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# FY05 LDRD Final ReportTime-Resolved Dynamic Studies using Short Pulse X-Ray Radiation

A. Nelson, J. Dunn, T. van Buuren, K. Budil, B. Sadigh, G. Gilmer, R. Falcone, R. Lee, A. Ng

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# FY05 LDRD Final Report
# Time-Resolved Dynamic Studies using Short Pulse X-Ray Radiation
# LDRD Project Tracking Code: 04-ERD-010


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**Abstract**

Established techniques must be extended down to the ps and sub-ps time domain to directly probe product states of materials under extreme conditions. We used short pulse (≤ 1 ps) x-ray radiation to track changes in the physical properties in tandem with measurements of the atomic and electronic structure of materials undergoing fast laser excitation and shock-related phenomena. The sources included those already available at LLNL, including the picosecond X-ray laser as well as the ALS Femtosecond Phenomena beamline and the SSRL based sub-picosecond photon source (SPPS). These allow the temporal resolution to be improved by 2 orders of magnitude over the current state-of-the-art, which is ~100 ps. Thus, we observed the manifestations of dynamical processes with unprecedented time resolution. Time-resolved x-ray photoemission spectroscopy and x-ray scattering were used to study phase changes in materials with sub-picosecond time resolution. These experiments coupled to multiscale modeling allow us to explore the physics of materials in high laser fields and extreme non-equilibrium states of matter. The ability to characterize the physical and electronic structure of materials under extreme conditions together with state-of-the-art models and computational facilities will catapult LLNL's core competencies into the scientific world arena as well as support its missions of national security and stockpile stewardship.


**Introduction/Background**

Ultrafast x-ray science is an emerging field of research in which x-ray techniques are used in combination with femtosecond lasers to probe in "real time" the electronic and structural dynamics of molecules, biological systems, liquids, solids, surfaces and interfaces. Implementation of these methods relies on the newest technological developments in x-ray sources and detection systems. Moreover, new concepts and new theoretical tools have to be developed to fully exploit the potential of time resolved techniques. These technique are ideal for LLNL's stockpile stewardship mission, which requires detailed knowledge of how materials respond to strong shocks or other extreme conditions on rapid timescales.

Our current success performing time-of-flight (ToF) photoemission spectroscopy with picosecond time resolution using the COMET x-ray laser [1-5] has allowed LLNL to establish a foothold in this exciting new field of research. Femtosecond X-rays from the new Sub-Picosecond Photon Source (SPPS) at SLAC

will now bridge the gap to the sub-picosecond time-scale, and allow us to look at the evolution of matter under conditions where electronic structure changes are the main driving force for reactions and phase changes instead of temperature.

**Research Activities**

We used high-energy laser drives (e.g. short pulse lasers, LLNL B174 facilities USP and COMET) to melt and shock materials, specifically ultra-thin foils of Cu, in combination with several X-ray diagnostic probes: (1) the LLNL COMET tabletop X-ray laser (XRL) with ToF photoemission spectroscopy for the low energy regime (<100 eV), (2) NEW Femtosecond Phenomena Beamline at ALS/LBNL (130-1800 eV) with x-ray absorption spectroscopy, and (3) the Sub-Picosecond Photon Source (SPPS) at SLAC (8 keV). We also began to characterize x-ray heating of materials to quantify the x-ray/material interaction.

*COMET e-ToF.* The COMET XRL source delivers $10^{12}$ photons in 2 – 5 ps giving a peak brightness of ~$10^{25}$ ph. mm$^{-2}$ mrad$^{-2}$ s$^{-1}$ 0.1% BW$^{-1}$. The electron time-of-flight (e-ToF) spectrometer is capable of high-energy resolution ($\Delta E/E<10^{-3}$) with high throughput. It has been successfully used to energy analyze shallow core level photoelectrons produced by XRL photoionization which allows picosecond time resolved measurements giving electron spectra in a single shot. We also used soft x-ray radiography to simultaneously characterize x-ray transmission, and therefore changes in material opacity, through the ultra-thin metal foils during melt.

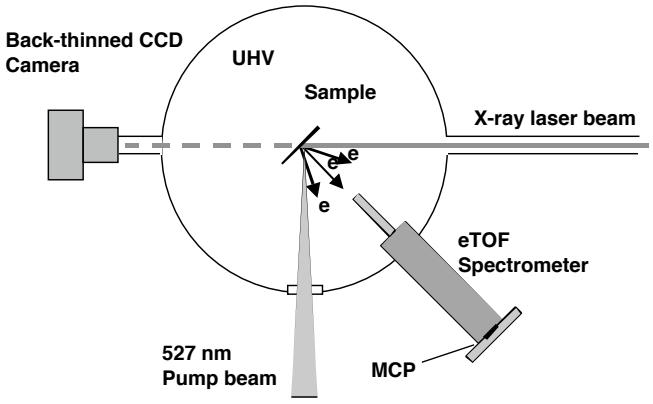


Figure 1: Schematic of optical pump/x-ray probe experiment with soft x-ray radiography.

