Coherent Diffraction Imaging of time-resolved strain induced by a high-repetition rate fs-laser.

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APS Technical Working Group meeting
Argonne, March 17, 2011
Outline

- Motivation and previous S7 work
- Brief Overview of beamline 7ID and APS timing experiments
- Progress in using Coherent Diffraction Imaging in laser-pump x-ray probe experiments.
APS bunch patterns are ideal for timing experiments.

- **3.68 µs**
  - 150 ps FWHM, 16 mA
  - Hybrid singlet

- **1.59 µs**
  - 90 ps FWHM, 4.25 mA

- **11.3 ns**
  - 60 ps FWHM, 0.31 mA
  - If laser here
  - Then gate this bunch (& previous round trip)
  - (& maybe the next one)

- **153 ns**
  - 24-bunch

- **244.5 µs**
  - 324-bunch
  - 60 ps FWHM, 0.31 mA
  - It is used with the Micra 88 MHz oscillator
Adapted from R. Trebino class notes, and Eric Landahl
Brief overview of Sector 7

Dedicated Sector for Time-Resolved Science

Sector 7: Time-Resolved Research Group
- scattering
- imaging
- spectroscopy

Talk by A. March

Talk by S. Kelly

ANL Chemistry+XOR

Ultrafast Spectroscopy

Laser Enclosure

7-ID-D 7-ID-C

Ultrafast Surface Diffraction

Ultrafast Microbeam scattering

ps-White/pink beam Imaging

White/pink beam scattering
Resolution limited by the bunch duration (or the timing jitter)
Arbitrary pump-probe delay (NOT limited by bunch separation)
7ID-C set up

- Response of condensed matter to coherent excitation
- Thermal and non-thermal strain generation and melting in semiconductors
- Acoustic propagation and impedances at boundaries
- Structural Phase Transitions
- Chemical reactions on surfaces
- Coherent diffraction imaging of laser-strained nanostructure

Huber 6-circle available
X-ray focused with 200mm-long KB system, APDs, or Pilatus 100K used in laser-pump x-ray probe experiments

Time-resolved Bragg Diffraction (laser pump/x-ray probe)

- Response of condensed matter to coherent excitation
- Thermal and non-thermal strain generation and melting in semiconductors
- Acoustic propagation and impedances at boundaries
- Structural Phase Transitions
- Chemical reactions on surfaces

Experiment: InSb 111, 10mJ/cm²

Reis et al. Phys Rev. Lett.(86) 2001

Impulsive Strain Generation
(Thomsen et al. Phys Rev. B (24) 1986.)
Imaging strain generation with coherent X-rays

- When l/w ~ 1, simple thermoelastic model is no longer valid, and strain field is no longer uniaxial.
- A coherent diffraction technique is used on 7ID with D ~ 7 by 35 µm, w ~ 1-6 µm. (the laser absorption length l < 1 µm). Focused energy density about 10 mJ/cm²
- A high resolution CCD in the far field images the coherent diffraction pattern. In 324 bunch mode, no need for gated CCD or fast x-ray chopper.

Fs-laser oscillator work from C. Rossignol et al., PRL 94, 166106 (2005)
Refresher on Coherent X-ray diffraction

- Modern 3rd generation sources provide enough partially coherent flux to illuminate a sample of say (10µm)^3 coherently.
- The coherent flux for a source like the APS, F ~ 4x10^{10} ph/s/eV, 50m from source in a 8.2µm x 197µm aperture at 9 keV.
- White or Pink beam provides factor 200 or more, suitable for SAXS.
Definitions: coherence lengths

\[ d_x \approx 0.64 \text{ mm} \]
\[ d_y \approx 0.021 \text{ mm} \]
For \( R \approx 50 \text{ m}, 9 \text{ keV} \)
\[ L_t \approx 0.008 \text{ mm (H)} \]
\[ 0.2 \text{ mm (V)} \]

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\( L_1 \) about 3 micron
at 9 keV with \( \frac{1}{2} \text{ eV} \) resolution
Speckle from coherent illumination

Static $\text{Fe}_3\text{Al}$ speckle pattern taken at ESRF from frozen antiphase domains.
One can also try to measure the 3D speckle and invert it through inverse methods.

Typical iterative method used to invert Coherent diffraction patterns

Reciprocal Space Constraints
- Experimental Amplitudes
- Oversampled
- Masked
- ZeroPadded

Direct Space Constraints
- Support
- Total Energy
- Mixing previous iterates
- Positivity

Chapter 3: Laser-heated CDI

- Coherent diffraction imaging experiment of microfocused laser-heated GaAs
- Preliminary results on ZnO nanocrystals
- Discussion
Time and spatial overlap

- Timing overlap done with 2 InGaAs photodiode (Judson Technology) and 16 GHz oscilloscope. One diode for laser trigger, one for x-ray and laser on sample.

