ESRF	**Experiment title:** The structure of Liquid Carbon	**Experiment number**: **HD- 428**
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1. Summary

The aim of the work was to implement a new method of investigating liquids which are formed under extreme conditions and to investigate the structure of liquid carbon. The work has to a large part been very successful. We have

1. Demonstrated the technique on InSb which is a heavy scattering material commercially available with mirror-like quality ***(Manuscript in preparation)***
2. Used time resolved x-ray diffraction to investigate the melting dynamics in amorphous carbon (a-C) ***(Manuscript in preparation)***
3. Laser-induced X-ray reflectivity changes in a thin film ***(Manuscript in preparation)***
4. Developed strategies for performing measurements on liquid carbon. In order to be successful we need crystalline samples of high quality, higher flux (4-bunch or hybrid mode) and thicker films than in the present study.

2. Experimental conditions

The experiments were conducted at the time-resolved studies beamline (ID09B) in the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. The multilayer monochrometer of ID09B was used to produce 18 keV x-rays with ~1.6% Bandwidth. X-ray slits placed before the sample were used to cut down the x-ray beam size to 20×200 μm. A custom built sample mount equipped with two extra stages was used in order to allow in-plane rotation and in-plane translation along the x-ray beam direction. The x-ray pulse duration is ~50 ps and the x-ray pulses are synchronized to the Ti:Sapphire laser system within ~100 ps.

3. Sample preparation

The main sample was a 46 nm thick a-C layer, coated on a 20×10 mm silicon substrate. The density of the a-C film is 2.0 g/cm^3 as characterized by x-ray reflectivity measurements, and its sp^3/sp^2 ratio is 0.2. InSb was also studied.

4. Excitation conditions

The sample was excited with 800 nm light at normal incidence. The melting fluence of the sample was calculated to be 125 mJ/cm^2 using the thermophysical properties of bulk graphite. The damage fluence was experimentally measured to be ~100 mJ/cm^2 by visual inspection of the sample after few seconds of excitation at 1 kHz repetition rate. It was observed that ~10% above the measured damage threshold buzzing noises were heard while ablating the sample. The experiments at ESRF were conducted at fluences ranging from 40 mJ/cm^2 up to 100 mJ/cm^2. At fluences higher than 100 mJ/cm^2, the a-C film was completely damaged in few seconds of excitation making it not possible to conduct the experiment by repetitively melting and regrowing of a-C on the same sample position.

5. Operating in grazing incidence and Time-resolved X-ray reflectivity from laser excited amorphous carbon

Carbon is a light element that does not efficiently scatter x-rays. In order to match the x-ray penetration depth and the optical skin depth in carbon, one has to go to extreme (near critical angle) grazing incidences. At a grazing angle of 0.09635°, the attenuation length of ~30 nm is comparable to the optical skin depth of 800 nm light. The experimental challenge, however, is highlighted in the fact that a change of 0.002° in angle of incidence results in a 100-fold change in the attenuation length, from 30 nm to 3 μm. ***In order to overcome this difficulty we have developed a technique for properly setting the grazing angle with accuracy better than 0.002°***. The grazing angle was set by measuring the x-ray reflectivity of the sample as a function of angle similar to Fig. 1a. Below the critical angle the reflectivity of the sample is 1, while above the critical angle the thin film interference effect is apparent from the interference fringes. The data was fitted using the well known thin film on a substrate interference model with one of the fitting parameters being the offset in stage position. The extracted offset was then used to calibrate the stage position yielding the absolute values of the grazing angle. The x-ray reflectivity of the sample as a function of grazing angle was also measured at various time delays and at excitation conditions comparable to the time resolved x-ray diffraction experiment. ***This was used to determine the density and thickness of the a-C film as function of time. This is presently prepared to be reported.***