*B174 Ultra Short Pulse (USP).* We successfully studied the disassembly dynamics of fs-laser heated ultra-thin Cu foils at the Ultra Short Pulse (USP) laser facility in B174 using our well developed femtosecond pump-probe techniques. Freestanding, 50nm Cu foils are illuminated with a 150fs (FWHM), 400nm pump laser pulse at normal incidence. For a focal spot diameter of 80mm (FWHM), the incidence laser irradiance can reach 5x$10^{13}$ W/cm$^2$ (or fluence of 7.5J/cm$^2$). Absorption of pump laser is determined from spatially resolved reflectivity and transmissivity measurements. The change in optical properties and the dynamic behavior of the heated foil are monitored using Frequency Domain Interferometry (FDI). This diagnostic measures the change in phase shift, $\delta f$, of the 150fs (FWHM), 800 nm probe reflected off the front surface of the foil. $\delta f$ is sensitive to both the dielectric function and hydrodynamic expansion of the heated material. By varying the relative time delay between the 400nm pump and the 800nm probe, we will be

able to determine the onset of target disassembly. This technique has been successfully used to verify the existence of a stable liquid phase in *fs*-laser heated Au foils for durations of 2-20 ps, depending on the energy density of the heated sample.

*Femtosecond Phenomena Beamline (ALS/LBNL)*. A NEW femtosecond phenomena beamline has been built at the Advanced Light Source (ALS). Beamline 6.0.2 specifics are 130 to 1800 eV (monochromatic) x-rays and ~3 x $10^6$ photons/s/0.1% BW. A short laser pulse (150 fs duration, 800 nm wavelength) will be used to rapidly heat (and melt) an ultrathin metal foil, producing a non-equilibrium state that persists while x-rays probe absorption through the sample. The resulting spectrum offers information on both the electronic structure via x-ray absorption near-edge spectroscopy (XANES) and the related atomic structure via extended x-ray absorption fine structure (EXAFS) of the transient state.

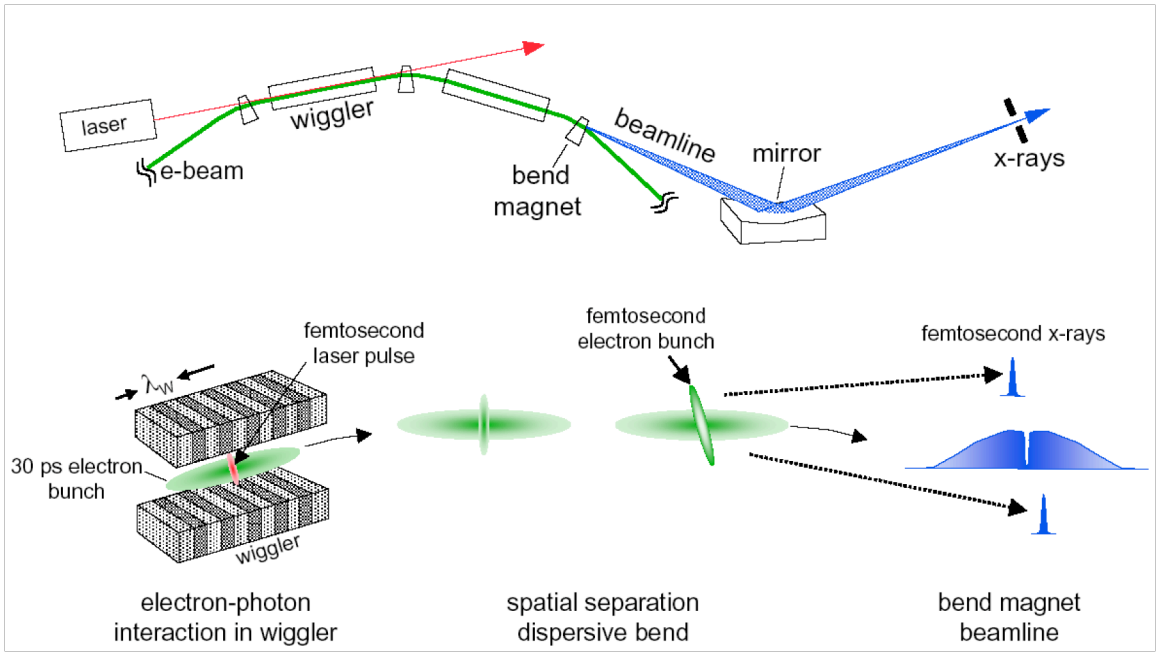


Figure 2. Schematic of Femtosecond Phenomena Beamline at the Advanced Light Source, Lawrence Berkeley National Laboratory.

