EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

http://193.49.43.2:8080/smis/servlet/UserUtils?start

Reports supporting requests for additional beam time

Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Structural dynamics in laser-excited Graphite close to the ablation threshold	**Experiment number**: **HS-4352**
Beamline:	**Date of experiment**: from: March 30 to: April 05, 2011	**Date of report**: 2011-06-15 *Received at ESRF:*
Shifts:	**Local contact(s)**: Michael Wulff	

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1. Summary

The aim of the work was to excite and measure laser-induced strains in graphite, and to extract the dependence of the lattice strain on excitation fluence, in light of recent studies that reported saturation in strain above 20 mJ/cm^2. The work has to a large part been successful, as we have been able to use time resolved x-ray diffraction to measure sizable strain effects in CVD-grown graphite films and to resolve the dynamics of strain with up to the ~60 ps time resolution possible with the instrument. We have also successfully carried the fluence dependence measurements and confirmed the existence of what appears to be a saturation effect in the strain above 20 mJ/cm^2. Extensive modelling is underway to confirm the preliminary analysis reported here.

2. Experimental set up

The experiments were conducted at the time-resolved studies beamline (ID09B) in the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. The silicon monochrometer of ID09B was used to produce 16 keV x-rays with ~2×10^{-4} Bandwidth. A custom built sample mount equipped with an extra stage was used in order to allow in-plane rotation. The x-ray pulse duration is ~70 ps and the x-ray pulses are synchronized to the Ti:Sapphire laser system within ~50 ps.

The sample was excited with 800 nm light at normal incidence. The experiments at ESRF were conducted at fluences ranging from 4 mJ/cm^2 up to 60 mJ/cm^2. The laser beam was focused along one direction using a cylindrical lens. The spot size at the sample was measured to be 3×0.2 mm, which is sufficiently larger than the x-ray foot print on the sample. X-ray diffraction profiles were captured using the Frelon 2D detector.

2. Sample preparation

The sample was a ~100 nm thick graphite film, coated on a 10×10 mm polished Nickel substrate. The crystallite structure of the sample is such that the graphite basal planes are parallel to the surface of the underlying substrate. In-plane, the sample consists of domains with different azimuthal orientations. The size of the domains, however, is not known. By using a thin crystalline film, one can limit the contribution of the x-ray signal to the optically pumped volume. This was a significant improvement over the use of thick graphite crystals, where x-rays penetrate deep into the bulk of the crystal even at grazing incidences due to the inherent roughness of the crystal surface.

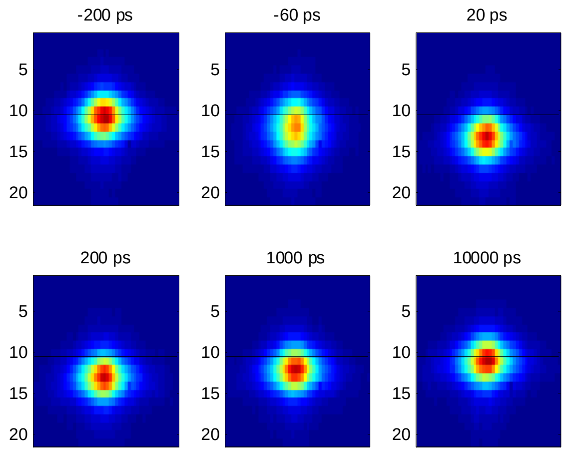

Fig 1. Selected snapshots of the 002 reflection of graphite, captured at different time delays relative to excitation with ~1.6 ps laser pulses centred at 800 nm and at an excitation fluence of 30 mJ/cm^2. The shift of the diffraction spot is indicative of *c*-axis strain.

