Final Scientific Report: DE-SC0008580

Term of award: 8/15/2012 – 8/14/2016
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The term of this grant, from 8/15/2012 – 8/14/2016, saw several notable scientific, technical, and organizational accomplishments. Below, we address these by the indicated category headings.

I Publications

Ten papers have appeared in print, and three additional manuscripts (discussed below) are in final stages of preparation. These will be submitted outside the support window but will acknowledge support from the present award. Papers in print are:

1. E. Klevak, F.D. Vila, J. Kas, J.J. Rehr, and G.T. Seidler, “Finite temperature calculations of the Compton profile of Be, Li, and Si,” Phys. Rev. B **94**, 214201 (2016).
2. G.T. Seidler, D.R. Mortensen, A. Ditter, N. Ball, and A. Remesnik, “A Modern Laboratory XAFS Cookbook,” Journal of Physics: Conference Series **712,** 012015 (2016).
3. D.R. Mortensen, G.T. Seidler, A.S. Ditter, P. Glatzel, “Benchtop Nonresonant X-ray Emission Spectroscopy: Coming Soon to Laboratories and Beamlines Near You,” Journal of Physics: Conference Series **712,** 012036 (2016).
4. R.A. Valenza and G.T. Seidler, “Warm Dense Crystallography,” Physical Review B **93**, 115135 (2016).
5. M.J. Lipp, J.R. Jeffries, J.-H. Park Klepies, W.J. Evans, D.R. Mortensen, G.T. Seidler, Y. Xiao, and P. Chow, “4f-electron behavior in the cerium oxides Ce_2O_3 and CeO_2 under pressure,” Phys. Rev. B **93**, 064106 (2015).
6. Oliver R. Hoidn and Gerald T. Seidler, “A Tender X-ray Camera Based on Mass Produced CMOS Sensors and Single-Board Computers,” Review of Scientific Instruments **86**, 086107 (2015).
7. G.T. Seidler, D.R. Mortensen, A.J. Remesnik, J.I. Pacold, N.A. Ball, N. Barry, M. Styczinski, O.R. Hoidn, “A Laboratory-based Hard X-ray Monochromator for High-Resolution X-ray Emission Spectroscopy and X-ray Absorption Near Edge Structure Measurements,” Review of Scientific Instruments **85**, 113906 (2014).
8. L. B. Fletcher, A. L. Kritcher, A. Pak, T. Ma, T. Döppner, C. Fortmann, L. Divol, O. S. Jones, O. L. Landen, H. A. Scott, J. Vorberger, D. A. Chapman, D. O. Gericke, B. A. Mattern, G. T. Seidler, G. Gregori, R. W. Falcone, and S. H. Glenzer, "Observation of Continuum Depression in Warm Dense Matter with X-ray Thomson Scattering," Phys. Rev. Lett. **112**, 145005 (2014).
9. Oliver R. Hoidn and Gerald T. Seidler, “Photometric Study of Single-Shot Energy-Dispersive X-ray Diffraction at a Laser Plasma Facility,” Physics of Plasmas **21**, 012707 (2013).

10. Brian A. Mattern and Gerald T. Seidler, “Theoretical Treatments of the Bound-Free Contribution and Experimental Best Practice in X-ray Thomson Scattering from Warm Dense Matter,” Physics of Plasmas **20**, 022706 (2013) (doi: 10.1063/1.4790659).

II Beamtime at LCLS/MEC and also at APS

Under funding from this award, the PI led two campaigns at the MEC endstation, LD67 and LK20, and also two beamruns at the APS aimed at prototyping x-ray heating target design issues..

II.A. MEC: LD67

The goal of the LD67 beamtime was to assess whether the problem of continuum lowering could be interrogated in detail by comparing its simultaneous effects on different elements in transition-metal alloys. Several complete data sets were collected using x-ray only configuration (no laser pulse), but technical difficulties with the target design and the irregularity of laser field illumination inhibited the laser-pump/ x-ray-probe portion of the beamtime. That being said, there were several notable outcomes.

The highlight example involve the numerous results obtained on x-ray heating of nanophase materials led to several new research directions. First, the observation of persistent crystallinity even at extremely elevated electronic temperature inspired the theoretical work, Valenza and Seidler (2016) [cite 4, above]. A central result of that paper is the prediction that, for low-Z materials, thermally excited electrons cannot be considered plane wave-like, in which case they would have zero contribution to x-ray diffraction. Instead, they are still in Block wave states, which are not free-momentum eigenstates, and consequently x-ray diffraction is still significantly sensitive to their thermal occupation of energy bands defined by the crystalline structure. This has the important consequence that diffraction, via changes in ratios of intensities in Bragg peaks, is therefore sensitive in significant detail to the energy distribution of electrons. This gives high potential for, e.g., testing finite-T DFT models in the very difficult transitional region $T \sim E_F$ where it is unclear how to best address finite-T exchange functions. These theoretical ideas, together with supporting experimental results from LD67, helped motivate the LK20 beamrun.