*Sub-picosecond Photon Source (SPPS/SLAC).* The high brightness and sub-picosecond pulse length of the SPPS will allow us to extend x-ray diffraction and x-ray absorption spectroscopy to study fast dynamics at the atomic level. SPPS

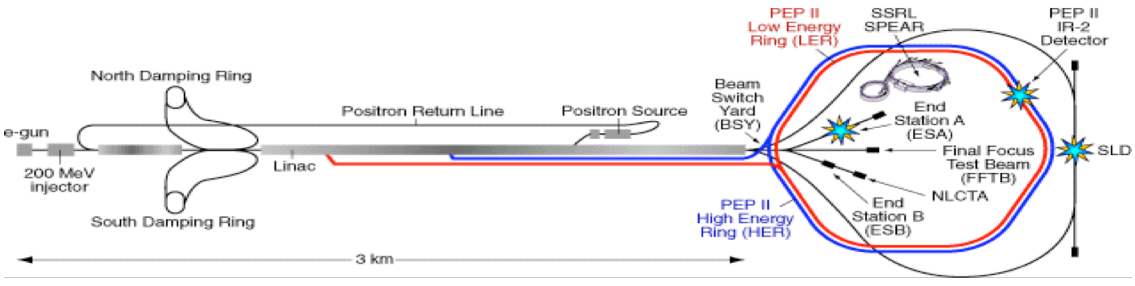


Figure 3. Sub-picosecond Pulse Source (SPPS) at Stanford Linear Accelerator (SLAC).

specifics are $10^8$ photons per pulse with a peak brightness of $10^{25}$ photons $mm^{-2}$ $mrad^{-2}$ $s^{-1}$ per 0.1% BW. A Ti:Sapphire short pulse laser is used for heating (100 mJ at 30fs). Our initial consortium experiment on x-ray pulse measurements by chirped pulse laser assisted Auger decay using our new ToF analyzer was to begin in November 2004 but has been rescheduled in July 2005 due to the SLAC accident and shutdown. We will also focus the SPPS x-rays to sub-micron spots to heat materials and probe with the ultrafast laser pulse.

*Materials Modeling & Simulation.* Modeling short-pulse laser ablation is challenging because of the diversity of rapid electronic and structural changes that occur. Short-pulse laser irradiation creates a non-equilibrium electron distribution at elevated temperatures while leaving the lattice at room temperature. In order to model laser interaction with metals, classical molecular-dynamics simulations need to be complemented with a model for the dissipative dynamics of the electron-ion system that can bring the two subsystems into equilibrium with each other. The most common treatment is through the phenomenological Two-Temperature Model (TTM) invented by Anisimov et al. This theory proposes that the relaxation of hot electrons is governed by two coupled non-linear differential equations. Thus, in a time equal to a few phonon oscillations (on the order of picoseconds) electron-phonon interactions will allow the two subsystems to exchange energy and reach local equilibrium. Subsequently, a slower relaxation process dominated by heat diffusion returns the system to ambient temperature. The electronic contribution to the thermal conductivity is furthermore essential for the redistribution of the laser energy in the irradiated substrate. Hence a conventional MD simulation where only lattice contributions are present leads to unphysical confinement of the laser energy to the surface region of the irradiated target.

**Results/Technical Outcome**

In the first year, we succeeded in the custom fabrication of ultra-thin (50 nm) metal foils critical for this experiment. Uniform heating of a metal 'slab' is required for a clear interpretation of ultrafast phenomena. We also improved the time response and low energy sensitivity of the e-ToF spectrometer by upgrading the detector and adding additional Mu metal shielding. Two x-ray CCD cameras were also procured and installed at the SPPS facility at SLAC that were to be used in our planned experiments on two-photon ionization of the Cu K-edge with subsequent observation of the Auger cascade of the excited state.

Also in the first year, we applied molecular dynamics models to examine void nucleation in Cu thin films for a range of temperature and pressure values, simulating the conditions after laser irradiation. We have further studied the dynamics of a shock-induced phase transformation in Ti, using a very recent and novel inter-atomic potential for this system. We observe a three-wave shock front where the initial alpha-Ti structure (hcp) becomes plastic and subsequently undergoes a transition to the omega phase. This simulation is the first of its kind, where a three-wave shock front is produced, with excellent agreement with experiments.

We also developed a new massively parallel hybrid atomistic-continuum model, where the classical molecular-dynamics simulation technique is combined with a continuum description of the laser excitation and subsequent relaxation of the conduction band electrons. It is the first code of this kind, where fully 3-dimensional treatment of the electronic heat conduction, as well as energy transfer between the electron and ion subsystems is implemented within a massively parallel paradigm.

