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 via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Reports supporting requests for additional beam time

Reports can 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:** Measuring the cross-plane thermal conductivity of graphite nano-layers	**Experiment number**: MA 2072
Beamline: ID09B	**Date of experiment**: from: January 23, 2014 to: January 27, 2014	**Date of report**: April 28, 2014
Shifts: 15	**Local contact(s)**: Dmitry Khakhulin, Michael Wulff	*Received at ESRF:*

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1. Summary

The experiment was aimed at measuring the dependence of cross-plane thermal conductivity of graphite on sample thickness. Strain was induced in nano-graphite films of various thicknesses by excitation with an ultrafast laser. The time dependent evolution and relaxation of strain along *c*-axis (i.e. cross-plane) of graphite was subsequently probed by following the x-ray 002 Bragg reflection. Dynamics of strain were fitted to an empirical model, from which key parameters relating to the heating and relaxation processes were extracted. Preliminary analysis suggests dependence of the relaxation timescale on both film thickness and incident fluence. A comprehensive model based on heat diffusion is needed in order to separate the competing effects that contribute to the observed dynamics, and consequently extract the thermal conductivities of the different-thickness samples.

2. Experimental setup

Measurements were conducted at the time-resolved studies beamline (ID09B) at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. The silicon monochrometer of ID09B was used to produce 15.2 keV x-rays with ~2×10^{-4} Bandwidth. The x-ray pulse duration was ~70 ps and the x-ray pulses were synchronized to the Ti:Sapphire laser system within ~50 ps.

Samples were excited with 800 nm light at normal incidence, while x-rays were incident at the 002 Bragg angle (~7^o) relative to the sample surface. The laser beam was focused along one direction using a cylindrical lens. The laser spot size at the sample was measured to be ~2.5×0.5 mm^2, which is sufficiently larger than the ~0.35×0.12 mm^2 x-ray foot print on the sample. The experiments were conducted at incident fluences in the 3.4 to 41 mJ/cm^2 range. The 002 Bragg reflection was captured on the frelon 2D detector. For each sample and excitation fluence, a series of images were recorded at different time delays in the -3 ns to 100 ns range as shown in Fig. 1.

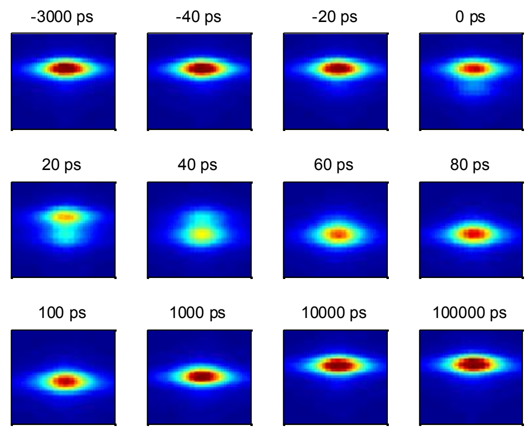

Fig 1. Selected snapshots of the 002 reflection of graphite, captured at different time delays relative to excitation at an incident fluence of 27 mJ/cm^2. The shift in the position of the diffraction spot to lower pixel values is indicative of *c*-axis expansion. Notice the "splitting" of the spot in the 20 ps and 40 ps frames. This is due to the x-ray pulse duration being larger than the timescale of thermal expansion in the film.

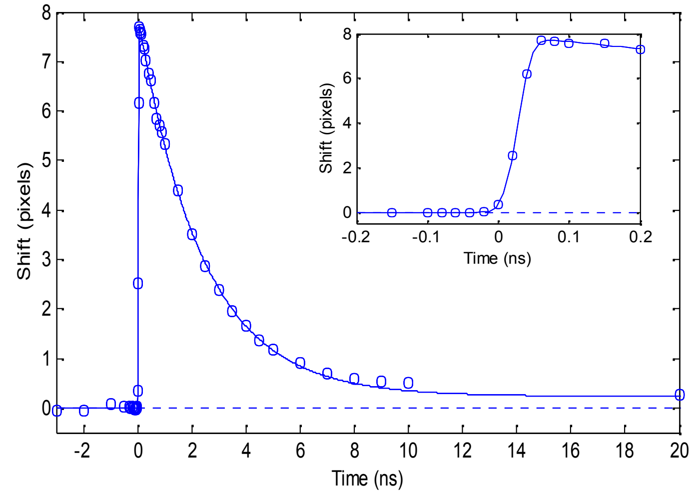

Fig 2. Dynamics of the shift in the position of the 002 reflection (open circles). The time dynamics are characterized by two features: an initial fast build-up of strain and a much slower relaxation. The inset is a blowout of the initial evolution of strain. The solid line represents a fit function comprised of an error function multiplied by an exponential decay function.

4. Results and discussion

Figure 2 shows the time dynamics of the shift in the position of the 002 reflection at an excitation fluence of 27 mJ/cm^2. The data shown in Fig. 2 is representative of all other measurements performed at different samples and over the whole range of incident fluences. The time dynamics feature an initial fast build-up of strain (~100 ps) and a relaxation to a level above room temperature occurring on a much slower timescale (~2 ns). The timescale of the initial build-up is limited by the instrument response time rather than the actual structural dynamics. Relaxation of strain, on the other hand, is dictated by heat diffusion through the nickel substrate. The data shown in Fig. 2 (open circles) were fitted to a function comprised of an error function multiplied by an exponential decay function (solid line).

Figure 3 is a summary showing the 3 key parameters obtained from the fit for measurements conducted over the whole fluence range. The top panel in Fig. 3 is the maximum amount of strain induced in the film. Strain increases with excitation, exhibiting a linear dependence as expected. Relaxation timescale (middle panel) increases with excitation level. The behaviour could be related to a change in thermal conductivity as function of sample temperature. It is known that *c*-axis thermal conductivity in graphite is dominated by phonons and exhibits a $1/T$ dependence (*K. Sun et al., Superlattices and Microstuctures 45 (2009) 60-64*). Lastly, the bottom panel shows the residual strain left in the graphite film. It is interesting that the film does not relax back to room temperature strain-level within the 100 ns time frame of the measurements. The residual strain suggests heating of the underlying nickel substrate. The next step of the analysis will focus on modeling heat diffusion through the film-nickel structure using a 1-D model akin to what has been previously employed (*M. Harb et al. Applied Physics Letters 101 (2012) 233108*). The $1/T$ dependence of thermal conductivity will be included in the model. The objective will be to extract the room temperature thermal conductivity for the various thickness samples and investigate the origin of thickness dependence which has been proposed by theory (*K. Sun et al., Superlattices and Microstuctures 45 (2009) 60-64*).

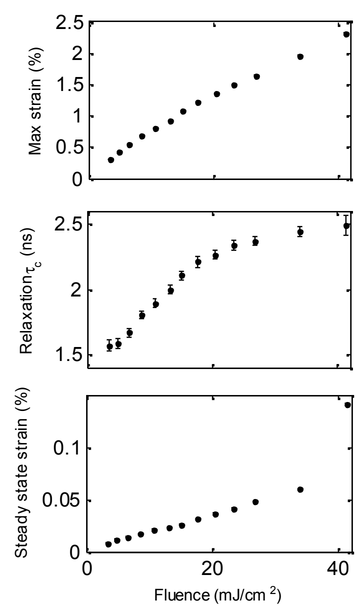

Fig 3. Fluence dependence of key parameters extracted from the fit to the strain dynamics.