Second, as shown in the Fig 1 (Ditter, in prep, finally near submission), we also observed an interested effect relating nanoscale target morphology to the effectiveness of energy deposition by x-ray heating. In Fig. 1, we show Kb x-ray emission spectra from nanophase and microphase Fe3O4 as a function of beam energy density incident on the target. The key observations are that at equivalent incident energy density, the nanophase target exhibits less electronic heating. We have subsequently performed a broad family of simulations of energy deposition into nanophase target designs (Hoidn, et al, very near submission), and have quantified predictions of this effect and recommendations for best practice to obtain highest electronic heating for given beam conditions. We believe that our work will have lasting impact on the design of x-ray heating targets for XFEL studies.

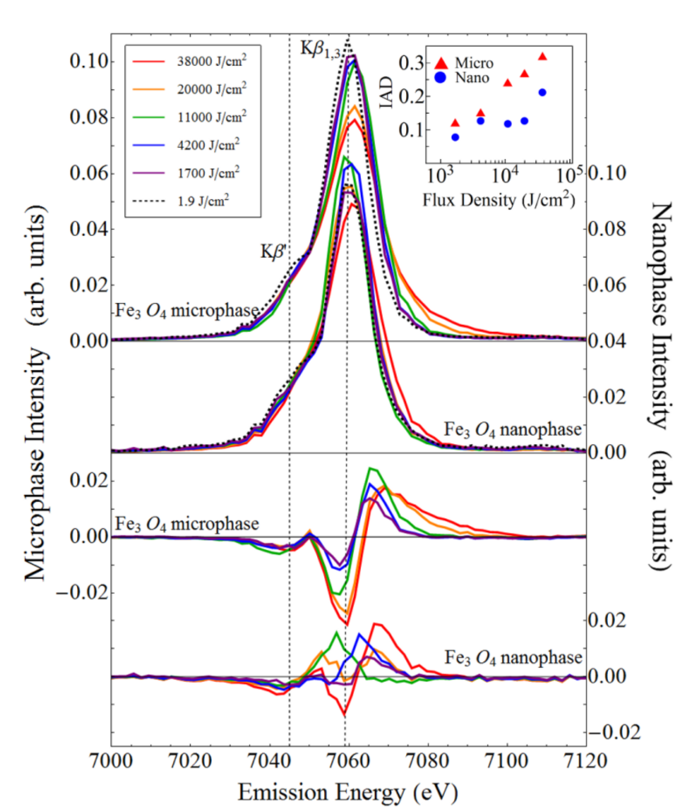

Figure 1: (Ditter et al, in prep) The Fe Kβ x-ray emission spectra from Fe3O4 as a function of incident energy density. The electronic heating effects are much stronger in the micron-scale sample than the nanoscale sample because the range of hot electron transport, ~50-100 nm, is sufficient to let much of the absorbed energy be deposited in the low-Z binder outside the Fe3O4 nanoparticles.

Third, the use of nanoparticles brings another issue to play: the possible expansion of the nanoparticles during ~20 fs time scale, which is modestly allowed by elementary speed-of-sound considerations. The relative absence of such a signal in our data, caused us to question the role of the surrounding polymer binder as an inertial tamper, in analogy to similar effects at much larger length scales for long-pulse laser shock experiments. In recent beamtime at the Advanced Photon Source (APS), we were able to demonstrate that such inertial coatings can be highly effective at containing highly heated matter – we are, in effect, performing "x-ray peening" studies, in analogy to the well-known process of "laser peening". The APS results (discussed in II.C) unambiguously demonstrated lattice temperatures above 0.1 eV (1000 K) via changes in Bragg peak intensities of a Cu-Au-Cu multilayer sample, giving clear evidence for interdiffusion and alloying of the metal layers.(Valenza, Hoidn, et al, in prep) This work strongly motivated the upcoming LP70 run at MEC, where we will use the full power of the LCLS to investigate the temperatures and *pressures* that can be achieved in x-ray peening. The planned studies will result in the capture of the illuminated sample regions, allowing the investigation of bulk-like amounts of several different material families that will have been x-ray heated to temperatures above 10,000K (10 eV) and then quenched at rates of ~10^{10} K/ s while likely experiencing reaction-shock pressures in the 100 GPa range. <u>We believe that this effort, spanning the completed grant and the new NSF/ DOE award, will demonstrate a new paradigm for creating and studying warm dense matter</u>.