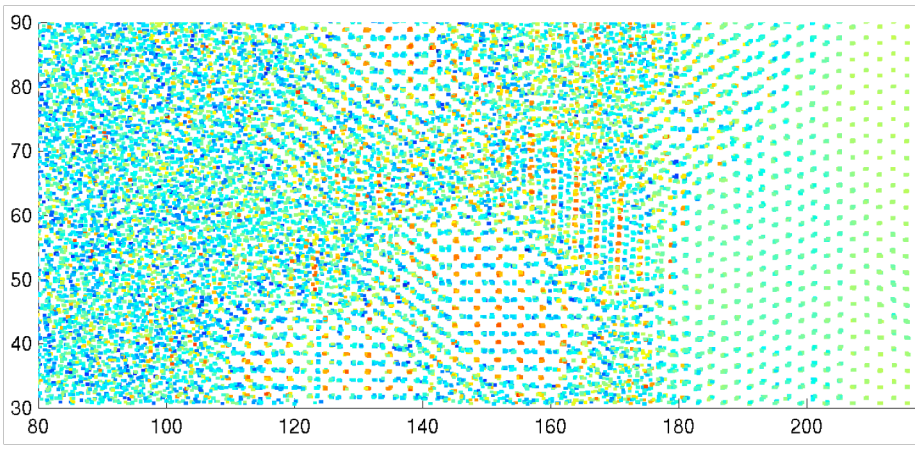


Figure 4. Cross-sectional view of a shock wave traveling through Ti, causing a phase transformation. The maximum pressure is 40GPa, and the piston is on the left of the figure moving at 1.5km/sec into a cold initial sample. The region near 200Å is the untransformed hcp Ti; and the omega phase is in the region around 160Å on the x-axis. The coloring is based on the average coordination of each atom obtained using Voronoi cell decomposition. The blue color indicates 10 fold coordinated atoms, green 12, and red 14.

Hence the simulation involves simultaneous calculations of the molecular-dynamics equations of motion for the ions, together with expressions to account for the interactions with the electrons. We are applying this to the evolution of photomechanical damage in large-scale simulations of laser spallation of various thin films of Ni and Cu.

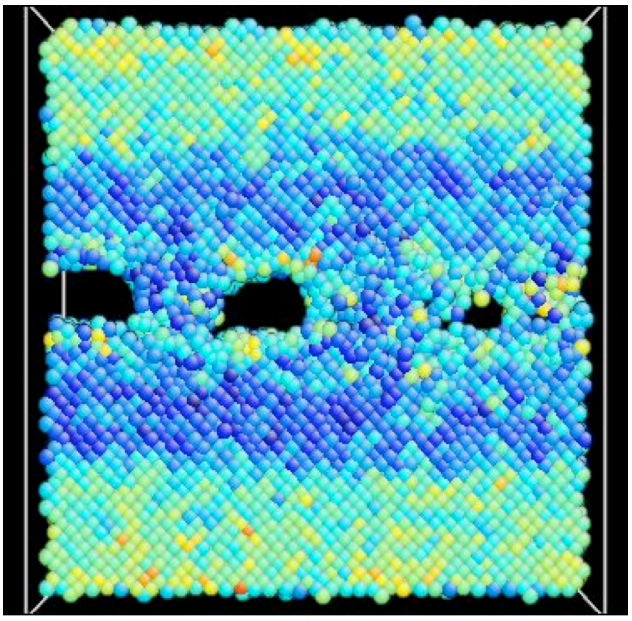

Figure 5. Void nucleation in a copper thin film (130A thick) upon release of an initial compression. The colors correspond to the atomic level hydrostatic pressure, with blue indicating tensile stress. In this case, the voids collapsed and the system recovered the initial crystal structure.

In addition, we studied the effect of electronic excitations on the phase stability, elasticity and lattice dynamics of various metallic systems. We assume that the dominant excitation of the electronic subsystem is in the form of Fermi-Dirac broadening of the conduction band electrons. We find that high electron temperatures can lead to considerable lattice expansion (hence hydrostatic pressure), and fairly modest renormalization of the shear elastic constants for simple systems such as Al and Cu. However for more complex transition metals such as Ti, we find that electronic temperature can dramatically modify the relative energy of various crystal structures, such as the Ti bcc and hcp phases. We introduced the notion of an electronic temperature-pressure phase diagram, different from the conventional phase diagrams where ion temperature is used.

In the second year of our project, our published work in Applied Physics Letters [APL 85, 6290 (2004)] was featured in the Virtual Journal of Ultrafast Science (http://www.vjultrafast.org), in the 2004 CMS Annual Report, and in the June 2005 Science & Technology Review. Results were also presented at the MRS Fall 2004 Meeting. Our work is also featured at Science Day with a poster presentation entitled "Ultrafast Science."

We also proceeded with optical measurements of the disassembly dynamics of *fs*-laser heated ultra-thin Cu foils showing the existence of a metastable liquid phase persisting 4-5 ps. This metastable phase was further studied using a *fs* laser pump pulse to create heated conditions that are probed with x-ray laser induced time-of-flight photoelectron spectroscopy.

