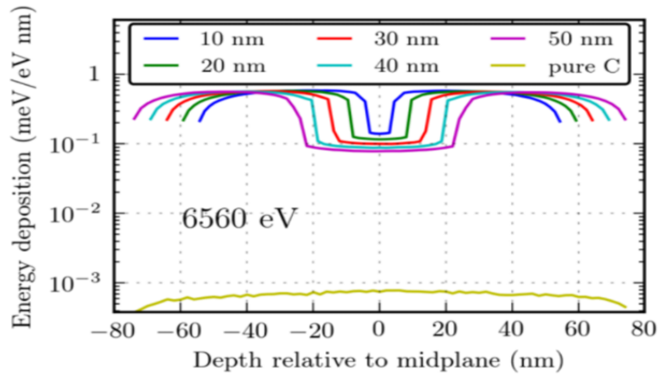

Figure 1: Hot Electron Furnace This plot shows the importance of using high-Z elements to ensure x-ray energy will be absorbed to deposit energy into low-Z elements. The C samples with varying thicknesses all were coated on both sides with 50 nm of Au and clearly have a much higher energy deposition level than the pure C.

2. *Proof of Principle: Inertial Confinement*

Beyond the hot electron furnace, the other important principle that was proved at APS was the importance of confinement as in laser peening. Valenza et. al. [5] discusses how this was demonstrated at APS. A commercially available sample of Cu-Au-Cu with each layer having .5 μm thickness was hit using the APS source and a hole was blown into it. However, when 100 μm of polymer was painted onto both sides of the tri-layer, the sample did not explode, but instead could be repeatedly hit with the x-rays to achieve an annealed effect. This principle of confinement was used to prevent explosions with the release of pressure and allows pressures to build within the sample.

B. XFELs and Shock Physics

An XFEL consists of a linear accelerator followed by an undulator. UV light hits a copper cathode which then emits electrons that travel through the linear accelerator to an undulator whose alternating north-south magnets cause the electron to oscillate giving off x-rays that give rise to the pulse in the XFEL. XFELs have predominantly been used to probe biological and chemical processes that occur on short time scales. In this sense, the x-ray source is used as a probe to understand processes. The Seidler group took the approach of using the XFEL as the mechanism that actually induces the change to the sample with analysis and probing of the samples done later with different techniques.
In order to demonstrate the pressure reached using the XFEL, samples with well characterized phase transitions were used. Meteor impacts are events that

involve high pressures and have been studied to the exteent that the pressures associated with some of the seen phase transitions are well known. The transition of quartz from coesite to stishovite and the transition of carbon and graphite to diamond have been found at meteor impacts and the pressure reached in each of these transistions is generally undisputed. Al plates were also prepared to demonstrate work hardening as mentioned earlier in relation to laser peening. The Cu-Au-Cu sample was studied again to compare results to the previous APS studies. The Al-Cu-Fe sample was chosen in hopes of creating the corressponding icosahedral i-Al-Cu-Fe quasicrystal based on Grenet, et. al. [2] **Table 1** lists the broad categories of prepared samples. The details of their preparation are discussed in the following section.

III. EXPERIMENTAL

A. Sample Preparation

With many samples to prepare, various methods of sample preparation were tried. Each sample type was prepared in a unique way to complement the goal transition of each study.

Alloy Samples	Meteor Impact	Work Hardening	Nanoparticles
Cu-Au-Cu	Quartzite	Al	Epoxy Cast TiO_2
Al-Cu-Fe	Glassy Carbon		ZrO_2 in PMMA
	Highly Oriented Pyrolytic Graphite		
	Pyrolytic BN		

Table 1: LP70 Samples This table shows the broad list of samples prepared and targeted during the beamtime. Each study contributes to the overall goal of demonstrating high pressure.

1. *Meteor Impact and Work Hardening*

The samples related to work hardening and meteor impact phase transitions were prepared using identical methodology. The glassy carbon, highly oriented pyrolytic graphite (HOPG) and pyrolytic BN (pBN) were 2 mm thick and commercially available from SPI Supplies. The quartzite was prepared by a collaborator as a thin plate. The Al plates were machined in the University of Washington (UW) machine shop. Because all of these substrates are composed of low-Z elements, it was important to make use of the hot electron furnace concept in preparing them. For this reason, 1 μm of Au was evaporated onto each of these substrates using the UW electron beam evaporator. Because these samples required 1 μm of Au and typical evaporation lengths are on the scale of nm, an attachment to the sample holder was machined. This attachment allowed the samples to be held closer to the Au crucible which led to a 3x gain in the thickness of Au evaporated. With this gain from the samples being closer to the crucible, less Au had to be evaporated so that money and time spent on the evaporation were saved. With less Au being evaporated, there was also less strain put on the sensors so that their lifetime was only minimally affected. The attachment was a rectangular $\frac{1}{8}$ in. thick Al sheet with $\frac{1}{4}$-20 holes drilled to allow screws to hold the samples in place. To ensure that the Au stuck to each sample, a 6-8 nm 'sticking layer' of chromium (Cr) was deposited onto the sample before the Au. While this Au is important for reaching high energy deposition levels, it does make post facto analysis more challenging by creating a large amount of background when using techniques like x-ray diffraction and a variety of microscopies. In order to be able to easily remove the Au from delicate samples like the quartzite, a thin layer of photo resist was spin coated onto the sample before evaporating Au onto the sample. This simplifies Au removal because photo resist can easily be wiped off using acetone. Therefore, washing the sample with acetone will remove the photo resist and thus the Au on top of it, significantly simplifying the analysis process. All shot quartzite and Al samples were coated with photo resist, while the glassy carbon, HOPG and pBN samples were all prepared both with and without photo resist so there would be control samples to determine if the photo resist was having an effect on target geometry outside of making the task of removing Au simpler. These samples were also confined and left without confinement to demonstrate the importance of confinement once again.

