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Quarterly progress report for Q2 FY06 for Complex Transient Events in Materials Studied Using Ultrafast Electron Probes and Terascale Simulation (FWP SCW0289)

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Quarterly progress report for Q2 FY06 for

Complex Transient Events in Materials Studied Using Ultrafast Electron Probes and Terascale Simulation (FWP SCW0289)

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Executive Summary

In this quarter (Q2 FY06), the DTEM underwent a substantial reconfiguration of its laser systems. The cathode laser system was changed to provide greater numbers of electrons per pulse by lengthening the time duration of the pulse to 30 ns. The greater number of electrons per pulse has allowed us to acquire high quality pulsed images and diffraction patterns. The spatial resolution in the single pulsed image has been measured at better than 20 nm. The diffraction patterns are now more comparable to conventional electron microscope operation. Examples are found in the body of the report. We summarize important achievements in the following list:

1. Instrument performance and design improvements:
   - The laser system was changed for the cathode photoemission system (75 ns at 1053 nm wavelength converted to 30ns at 211 nm wavelength) to give longer electron pulses at the same current to yield more electrons per pulse.
   - New specimen drive laser constructed.
   - New computer monitored and controlled alignment systems installed for both laser systems to facilitate laser alignment through a user friendly computer interface.

2. Experimental Progress:
   - The spatial resolution of pulsed images was tested by imaging a cross-section of multilayer thin foils with 30 nm and 20 nm periods. Single pulse images were observed to have spatial resolution better than 20 nm. This combination of 20 nm spatial and 30 ns temporal resolution is thought to be highest combined spatial and temporal measurement ever made.
   - The quality of single pulse electron diffraction patterns have been improved to the point where differentiating the HCP from BCC patterns in Ti is substantially easier. The spatial coherence of the electron illumination on the specimen was improved to give much smaller diffraction spots in the pattern.

Laser change over and pulse lengthening

Since the electron yields produced by 2 ns UV (213 nm) laser pulses generate insufficient signal for imaging (the images are noise limited) and the resolution of the diffraction patterns is degraded due to a converged electron beam on the specimen to increase signal levels, efforts to increase the number of electrons per pulse were continued in the months of January to mid-March. The most feasible, quickest to implement, and cost-effective solution was to lengthen the laser pulse. With longer laser pulses, the total charge and signal levels increase without degradation of the image spatial resolution due to space charge effects. However, this is, of course, a trade-off with temporal resolution. In principle, the total charge per pulse has a linear, 1 to 1 correlation with the laser pulse duration, i.e. increasing the pulse duration by a factor of 10 should increase the charge per pulse by 10. Since our Q-switched laser system has a fixed cavity length and therefore a fixed pulse duration, the cathode laser system was replaced with a system able to produce significantly longer pulses. The laser system change was necessary from the logistic standpoint as well, due to increased demands on the previous cathode laser system from other programs (the Coherent Infinity laser is the property of the NIF facilities), limiting its availability for the DTEM project. To meet the laser requirements in short order, an amplification stage was installed on the previous specimen drive laser system, which produces 75 ns, 1053 nm laser pulses, and the beam path of the laser was rerouted to the cathode laser beam path. Since the laser was an in-house, custom built LLNL system, several weeks were required to implement the changes and stabilize the system to meet the stringent UV laser beam quality and energy needed for photoemission. Due to the change over of the specimen drive laser to the cathode laser system, a new drive laser was constructed from salvaged systems and spare parts. The laser parameters of the drive system are less stringent in energy and pulse duration requirements, and a 10 mJ, 1064 nm, 12 ns pulse duration laser was used, which is more than sufficient to thermally drive the alpha to beta transition in Ti. The following list summarizes the notable changes to laser systems and DTEM in the past months.

- January 01-31, Cathode laser change over from the Infinity system (1064 nm, 3ns/ 213nm, 2 ns pulses) to the LLNL system (1053 nm, 75 ns/ 211 nm, 30ns pulses).
    - Amplification stage added to the LLNL system to increase pulse energy from 30 mJ to **600 mJ**.
    - Laser pulse slicer installed for arbitrary pulse duration selection from 75 ns down to 0.5 ns.
    - Transport optics installed to relay image the output of the laser 15 m to the conversion crystals (relay imaging is important since it preserves the spatial quality of the beam). The beam was demagnified from 8 mm to a 4 mm diameter to increase the beam intensity and the conversion efficiency to the 5th harmonic (211 nm) light.
    - It is important to note that the frequency conversion is a non-linear optical process and that the pulse duration for the UV beam is shorter than the fundamental, i.e. 75ns, 1053 nm pulses are converted to 30 ns,

211 nm pulses. **Thus, the pulse duration was increased by a factor of 15**.