Changes in the optical properties and the dynamic behavior of the heated foil were monitored using Frequency Domain Interferometry (FDI). [6,7] This diagnostic measures the change in phase shift, $\delta\phi$, of the 150-fs (FWHM), 800-nm probe reflected off the front surface of the foil which was heated by a 150-fs (FWHM), 400-nm pump pulse at an intensity of 4 – 8 X $10^{12}$ W/cm$^2$. The average energy of the

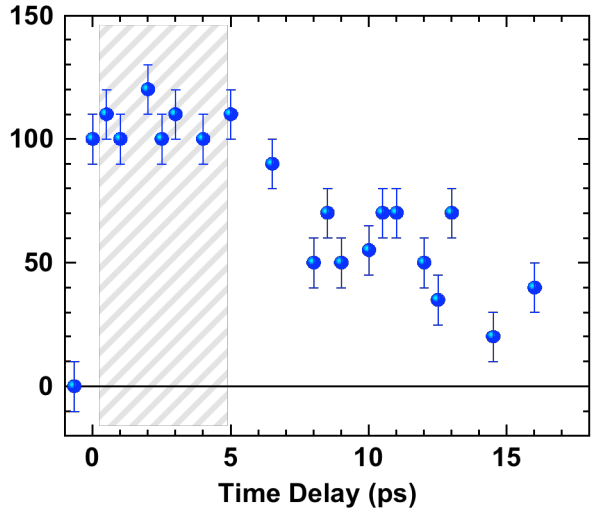


Figure 6. Temporal behavior of the relative phase shift (dF) of the 800-nm probe pulse reflected off the isochorically heated ultra-thin Cu slab. The hatched area indicates the existence of a metastable liquid phase.

pump pulse at the target was 30 μJ. $\delta\phi$ is sensitive to both the dielectric function and hydrodynamic expansion of the heated material. By varying the relative time delay between the 400-nm pump and the 800-nm probe, we were able to determine the onset of target disassembly as shown in Figure 6. Note that the 800 nm probe and 400 nm pump wavelengths are below and above the interband absorption threshold, respectively, for Cu. [8] Also, for reference, the 1/e penetration depth of 2w light (400 nm, 3.10 eV) in Cu is 14.9 nm. Furthermore, the ballistic electron range in Cu is 70 nm, thus resulting in a uniformly heated 50 nm thin Cu slab for these experiments. The FDI measurements show the existence of a metastable liquid phase in the *fs*-laser-heated ultra-thin Cu foils persisting for approximately 4-5 ps (hatched area in Figure 1). Similar results have been observed for Au and Al. [9] The persistence of this single state for several picoseconds allow us to accurately probe the dynamic conditions of the foil with picosecond duration x-rays.

Figure 7 presents the single-shot e-ToF photoemission spectra of the 3*d* valence band of static and laser heated ultra-thin polycrystalline Cu foil illuminated with $10^8$ – $10^9$ x-ray laser photons. This sample is sufficiently thin to observe the transmitted x-ray laser beam simultaneously with the photoelectron signal.

The valence band maximum of Cu is composed of *s*, *p*-like states above the dominant Cu 3*d* shell, which is the strongest event in the e-ToF spectrum. The strong Cu *d*-state emission observed at the valence band maximum, i.e. first e-ToF event, is indicative of a high density of states (filled *d*-states) below the *s*, *p*-like states and Fermi level.

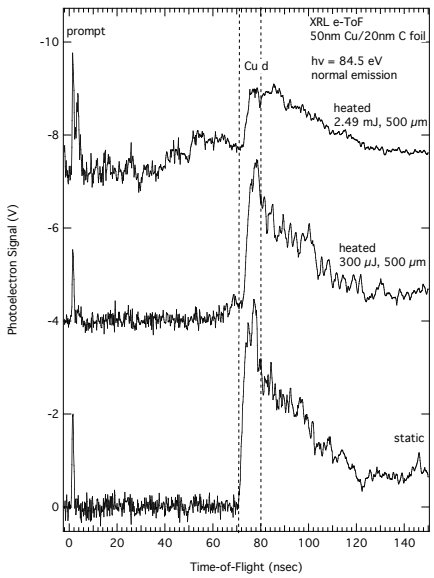


Figure 7. X-ray laser induced photoelectron spectra (signal versus time-of-flight) for static and laser heated ultra-thin polycrystalline Cu foil