II.B. Beamtime LK20 at MEC/LCLS

The purpose of the LK20 experiment was to demonstrate a clear experimental modality for 2-color x-ray diffraction over a wide momentum-transfer range, while at the same time investigating nanoscale effects in x-ray heating target design. Unfortunately, the 2-color operational mode of the accelerator could not be stabilized in the later shifts, and the 2-color data taken in the early time window of the study was somewhat corrupted by an error in the local endstation software. That being said, the single-pulse x-ray heating studies in the last shift of the beamtime have proven fruitful. As one example that is serving as the capstone project for a graduate student supported under this award (Hoidn, et al., in prep), the x-ray heating of nanophase MgO is showing clear evidence for maintained crystallinity (due to confinement) and also for simple electronic reorganization as a thermalized consequence of simple x-ray heating to electronic temperatures up to ~20 eV. This beamtime also provided additional x-ray diffraction data that has been included in the Fe_3O_4 manuscript in preparation from the LD67 beamrun.

II.C. X-ray Heating Study at the Advanced Photon Source

When using a focused insertion device beam in ‘pink beam’ mode, single pulses at the APS can achieve energy densities less than 0.1% that of the LCLS, but still sufficient for studying some “nearly” WDM conditions and for proof of principle of some target design issues. The pulse duration of ~100ps is of course much longer than at the LCLS, so that this type of heating study is heavily thermalized with the lattice during x-ray exposure. Last summer, under support from this award, we completed the second

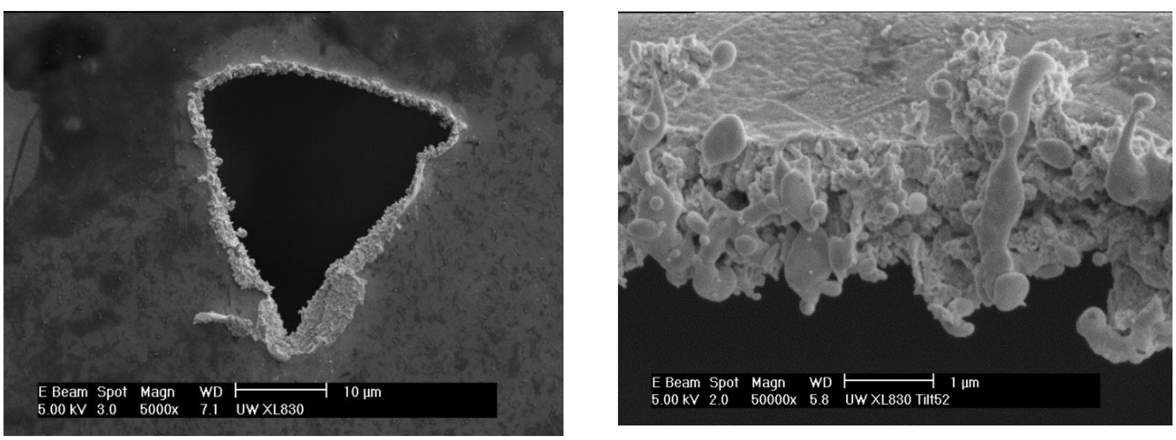

Figure 2:
Left top panel: SEM micrograph of a 2-μm thick gold foil after exposure to a single 90 μJ pulse with 25 μm FWHM at ~12 keV at the Advanced Photon Source.
Left bottom panel: SEM micrograph at higher magnification of the edge of the hole in the Au foil target from the left-top image. Note the globular morphology, indicating melting and a rapid quench. Note that the melting point of Au is ~1300 K while the boiling point is ~3000 K.

iteration of a study to heat high-Z metals with such a focused beam at the BIOCARS beamline. Similar capability is now available at the Dynamic Compression and Shock beamline, also. As shown in Fig. 2, we had clear evidence for heating to temperatures well above the melting point of Au (1064 C).

This led to a study of confined heating in a Cu-Au-Cu multilayer. As shown in Fig. 3, the inertial confinement with a ~0.1mm thick polymer layer allowed multiple x-ray shots to illuminate the same sample spot, with thermal quenching after each shot. The changing structure and intensity of the x-ray diffraction is simply explained as a consequence of alloying of the Cu and Au over a ~100nm length scale after 10 pulses. The sample was also observed to exhibit a clear mechanical recoil to the x-ray pulses, again suggesting the generation of a reaction shock, as in laser peening. Back-of-the-envelope calculations suggest pressures above 50 GPa in the APS low-power experiment, and the possibility of much more intense reaction shock waves in the upcoming LCLS work. A manuscript is in preparation (Valenza, Hoidn, et al).