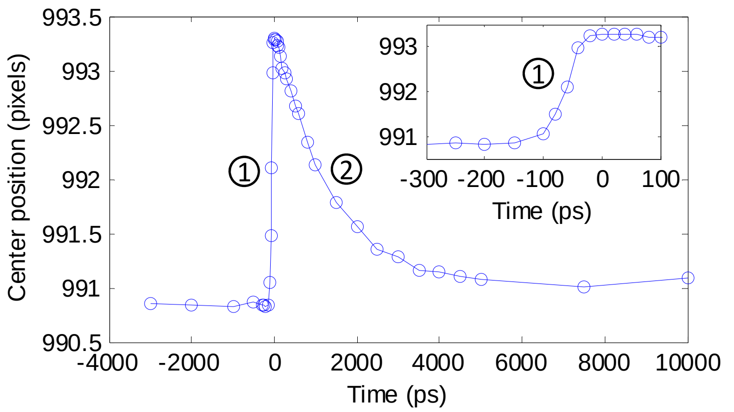

Fig 2. Dynamics of the shift in the position of the 002 reflection at 30 mJ/cm^2. The time dynamics are characterized by two features: an initial fast build-up of strain and a much slower relaxation. The inset is a blowout of the initial evolution of strain. Data in the inset was fitted to an error function. The extracted time scale of 55 ps (FWHM) represents the instrument response time, which is limited by the duration of the x-ray pulses and the jitter in the x-ray-laser synchronization scheme.

4. Results and discussion

The experiment was conducted by recording the diffraction pattern of the 002 and the 004 reflections of graphite at differnet time delays relative to the arrival of the laser exciation pulse. Fig. 1 shows sample images of the 002 reflection spot for selected time delays. The full data set contains time points in the range of -3 ns to +10 ns, with sufficient sampling to map out the dynamics of strain. Fig. 2 shows the full time dynamics of the shift in the position of the 002 reflection. The time scale of the intial fast build-up of strain is 55 ps as extracted for an errror function fit to the data in the inset of Fig. 2. As strain waves propagate at the 5000 m/s speed of sound in graphite, it is expected to take ~20 ps for strain to fully develop within the 100 nm film. Therefore, it is clear the that the time scale of the initial build-up is limited by the instrument response time rather than the actual structural dynamics. Following the build-up of strain, the sample relaxes back to room temperature on a much slower time csale, dictated by heat diffusion through the nickel substarte. Modelling of this effcet is underway in order to reproduce the measured dynamics.

Fluence dependence measurements were also carried out to extract the peak strain (defined as strain after the initial build-up) as function of fluence. The preliminary results shown in Fig 3 suggest a saturation in strain around 20 mJ/cm^2. This effect has been previously observed by Ruan *et al.*, though no explanation was offered. We believe that the saturation could be due to an increase in the optical absorption length with fluence. To explore this possibility, we will use the optical absorption depth as a free fit parameter while modelling the heating and subsequent cooling of the film. The modelling process will involve two steps: First, we will solve for the temperature of the graphite film as function of time and depth. Second, we will reconstruct the x-ray diffraction pattern of the 002 reflection by adding contributions from different atomic layers, in which at each layer a shift (strain) in the diffraction profile is introduced according to the layer's temperature. The Debye-Waller effect will also be accounted for by reducing the intensity of the reflection; though this effect is expected to be significant only at large scattering vectors (e.g. for the 004 reflection). The simulated diffraction profile will be compared to the measured one and the first step in the modelling repeated by changing the optical absorption depth until satisfactory agreement between measurement and simulation is obtained. The ultimate objective is to separate the dynamics of strain from the laser-induced changes in the optical properties of the sample.

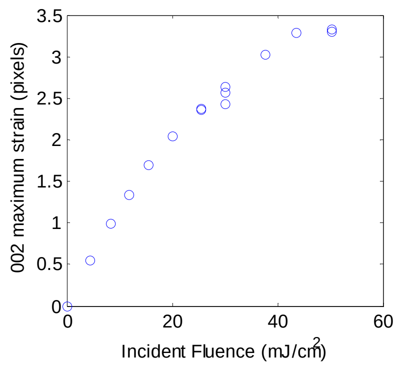

Fig 3. Strain of the 002 reflection as function of laser incident The data suggests a deviation from linearity above ~20 mJ/cm^2.