Based on our separate FDI measurements showing the existence of a metastable liquid phase in the *fs*-laser-heated ultra-thin Cu foils persisting for approximately 4-5 ps, the x-ray laser probe was now used to investigate the evolution of the electronic structure of the metastable state. The pump-probe experiment was performed with the COMET platform 400 fs, 527 nm (2.35 eV)

optical pump beam focused onto a large 500 ´ 700 $\mu m^2$ (FWHM) spot on the ultra-thin (50 nm) polycrystalline Cu foil. Note that the optical pump energy is above the interband absorption threshold and has a 1/e penetration depth in Cu of 16.2 nm. The x-ray laser probe and optical pump were timed to coincide on the sample. Two laser heating conditions were applied (1) 300 μJ of laser energy corresponding to 2 ´ $10^{11}$ W $cm^{-2}$ intensity and (2) 2.49 mJ of laser energy corresponding to 1.8 ´ $10^{12}$ W $cm^{-2}$ intensity. To estimate the temperature of the 50 nm Cu foil for both pump energies taking into account reflection and transmission at 527 nm and using the room temperature heat capacity, we calculated the final temperatures for the two cases to be (1) 1929 K and (2) 15,000 K. It is noted that the long thin aspect ratio of the x-ray laser beam overfilled the optical beam on the sample in the vertical direction so that there are detected photoelectron signals from heated and unheated regions of the foil.

The single-shot e-ToF photoemission spectra representing the evolution of the valence band electronic structure of the femtosecond laser heated ultra-thin polycrystalline Cu foil (Fig. 7) clearly show that the observed 3*d* peak intensity decreases with increasing laser energy and that there exist features before the strong onset that represents the valence band maximum. For laser heating condition (1) at 300 μJ, the decreasing Cu 3*d* peak intensity is due to a rapid depopulation of the *d*-band as the electron temperature $T_e$ increases for the resultant metastable phase. [10,11] The temperature dependent depopulation of the valence band *d* states also creates vacancies in the conduction band (CB) thus allowing intra-atomic absorption below the edge (e.g., 3*d*-4*p* transitions in Cu) as observed. In addition, we observe that the Cu 3*d* peak shifts towards lower kinetic energy (higher binding energy) indicating that the band is 'sinking' in the overall electronic structure of the persistent metastable phase. Depopulation of the *d*-band is predicted to affect its binding energy in this manner. [10,11] Also, note that there is no broadening of the Cu 3*d* upon heating which implies a nonequilibrium distribution of occupied states i.e. spreading of the Fermi-Dirac electron energy distribution. Increasing the pump energy by a further factor of ten [laser heating condition (2)] further depopulates the *d* band and generates a strong electron signal before 50 ns (Fig. 7) that indicates the sample is now in an ionized state. Also, note the two prompt peaks separated by 2 ns, that we attribute to multiple scattering of the incident x-rays during skin layer expansion and ablation of the Cu. Post mortem electron microscopy did indeed reveal that Cu ablation had occurred with the 2.49 mJ pump energy.

Modeling accomplishments in the second year included applying the newly developed hybrid atomistic-continuum model to the evolution of photomechanical damage in large-scale simulations of laser melting and spallation of various thin films of Ni and Cu. Figure 8 shows an example of results from this model. The temperature contour plot for simulation of a 50 nm free-standing metal film heated by a 200 fs pulse at a laser fluence of 0.043 $J/cm^2$ is shown. Laser energy is deposited during the laser pulse to the conduction band electrons and the energy transfer from hot electrons to the lattice leads to the initial temperature increase during the first few ps. Strong electron-phonon coupling leads to a large temperature gradient in the film. After a few ps the surface region of the irradiated target is heated far above the equilibrium melting point.

Structural changes in a thin copper film resulting from femtosecond laser ablation are shown in Figure 9. These results illustrate the ability of the model to treat rapid transformations. As can be seen in this sequence, the rapid heating causes the film to melt and expand in thickness. This expansion accelerates the top half in the upward direction, and the lower half downward. If the energy density of the pulse is sufficient, the two halves will pull apart and nucleate voids in the middle. These may collapse later as a result of surface tension, or continue to expand until

the film splits into two pieces. Our simulations show that the threshold energy density to split the film in two is a sensitive function of the film thickness. Although very thin films clearly have reduced strength, they will also experience very weak tensile stress because of the relaxation at the free surface. We are presently examining the balance between these effects as a function of film thickness using simulation of ablation in films over a wide range of thicknesses. Since the atomistic model can treat both the highly non-equilibrium conditions and the nucleation of voids and defects, it can provide an accurate description of these processes.

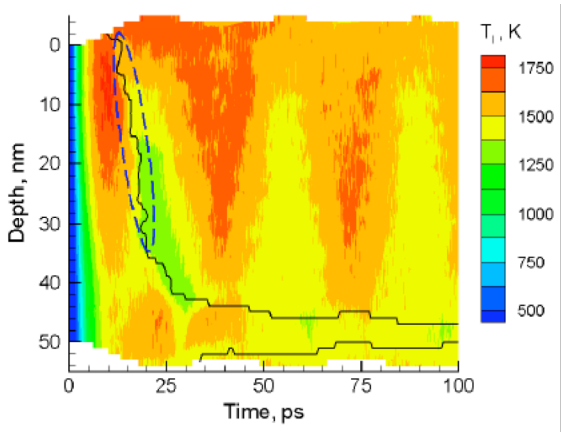


Figure 8. Temperature contour plot for simulations of laser melting of a 50 nm Ni film irradiated with a 200 fs laser pulse with a fluence of 0.043 J/cm$^2$ is shown.