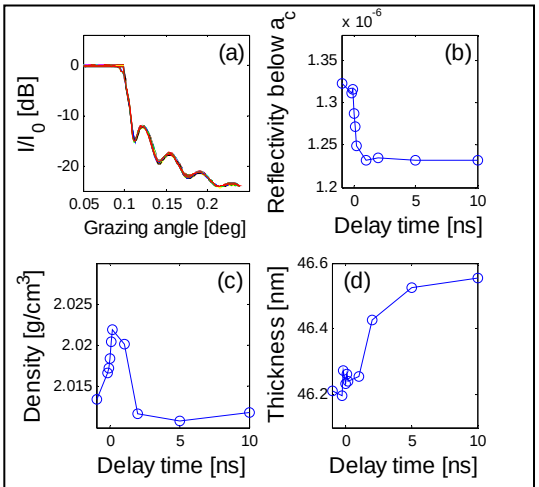


Fig 1. (a) Intensity of the specular reflection from a-C film as function of angle while excited at 90 mJ/cm^2. The multiple curves represent different delay points. Dynamics of the reflected intensity below the critical angle (b), film density (c) and film thickness (d) as extracted from the fit to the thin film model.

6. Time resolved x-ray diffraction from flash-molten InSb

Because of its fast dynamics, non-thermal melting of InSb was used as a time diagnostic experiment to determine "time zero" (the instance at which x-rays and laser arrive simultaneously at the sample). In this experiment, the InSb sample was excited at 28 mJ/cm^2. Fig. 2 shows the intensity profile differences of InSb for selected time delays. At positive delays, the liquid diffraction profile of InSb emerges. The data is of sufficiently high quality to determine radial distribution functions. A methodology paper is being prepared.

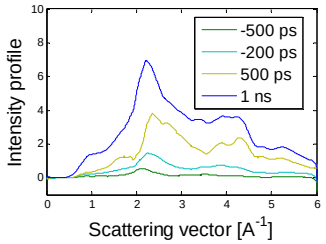
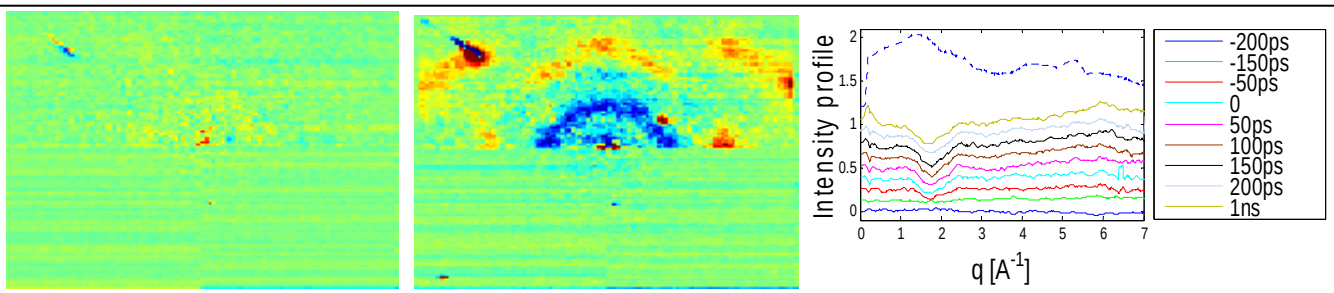


Fig 2. Scattering from laser excited InSb at different times. Liquid InSb is formed following excitation.

7. Time resolved x-ray diffraction from laser-excited amorphous carbon

We present the results of the experiment carried out at an excitation fluence of 80 mJ/cm^2. Fig. 3 shows selected diffraction image differences constructed by subtracting a reference image representing negative delays ($t < 0$) from images taken at a specific time delay (t). The rightmost panel displays the intensity profile differences for the whole data set. One can clearly see the onset of the dynamics in the -50 ps intensity profile difference, confirming the validity of the timing. Furthermore, there are two distinct features in these dynamics: increase in the intensity (red) of the different diffraction spots corresponding to the Si substrate, and decrease in the intensity (blue) of the a-C carbon diffraction ring. Modelling is underway in order to decide if the drop in intensity is due to bond breaking or due to heating. Since the signal is constant over 5 ns, where the temperature changes from 5000 to 100 degrees above room temperature, we believe that we directly observe the fraction of broken sp2 bonds to be 1%.



-200 ps ***200 ps*** ***Radial lineouts***

Fig 3.Laser-excited amorphous carbon (a-C). Selected diffraction image differences taken at a specific time delay and the intensity profile difference for the whole data set.