- February 01-28, new specimen drive laser construction and testing and modification of the LLNL cathode laser system
  - Used parts from existing out-of-service laser system. Laser capable of producing **10 mJ of 1064 nm light with 12 ns pulse duration.**
  - Transport optics installed to relay image 12 m from the laser output to the final focusing lens (near the final steering mirror and laser port of the DTEM).
  - Drive laser focusing optics modified to improve beam quality and reduce spot size on specimen. **The $1/e^2$ drive laser spot size at the specimen plane is 125 µm by 190 µm**.
  - New computer monitored and controlled alignment systems installed and the existing pointing and centering optics were modified in the cathode laser beamline to facilitate laser alignment through a user friendly computer interface.
- March 01-24, installation of laser system diagnostics and initial Ti dynamic experiments with longer laser pulses.
  - Due to instabilities in the laser systems, the new lasers cannot run in single shot mode and must continually pulse at a 10 Hz repetition rate. For controlled single-shot, pump/probe experiment, computer controlled mechanical shutter system was installed and new Labview® programs were developed to control the laser timing and shutters.
  - Photo diodes, power meters and cameras were installed in the specimen drive laser beam path to monitor the timing, energy, and beam size and alignment, respectively. These diagnostics are necessary for pump/probe experiments.
  - Resolution studies were preformed to assess the DTEM imaging capabilities with longer pulses.
  - In the latter part of the month, the $\alpha \leftrightarrow \beta$ transition in Ti was studied in diffraction mode using nanocrystalline thin films and electrolytically-thinned, coarse-grained, bulk Ti samples.

Results with longer electron pulses

Lengthening the UV pulse by a factor 15 (2 ns to 30 ns) yielded raw electron counts per pulse that exceed 5×10^7 e^-, which is lower than expected. Using 2 ns UV pulses, the electron yields exceeded 8×10^6 electrons per pulse. Assuming the 1 to 1 correlation with laser pulse duration (linear photoemission), the electron pulse should be in excess of 10^8 electrons per pulse. The lower yields are not yet understood, but may be related to non-linear photoemission processes coupled with the electron optical limitations of collection. Nonetheless, with the increased number of electrons per pulse, image and diffraction pattern resolution increased. The raw pulsed electron beam is highly aberrant and must be spatially filtered, i.e. a C2 aperture used, to obtain interpretable images and diffraction patterns with reasonable resolution. However, increased beam coherency leads to lower signal levels. In pulsed mode, there are two additional ways (other than inserting a smaller C2 aperture) to spatially filter the beam while maintaining reasonable signals. The first method is to use an optimal C1 lens setting which gives both high current and spatial filtering by the fixed apertures in the condenser portion of the microscope column. The second method is less intuitive from the nominal microscope operation point of view and involves the Wehnelt bias settings. Under pulsed mode, the bias setting strongly affects the beam current, where the highest bias settings (low bias voltage) give the highest pulsed beam current. However, the low bias voltage adversely affects the beam quality; the beam is highly aberrant. This is primarily due to the large laser spot and photoemission region, which allow more off-axis electrons to be accelerated and collected. Thus, an optimum exists between the laser spot size, C1 setting, C2 aperture size, and the bias voltage, and there is a continual trade-off between beam current and quality. Additionally, with the high bias settings, the beam spot size is large and the electron beam drifts several microns shot to shot. Although the signal levels are lower, the intermediate bias settings offer a good compromise between signal current and beam quality. Under such settings, $5x10^6$ 'high-quality' electrons per pulse are routinely obtained, and the resolution of single-shot image is improved.

Figure 1 shows a montage of TEM images of a cross-section of a multilayer. The foil is comprised of alternating layers of carbon and gold with two different layer thicknesses, 20 nm and 30 nm. The 30 nm layers are clearly visible in the single shot image, and the 20 nm layers are faintly visible. The single shot image is noise limited since only 5×10^6 electrons (in a 30 ns pulse) arrived on the CCD. By averaging 50 shots, the image quality improved dramatically, image resolution and contrast were similar to the conventional image.