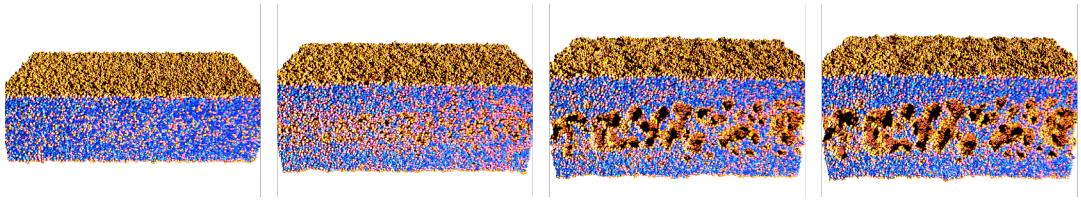

Figure 9. A time sequence of snapshots taken at 1, 14, 24, and 33 ps after irradiation showing a cross-section through a thin film. The energy density of the pulse was slightly below threshold, and the voids eventually collapse. The void nucleation rate is a maximum in the center of the film, where the maximum tensile stress develops.

**Exit Plan**

This ultrafast materials response project combined the resources of DNT, CMS and PAT in collaboration with UC Berkeley and provided a clear vision for ultrafast examination of extreme states of matter, investigations that are directly related to LLNL's missions of national security and stockpile stewardship. We delivered sub-picosecond pump-probe spectroscopic capabilities with advanced diagnostics and new high-performance modeling/simulations for the prediction of the physical and electronic structure of materials under extreme conditions. The experimental capabilities together with state-of-the-art models and computational facilities will enhance the scientific foundation for LLNL's evolving missions.

The now established LLNL expertise in ultrafast science will be applied to experiments at the Linac Coherent Light Source (LCLS) at SLAC, the world's first

hard x-ray free electron laser when it becomes operational in 2009. The unique x-ray laser-like qualities of the LCLS have the potential to revolutionize the experimental investigation of structural dynamics with x-ray techniques by directly following the time evolution of the electron density during the course of a biological, chemical, or physical transformation. It will also be used to directly create extreme states of matter at high temperature and density, and to probe the ablation/damage process to study structural changes and disintegration. LCLS will be a new platform to study the interaction of intense x-rays with matter.

## Summary

We have successfully demonstrated the optical pump-x-ray laser probe characterization capability to observe the evolution of changes in the electronic structure of laser heated ultra-thin Cu with picosecond time resolution. The first preliminary measurements of the disassembly dynamics of fs-laser heated ultra-thin Cu foil were also performed. Single-shot spectra have been achieved showing that the incident photon number is more than enough for characterizing dynamical events. The dynamic pump-probe results reveal depopulation of the Cu *d*-band and spreading of the Fermi-Dirac electron energy distribution. We will further investigate and characterize the range of x-ray laser intensities on the sample and the effect of space charge on the nonequilibrium electron energy distribution in future studies.

## Acknowledgements

We would like to acknowledge the many contributions and excellent technical support to this work by Jim Hunter and Roger van Maren, and we would also like to thank Mark Eckert and Al Osterheld for their continued support. In addition, we appreciate and thank the SSRL and ALS staff for their assistance during experiments at their facilities. Work conducted at the Stanford Linear Accelerator Center and the Advanced Light Source at Lawrence Berkeley National Laboratory, was supported by the U.S. Department of Energy under contract nos. DE-AC03-76SF00515 and DE-AC03-76SF00098, respectively.

## References

1. J. Dunn, R.F. Smith, J. Nilsen, A.J. Nelson, T.W. Van Buuren, S.J. Moon, J.R. Hunter, J. Filevich, J.J. Rocca, M.C. Marconi, V.N. Shlyaptsev, A picosecond 14.7 nm x-ray laser for probing matter undergoing rapid changes, *X-Ray Lasers 2002: 8th International Conference on X-ray Lasers*, ed. J.J. Rocca et al, AIP Conference Proceedings No. **641**, 481 (2002).
2. A.J. Nelson, J. Dunn, T. van Buuren and J. Hunter, Appl. Phys. Lett. **85(25)**, 6290 (2004).
3. A. J. Nelson, J. Dunn, T. van Buuren and J. Hunter, Single-state electronic structure measurements using time-resolved x-ray laser induced photoelectron spectroscopy, Neutron and X-Ray Scattering as Probes of Multiscale Phenomena, Materials Research Society Conference Proceedings No. **840**, Q4.6 (2004).
4. A.J. Nelson, J. Dunn, J. Hunter and K. Widmann, Appl. Phys. Lett. **87(15)**, 154102 (2005).
5. A.J. Nelson, J. Dunn, K. Widmann, T. Ao, Y. Ping, J. Hunter, and A. Ng, Advancements in time-resolved x-ray laser induced time-of-flight photoelectron spectroscopy, Presented at the SPIE 50th International Symposium on Optical

