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Ultrafast probing of the x-ray-induced lattice and electron dynamics in graphite at atomic-resolution

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Disclaimer

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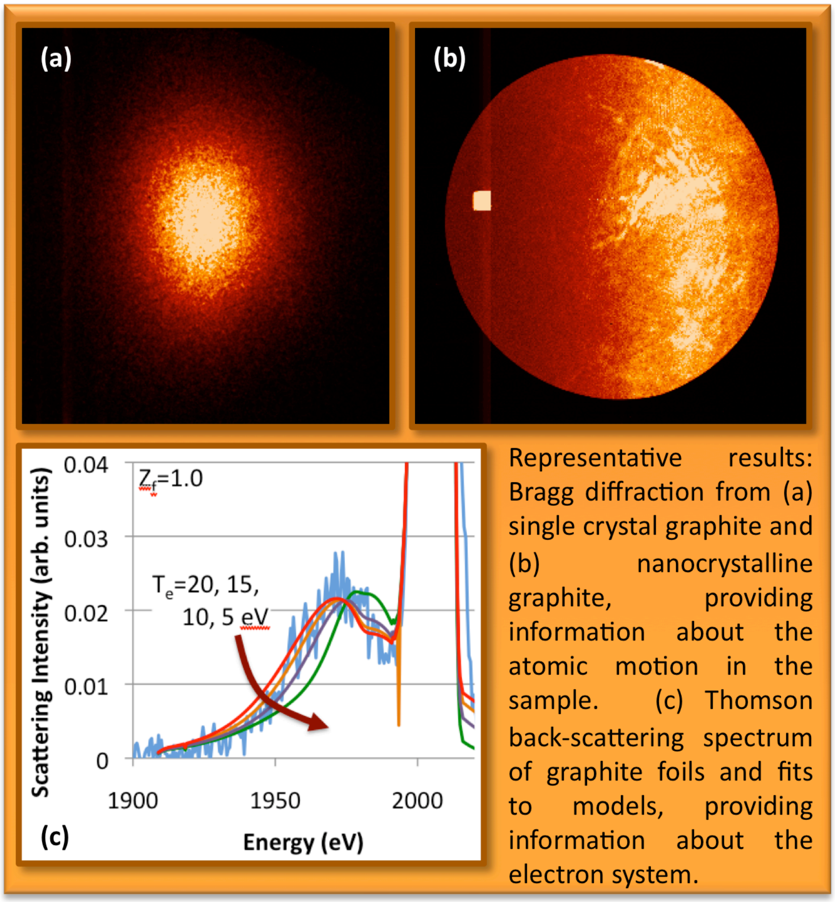
LCLS beamtime report

Ultrafast probing of the x-ray-induced lattice and electron dynamics in graphite at atomic-resolution

Run L123, July 2010, Stefan Hau-Riege (LLNL)

Objective:

We used LCLS pulses to excite thin-film and bulk graphite with various different microstructures, and probed the ultrafast ion and electron dynamics through Bragg and x-ray Thomson scattering (XRTS). We pioneered XRTS at LCLS, making this technique viable for other users.



Representative results: Bragg diffraction from (a) single crystal graphite and (b) nanocrystalline graphite, providing information about the atomic motion in the sample. (c) Thomson back-scattering spectrum of graphite foils and fits to models, providing information about the electron system.

Shift-by-shift progress:

Shift 1: Due to the rather short setup time of two days, we had to overcome in-vacuum-CCD-cooling and cooling-line-leak issues during the first shift. Toward the latter part of the shift, we collected the first Thomson scattering spectra of graphite films, demonstrating that our experimental setup works.

<12 hours break>

Shift 2: We stabilized the leak and cooling issues. We then detected a Bragg signal from the bulk HOPG samples, the authenticity of which we verified by varying the LCLS wavelength. We selected the CCD attenuator setting so that it matched the CCD dynamic range. We collected high-and low-intensity rocking curves of bulk single-crystal graphite. We collected the strontium and silicon emission spectra to calibrate our spectrometers.

<12 hours break>

Shift 3: We measured forward and backward Thomson scattering spectra of graphite films while varying the LCLS wavelength to verify the validity of the spectra. We measured the powder diffraction pattern from graphite nanocrystals and selected the matching CCD attenuator settings. We performed an LCLS gas attenuator scan to measure the Bragg reflection from single-crystal bulk graphite as a function of LCLS fluence for nominally 70 and 300 fs pulse lengths.