The graph in Figure 2 shows the variation in the pixel intensity across the multilayers. Intensity from both the 30 nm and 20 nm thick layers are clearly visible above the background, indicating that the resolution is at least 20 nm if not better. This is the best resolution, single-shot image obtained in the LLNL DTEM to date and is possibly the highest temporal/spatial resolution image ever to be recorded. As can be seen by the amount noise in the single-shot image, signal levels continue to be a problem and are still too low for effective diffraction contrast imaging, which require insertion of an

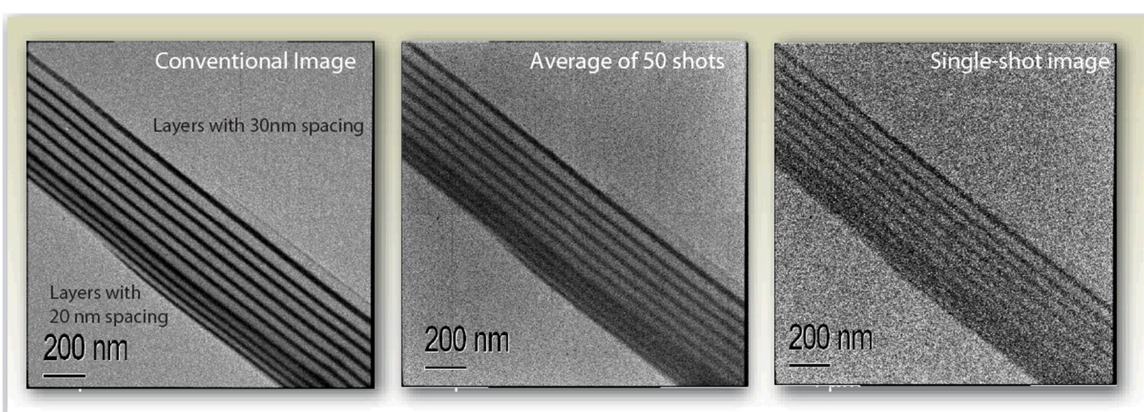

Figure 1. Composite figure of three images of a cross section of a Au/C multilayer foil. The image at left is a conventional TEM image. The image at center is an average of 50 DTEM pulses. The image at right is a single-shot image of the same area.

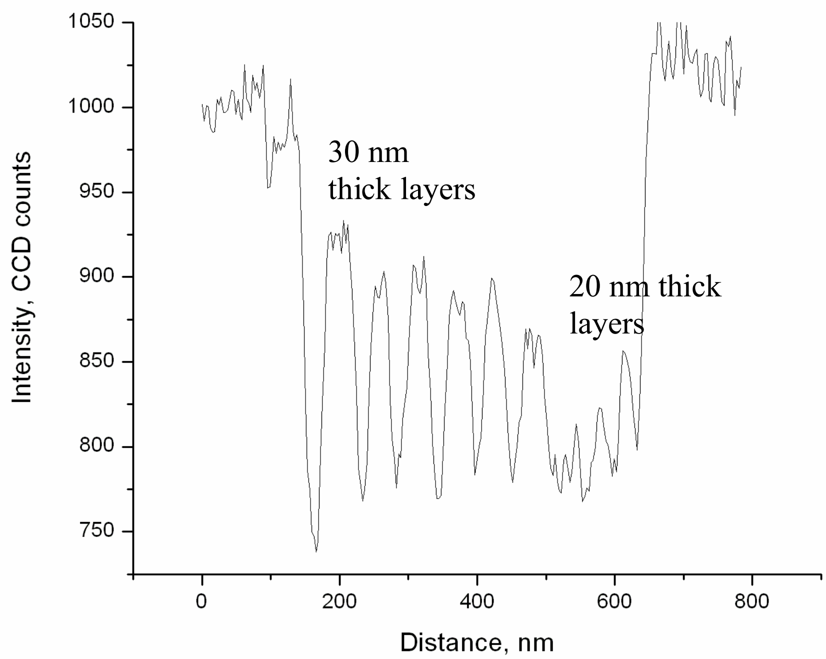

Figure 2. Graph show the pixel intensity as a function of distance across the multilayers.

objective aperture that further limits the signal reaching the CCD.

The diffraction pattern resolution also increased with longer electron pulses and increased signals. The main method for increasing signal on the CCD in diffraction mode has been to converge the beam on the specimen and form a pattern with a small illumination area and no selected area aperture. A converged beam has the adverse effect of increasing the breadth of the diffraction lines. With longer electron pulses, a more parallel illumination is used, and the diffraction lines are narrower. The diffraction line breadth is important for the observation of the $\alpha \rightarrow \beta$ transition. Broad diffraction lines or spots make it difficult to distinguish between the two phases. Being able to deconvolute the diffraction patterns and track the individual phase fractions as function of time is important experimental capability. Figure 3 illustrates the change in the diffraction pattern quality by stretching the UV laser pulse from 2ns to30ns.

There is an obvious increase in resolution diffraction pattern formed with 30 ns electron pulses; the spots are smaller and similar in diameter to CW pattern. In addition, the 1.5 ns pulsed electron diffraction patterns are more noisy due to low signal levels.