Science & Technology, Soft X-Ray Lasers and Applications VI, San Diego, Proc. SPIE Int. Soc. Opt. Eng. **5919**, 139 (2005).
6. J.P. Geindre, P. Audebert, A. Rousse, F. Falliès, J.C. Gauthier, A. Mysyrowicz, A. Dos Santos, G. Hamoniaux, and A. Antonetti, Opt. Lett. **19**, 1997 (1994).
7. K.Widmann, G. Guethlein, M.E. Foord, R.C. Cauble, F.G. Patterson, D.F. Price, F.J. Rogers, P.T. Springer, R.E. Stewart, A. Ng, T. Ao, A. Forsman, Phys. Plasmas **8**, 3869 (2001).
8. J.F. Janak, A.R. Williams and V.L. Moruzzi, Phys. Rev. **B11**, 1522 (1975).
9. K. Widmann, T. Ao, M. E. Foord, D. F. Price, A. D. Ellis, P. T. Springer, and A. Ng, Phys. Rev. Lett. **92**, 125002 (2004).
10. D.F. Price, R.M. More, R.S. Walling, G. Guethlein, R.L. Shepherd, R.E. Stewart, W.E. White, Phys. Rev. Lett. **75**, 252 (1995).
11. D. Fisher, M. Fraenkel, Z. Zinamon, Z. Henis, E. Moshe, Y. Horovitz, E. Luzon, S. Maman and S. Eliezer, *28th European Conference on Laser Interaction with Matter*, Rome (2004).

**Appendix: Publications related to LDRD work**

**Journal Publications**

1. A.J. Nelson, J. Dunn, T. van Buuren and J. Hunter, X-ray laser induced photoelectron spectroscopy for single-state measurements, Appl. Phys. Lett. **85(25)**, 6290 (2004). UCRL-JRNL-205272.

2. A.J. Nelson, J. Dunn, J. Hunter and K. Widmann, Time-Resolved X-ray Laser Induced Photoelectron Spectroscopy of Isochoric Heated Copper, Appl. Phys. Lett. 87(15), 154102 (2005). UCRL-JRNL-207542.

3. James Dunn, Jorge Filevich, Raymond F. Smith, Stephen J. Moon, Jorge J. Rocca, Roisin Keenan, Joseph Nilsen, Vyacheslav N. Shlyaptsev, James R. Hunter, Andrew Ng, and Mario C. Marconi, "Picosecond 14.7 nm interferometry of high Intensity laser-produced plasmas", Laser and Particle Beams **23**, 9 - 13 (2005). UCRL-JRNL-207241.

**Conference Proceedings and Invited Presentations**

1. A. J. Nelson, J. Dunn, T. van Buuren and J. Hunter, Single-state electronic structure measurements using time-resolved x-ray laser induced photoelectron spectroscopy, Neutron and X-Ray Scattering as Probes of Multiscale Phenomena, Materials Research Society Conference Proceedings No. **840**, Q4.6 (2004). UCRL-ABS-204842.

2. A.J. Nelson, J. Dunn, K. Widmann, T. Ao, Y. Ping, J. Hunter, and A. Ng, Advancements in time-resolved x-ray laser induced time-of-flight photoelectron spectroscopy, Presented at the SPIE 50th International Symposium on Optical Science & Technology, Soft X-Ray Lasers and Applications VI, San Diego, Proc. SPIE Int. Soc. Opt. Eng. **5919**, 139 (2005). UCRL-PROC-214158.

3. J. Dunn, R. Keenan, S. Moon, A. Nelson, J. Nilsen, R. Shepherd, R. Smith, F. Weber, H. Fiedorowicz, A. Bartnik, J. Rocca, J. Filevich, V.N. Shlyaptsev, P. Zeitoun, "X-ray Laser Source Development and Applications at Lawrence Livermore National Laboratory", Invited talk given at Institute for Optoelectronics, Military University of Technology, Warsaw, Poland 6 September, 2005. UCRL-PRES-216144.

4. J. Dunn, R. Keenan, S. Moon, A. Nelson, J. Nilsen, R. Shepherd, R. Smith, J. Rocca, J. Filevich, V.N. Shlyaptsev, P. Zeitoun, "Characterization and Applications of Laser-Driven X-ray Lasers", Invited talk given at Atomic Spectroscopy in High Fields Workshop Piaski, Poland 6 - 11 September, 2005. UCRL-CONF-216064.