Shift 4: We performed YAG intensity scans to measure the beam focal position and to characterize the beam intensity distribution. We collected a detailed data set for Thomson forward and backward scattering from graphite films. We measured Thomson backward scattering spectra from nanocrystals and bulk graphite.

Shift 5: We exchanged sample (since there is no shift break during shifts #3-5). We utilized the downtime to measure LCLS x-ray spectra in the SXR beamline. We then measured the forward and backward Thomson spectra of graphite films for nominally 70, 300, and 850 fs pulse lengths. We again characterized the beam size with YAG intensity scans since the chamber z-motion did not function properly during the first scan in Shift 4.

Achievements:

Instrumentation. We demonstrated for the first time that the LCLS can be used to characterize warm-dense-matter through Bragg and x-ray Thomson scattering. The warm-dense-matter conditions were created using the LCLS beam. Representative examples of the results are shown in the Figure above. In our experiment, we utilized simultaneously both Bragg and two Thomson spectrometers.

Results. The Bragg measurements as a function of x-ray fluence and pulse length allows us to characterize the onset of atomic motion at 2 keV with the highest resolution to date. The Bragg detector was positioned in back-reflection, providing us access to scattering data with large scattering vectors (nearly $4\pi/\lambda$). We found a clear difference between the atomic dynamics for 70 and 300 fs pulses, and we are currently in the process of comparing these results to our models. The outcome of this comparison will have important consequences for ultrafast diffractive imaging, for which it is still not clear if *atomic* resolution can truly be achieved.

The backward x-ray Thomson scattering data suggests that the average graphite temperature and ionization was 10 eV and 1.0, respectively, which agrees with our models. In the forward scattering data, we observed an inelastic feature in the Thomson spectrum that our models currently do not reproduce, so there is food for thought.

We are in the process of writing these results up. Depending on if we can combine the Bragg and Thomson data or not, we plan to publish them in a single paper (e.g. Nature or Science) or as two separate papers (e.g. two Phys. Rev. Lett.). We will present the first analysis of the results at the APS Plasma Meeting in November 2010.

Problems:

We had a fantastic experience performing our experiment at the LCLS, and we are grateful to the beamline scientists and all the support personnel for enabling this experiment. A major hurdle was the very short transition time of two days, which despite all our preparations did not give us sufficient time to test the full system before the start of the beam time. We further were not able to make optimal use of the beam time since we had to exchange samples in the middle of the 36-hours shift. An additional 12-hours break could have avoided this. Finally, our experiment would have benefitted from the best possible focus, but 5 shifts do not allow performing the experiment while fine-tuning the focusing optics.

Feedback for LCLS as a user facility:

It is very sensible to put the main focus of the facility support teams on the next upcoming experiment. However, it is difficult for users at times to not get responses from experts in the various fields (which are clearly overworked) until one week before the beam time. At that point, there are so many other things for the experimental team to take care of, that it is overwhelming, whereas lots of problems could have been solved weeks in advance.

People involved during the experimental run and their role:

Name	Role at LCLS	Position	Institution
S. Hau-Riege	PI, DAQ, controls, sample prep	Scientist	LLNL
A. Graf	Overall experimental setup, DAQ	Postdoc	LLNL
T. Doeppner	DAQ	Scientist	LLNL
M. Frank	DDC cooling, vacuum	Scientist	LLNL
R. London	DAQ	Scientist	LLNL
K. Sokolowski-Tinten	DAQ, CCD software	Scientist	U Duisburg-Essen
A. Milev	sample prep	Professor	U Western Sidney
J. Krzywinski	DAQ	Scientist	SLAC
M. Messerschmidt	DAQ	Scientist	SLAC
C. Bostedt	beamline scientist, experimental setup	Scientist	SLAC
S. Schorb	experimental setup	Postdoc	U Berlin
M. Seibert	DAQ	Scientist	SLAC
D. Rolles	CAMP integration	Scientist	MPI ASG CFEL/DESY
A. Rudenko	CAMP integration	Scientist	MPI ASG CFEL/DESY
B. Rudek	CAMP integration	Postdoc	MPI ASG CFEL/DESY

Additionally, S. Glenzer and C. Fortmann (both LLNL) participated in the preparation of this experiment.

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