2. *Alloy Samples*

The Cu-Au-Cu sample was a tri-layer with each layer being 0.5 μm thick identical to the sample used at APS. This thin sample was inertially confined on both the front and back of the sample. The Au-Cu-Fe sample was prepared using e-beam evaporation onto 1 mil Kapton with the overall composition of the sample being $Al_{62.5}Cu_{25}Fe_{12.5}$ as that corresponds to the desired quasicrystal. Kapton was useful to deposit onto because it does not succumb to radiation damage. As in Grenet, et. al.[2], multiple "sandwiches" of these layers were made in order to give the sample multiple interfaces for the quasicrystals to form at. The order of the deposition was Al-Cu-Fe-Cu-Al-Cu-Fe-Al so that there were four regions of Al-Cu-Fe. It was important to keep the order so that for example Fe was never deposited directly onto Al. As with the Au deposition, a small Cr sticking layer was initially used. Since these thicknesses were on the nm scale, the sample holding attachment was not used so the samples were the typical distance from the crucible. The final Al layer was slightly thicker than the ratio required as to act as a buffer against oxidation. The quasicrystal sample was also stored under vacuum in an effort to minimize oxidation effects. Polymer was painted onto the front of the sample to inertially confine it.

3. Nanoparticles

The nanoparticle samples did not require an e-beam deposition. The commercially available TiO_2 nanoparticles were epoxy cast onto kapton. The ZrO_2 nanoparticles, which were also commercially available, were in PMMA on the same kapton. These samples were prepared in a variety of concentrations. Preliminary x-ray diffraction (XRD) was done on the samples. Comparing the XRD results of the various concentrated samples revealed which concentration showed strong enough Bragg peaks that the nanoparticle signature could be picked up from bulk XRD.

B. LCLS LP70 Beamtime

The current study was carried out using the SLAC LCLS XFEL under the LP70 beamtme at the Matter in Extreme Conditions (MEC) endstation. This endstation has been designated to shock physics among other disciplines and its main feature entails combining high power optical lasers with the XFEL. The endstation contains a large vacuum chamber where the samples were placed to be exposed to the XFEL.

1. Sample Mounting

With time being of high value, sample mounting geometry was vital to ensure time was used efficiently. To make the best use of time, aluminum plates that could hold from three to six samples were designed by the Seidler group and machined by the UW machine shop instrument makers. Multiple of these aluminum plates were screwed into the sample holder at a time so that vacuum would have to be broken as little as possible to exchange samples. To attach the samples to these Al plates, nail polish was used for the alloy samples, the nanoparticles and the quartzite. All other samples were attached using kapton tape. Black nail polish was used to place small symmetry breaking fiducials on the samples so that when shooting the sample with x-rays, the group was able to orient itself. It was also important to include the fiducials so that when the samples were removed from the plates for analysis, sample parts were note confused.

2. XFEL Exposure

While using the XFEL, the spot size was 25 μm in diameter. A YAG crystal was used to calibrate the equipment. There were two different ways in which the XFEL was used to shoot the sample. The first was referred to as 'grid exposure'. With this type of exposure, multiple shots were done at the exact same spot. The sample would then be moved to a new spot 250 μm to the right of the first and multiple shots would be one at that spot. A row of these shots would be done at one intensity and then a new row of shots would be started and the intensity was brought down an order of magnitude. This was repeated for multiple intensities. The second type of exposure was referred to as 'area exposure'. In this case, a single shot was fired at a spot and then the sample was moved slightly and another single shot was fired that had a slight overlap with the first shot. This process continued rastering the XFEL back and forth across multiple rows at the same intensity. After an area exposure was completed, the XFEL was attenuated down an order of magnitude in intensity and another area exposure was done at the lower intensity. The XFEL itself was also used to make fiducials to indicate where each intensity of shot was on each sample.

C. Analysis

These samples were designed for post facto analysis, which took place at the UW Molecular Analysis Facility (MAF). Bulk XRD was done on each of the samples as an initial diagnostic of post facto analysis. Each sample then will require further analysis unique to that sample. Considering these preliminary results, various other forms of microscopy were used. Scanning electron microscopy (SEM) was used for a qualitative look at what happened to the sample. Taking a look at the physical changes to the samples helped to indicate which portions of the samples should be looked at using other techniques. Raman spectroscopy followed SEM and gave a quantiative look at the changes to the sample. Findings from this analysis will be detailed in future Seidler publications.

IV. FUTURE WORK

The preliminary results of UW MAF analysis indicate that the immediate future will be an exciting time in the Seidler group. The most immediate form of planned analysis is electron backscatter diffraction. The samples will require a significant amount of further analysis and beyond that the Seidler group aslo looks forward to another upcoming beamtime at APS.

On the LCLS front, construction was started on a second generation source called LCLS-II in April of

2016. The upgrade will involve putting another accelerator and undulating magnet next to the existing one. The new accelerator will be superconducting. The expected output is that there will be 1 million pulses per second instead of 120 and that the x-rays will be 10,000 times as bright. [3]

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