Yellow: laser trigger, rise time ~100 ps
Blue: laser and weaker x-ray pulse with 2 ns delay

X5 microscope objective focuses laser to 7 µm FWHM on sample
CXD, bulk GaAs (200) 10 keV

- Results obtained with Si (111) monochromatic beam, with 7µm x 7µm unfocused coherent beam.
- Laser power 68 mW in 6 µm FWHM Gaussian laser beam.
- X-ray peak intensity reduced by factor 5. Angular split implies a peak temperature difference of 30 K, FEA results, 137 K. Reasonable due to long footprint. No obvious time dependence when changing laser-x-ray delay.
Preliminary results on ZnO capped with Ni film

• Work lead by Ross Harder, Steve Leake, on ZnO nanoparticles.
• Coated with Ni to absorb 800 nm light
• 135 mW light into 5.9 µm spot or ~6 mJ/cm².
• CDI clearly sensitive to laser on/off as speckle pattern moves in 2θ with laser on.
• Exposure time 30 sec/angle in 80 pts scan of 0.16 degrees, 86 min total acquisition. (Unfocused x-rays.)
Recent result on InAs nanowires

Phonon dynamics in nanowires

- Oscillations of straight-up InAs nanowires
- Three acoustic modes found
- Preliminary data collected at 7ID
- University of Copenhagen
ZnO (Recent Electron diffraction)

Fig. 3. Structural dynamics. Changes of the c-axis expansion, diffraction intensity, and vertical width of the (000) Bragg spot with time and at different excitation fluences. All solid lines are fits to the data (see text and 50M). (A) The intensity decrease (blue dots) and width increase (green dots) behave similarly and precede the buildup of structural expansion (red dots). In ~200 ps the former two diffraction features almost recover the original values, whereas the decay of expansion appears on a longer time (ns) scale. (B) A significantly larger c-axis expansion was obtained at higher excitation fluences. The fitted slope in the log-log plot (inset) indicates that the maximum expansion is proportional to the fluence to the power of 2.5. At all fluences, however, the expanded nanowires would return to their original structure in ~1 ns. (C and D) A more significant intensity decrease and a larger vertical width increase were observed after a stronger excitation. At the lowest fluence used, however, no appreciable diffraction changes were observed.

Average heating

From E. Dufresne et al. NIM A (in print) DOI: 10.1016/j.nima.2011.01.050

The angular shift in $2\theta$ can be used to focus the laser
Most recent results

3 sec. exposure, 50 ps sampling, 23 min scan.

From E. Dufresne et al. NIM A (in print)
DOI: 10.1016/j.nima.2011.01.050

Integrated intensity strongly modulated by timing of laser and x-rays! Although not shown, the fast oscillation are significant, clearly above noise.
Cuts through reconstructions

Figure 8.1: The reconstructed solutions at a laser to x-ray offset of 0ps, 100ps and 200ps, the translucent solid object shows the direct space amplitudes, the black bar represents the expected location of the substrate. Cut planes through the phase are shown at regular intervals along the beam direction, the crystals dimensions are 1.5µm x 0.5µm.
Discussion

• We are developing a CDI technique to image strain fields generated by pulsed laser heating which enable strain mapping with a fraction of a micron resolution.
• In GaAs, fast (100ps) time resolved effects were not observed, possibly because the sample does not have time to recover after 11 ns or illuminated volume is too large.
• ZnO results are encouraging. More systems need to be explored to determine the usefulness of oscillator driven experiments at the APS.
• With a long cavity oscillator running at 6.5 MHz, one could extend the relaxation time between two laser pulses to 153 ns and get more energy per pulse (x50). This laser is now available commercially.
5-10 years outlook

• Brighter 3rd generation are under construction or planned with short pulse options. (NSLS-II, APS-U).
• Many hard-xray FEL in construction. LCLS is being commissioned now. More in construction (XFEL, Japan, Taiwan).
• These new sources will allow to probe matter with fs resolution in and out of equilibrium.
Acknowledgements

• CDI work:
  • Steven Leake, Loren Beitra, Xiaojing Huang, Prof. Ian Robinson, University College London
  • Ross Harder, Bernhard Adams, Matthieu Chollet, Eric Landahl, Yuelin Li (APS)
  • Prof. David Reis group (S. Lee, D. Fritz) (Univ. of Michigan)