NEXT QUARTER GOALS

With increased resolution and signal in the single-shot time resolved diffraction patterns, the phase fractions can more accurately be determined, especially with low phase content. The next step is to track the phase transformation in incremental steps, i.e. 25 ns steps, and determine the phase fractions at various times and laser energies (temperature). Experiments using single-crystalline or coarse-grained Ti samples will be

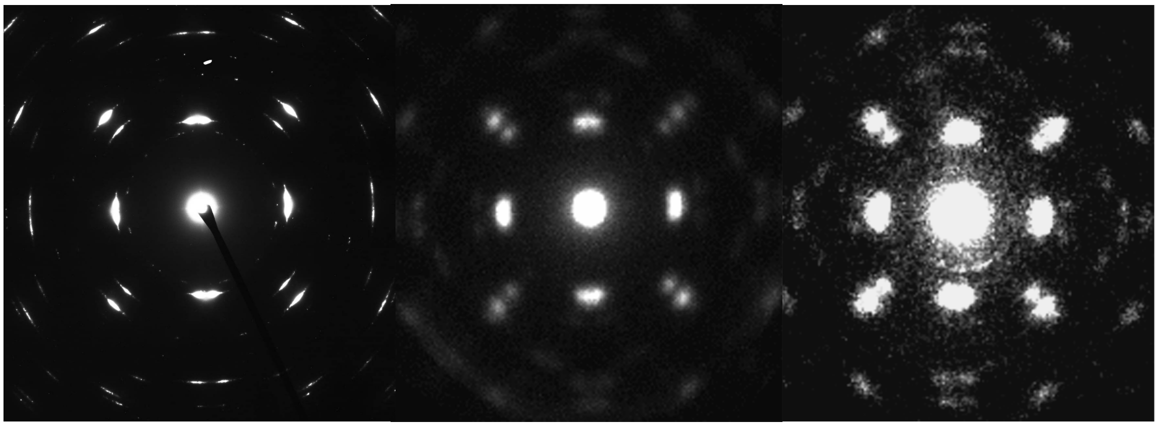

Figure 3. Composite figure of three diffraction patterns 40nm Ti foil showing the nominal room temperature HCP structure. The image on the left is a conventional TEM diffraction pattern taken with selected area aperture. The image in the center is a single-shot diffraction pattern using a 30 ns electron pulse. The image on the right is a single-shot diffraction using a 1.5 ns electron pulse.

conducted since sharper spots can be obtained and fewer BCC variants are formed, making it easier to track the transition and deconvolute the phases.

Publications Progress

Publications from our work with the DTEM have begun to appear. Conference proceedings are the first to appear. First from the PTM 2005 meeting:

1. G.H. Campbell, T.B. LaGrange, W.E. King, J.D. Colvin, A. Ziegler, N.D. Browning, H. Kleinschmidt, and O. Bostanjoglo, "The HCP to BCC Phase Transformation in Ti Characterized by Nanosecond Electron Microscopy," in Proceedings of the Solid-Solid Phase Transformations in Inorganic Materials 2005, Vol. 2, edited by J.M. Howe, D.E. Laughlin, J.K. Lee, U. Dahmen, and W.A. Soffa (TMS, Warrendale, PA, 2005) p. 443 - 448.

The next proceedings are from the MRS Fall 2005 meeting and has now appeared on the MRS web site:

2. Thomas B. LaGrange, Geoffrey H. Campbell, Jeffrey D. Colvin, Wayne E. King, Nigel D. Browning, Michael R. Armstrong, Bryan W. Reed, Judith S. Kim, and Brent C. Stuart, "*In-situ* Studies of the Martensitic Transformation in Ti Thin Films using the Dynamic Transmission Electron Microscope (DTEM)," in *In-Situ* Electron Microscopy of Materials, eds. P.J. Ferreira, I.M. Robertson, G. Dehm, and H. Saka (Materials Research Society Symposium Proceedings Vol. 907E) p. 0907-MM05-02.1 - 6.

The proceedings of the FEMMS2005 meeting are being published in a peer reviewed archival journal and so are taking longer to appear. Our paper has passed peer review and has been accepted:

3. Thomas LaGrange, Geoffrey H. Campbell, Jeffrey D. Colvin, Bryan Reed and Wayne E. King, "Nanosecond Time Resolved Electron Diffraction Studies of the α to β Transition in Pure Ti Thin Films using the Dynamic Transmission Electron Microscope (DTEM)," in the Proceedings of the Frontiers of Electron Microscopy in Materials Science, *Journal of Materials Science*, in press.

We have other manuscripts in progress and will report on them as they are completed and submitted.