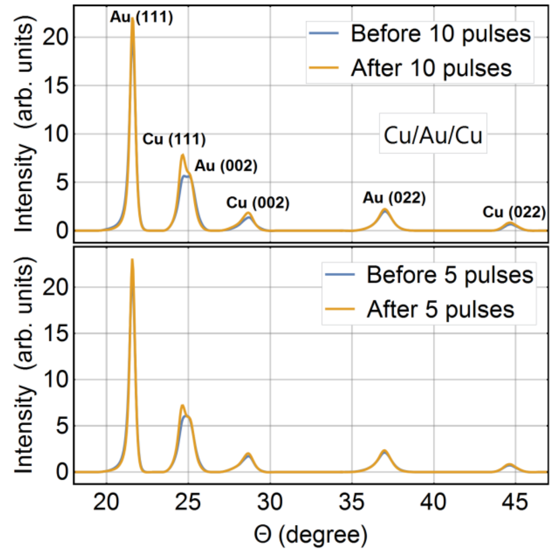

Fig. 2: XRD results from APS taken before and after 5 pulses (bottom) or 100 pulses (top) of 7 J/cm^2 on a target consisting of a (0.5-μm Cu)/(0.5-μm Au)/(0.5-μm Cu) trilayer coated on both faces with ~100 μm of polymer cladding.

III. Theoretical Work and Connection with Laser Plasma Physics

Three of the publications under this award address, or make use of, improved theoretical treatment of x-ray Thomson scattering. The most recent paper (Klevak, et al) investigates the role of thermal lattice disorder on the Compton scattering profile. That study makes use of finite-T DFT applied to MD-simulated Be, Li and Si to isolate the relative importance of electronic-thermal effects and ion-core (melting/ disorder) thermal effects on Compton scattering in the 'tepid' WDM regime of a few eV temperature. This result will be useful for constraining the use of crystalline models, where finite-T calculations are simpler, as testing grounds for predictions for disordered systems, such as result from long-pulse laser studies.

An additional paper (Mattern and Seidler 2013) which appeared early in this award has had significant effect in the field, correcting a long-standing faulty practice in the modeling and fitting of the core-shell contributions to x-ray Thomson scattering.

Finally, early in the award we pursued a project with the LLE and collaborators at LLNL to try to implement energy-dispersive x-ray diffraction in OMEGA. This resulted in a careful photometric study (Hoidn and Seidler 2013), but the ride-along shots at OMEGA gave inconclusive results about the ability to sufficiently screen the TIM's for this method to function by the most direct methods.

IV. Technical Development

Our research group is well known for development of novel instrumentation for use at x-ray facilities. During the term of this award, a spin-off of the effort to develop a portable, multi-analyzer spectrometer for potential use at LCLS resulted in unexpected advances in lab-based x-ray spectroscopy, including several patents and a nascent commercialization effort (See easyxafs.com). It has also resulted in the development of a 'disposable' energy-resolving x-ray camera (Hoidn and Seidler 2015) that we expect will be used at LCLS in the LP70 beamrun as part of a new beam spectral/ intensity monitor, and which may also see future application in several laboratory-scale facilities doing laser-generated x-ray physics.

Filed patents that made use of some partial FTE under this award:

U.S. Patent Application No. 15/ 392,430 filed 12/ 28/ 2016
Entitled: "X-Ray Spectrometer with Source Entrance Slit"
Inventors: Devon Mortensen, Gerald Seidler

International Patent Application No. PCT/ US2016/ 068837 filed 12/ 28/ 2016
Entitled: "Methods for Aligning a Spectrometer"
Inventors: Devon Mortensen, Gerald Seidler

U.S. Provisional Patent Application No. 62/ 394,981 filed 9/ 15/ 2016
Entitled: "A Miniature Spectrometer for Wavelength Dispersive X-ray Emission Spectroscopy of Air-Sensitive and Hazardous Materials"
Inventors: Gerald Seidler, Oliver Hoidn, William Holden

U.S. Provisional Patent Application No. 62/ 271,989 filed 12/ 28/ 2015
Entitled: "Improved X-ray Emission Spectrometer"
Inventors: Devon Mortensen, Gerald Seidler

U.S. Provisional Patent Application No. 62/ 271,992 filed 12/ 28/ 2015
Entitled: "Improved X-ray Optic Alignment by Controlled Azimuthal Rotation"
Inventors: Devon Mortensen, Gerald Seidler