ESRF	**Experiment title:** NUCLEATION MECHANISM FOR THE DIRECT LIGHT-DRIVEN TRANSITION FROM GRAPHITE TO NANODIAMOND	**Experiment number**: MA 3035
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Report:

The experiment was aimed at investigating the nucleation mechanism for the direct light driven transition from graphite to nanodiamond. We used high-order pyrolythic graphite HOPG flakes and natural graphite flakes as the samples. This requires single shot operation mode for the experiment. Therefore we worked in the high-flux 4-bunch mode. Unforturnately, even in this high-flux mode it was not possible to see convincing evidence for diamond formation from the dynamic measurement.

However, we were able to see nano-diamonds from pre-exposed samples and in dynamic measurements we were able to observed large strain wave which could give rise to diamond formation.

During the experimental run, we acquired static X-ray diffraction data from a HOPG sample that had been pre-exposed by a femtosecond laser. The data is shown in figure 1. The observed diffraction spots (identified to be cubic diamond) are marked with black circles.

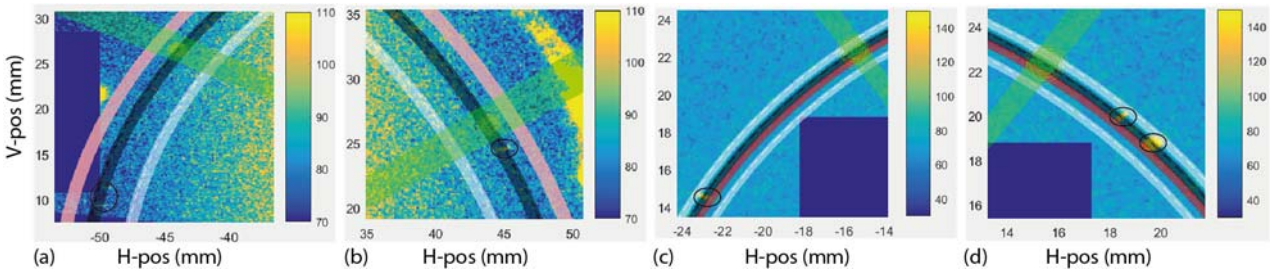

Figure 1 X-ray diffraction images taken from pre-exposed HOPG. 1(a) and 1(b) give the diffraction spots on cubic diamond (2 2 0) ring with q value 4.984 Å^{-1}, and 1(c) and 1(d) show the diffraction spots on cubic diamond (1 1 1) ring with q value 3.052 Å^{-1}. The dash lines correspond to the tabulated q values of cubic diamond (CD), hexagonal graphite (HG) and rhombohedral graphite (RG). The shade color is the error bar, black for CD, white for HG, red for RG, and greeen for angular error bar. The error bar for q values was calculated by the convolution of X-ray bandwidth, footprint, penetration depth and sample flatness. The angular error bar was calculated by sample flatness.

Then we started the time-resolved measurements on natural graphite with the aim of following the dynamics in nanodiamond formation. One potential mechanism of graphite-diamond transition involves restacking to rhombohedral graphite, follwed by shock-wave compression and subsequent sp^3 bond formation[2]. From the carbon phase diagram, the pressure, which is needed to induce graphite-diamond transition, varies from 2 GPa to 10 GPa depending on temperature[3]. The dynamical measurements were excuted in single shot operation mode. The pulse duration of laser is 1.2 ps, and the FWHM of X-ray is 100 ps. The wavelength of laser is 800 nm, and the X-ray energy is 15 keV. The incident angle of X-ray was set to be 12 degree, and we could see the non-coplanar (1 0 3) reflection of graphite by optimizing the azimuth angle. The data was taken at different time delays with different fluences.

To investigate the pressure generated by the laser, we calculated a difference image by subtracting an image when the laser was on and a refereence image which the laser was off. Figure 2(a) and 2(b) show the expertimental (Fig. 2a) and simulated (Fig. 2b) difference images. The experimental data was taken at 100 ps time delay with a fluence of 16.3 J/cm^2.

The simulation is based on a diffraction code which enables simulattion of non-coplanar diffraction for a pre-defined strain profile. The simulation result has good agreement with the experimental data. The simulation is based on a strain profile which can be seen in figure 2(c). From figure 2(c), we can see that the maximum strain in the sample is -12%, which means the hexagonal graphite lattice is compressed by 12%. According to the bulk modulus 36.4 GPa[4], this 12% compression strain corresponds to 4.4 GPa. This pressure is in the range for the graphite-diamond phase transition.

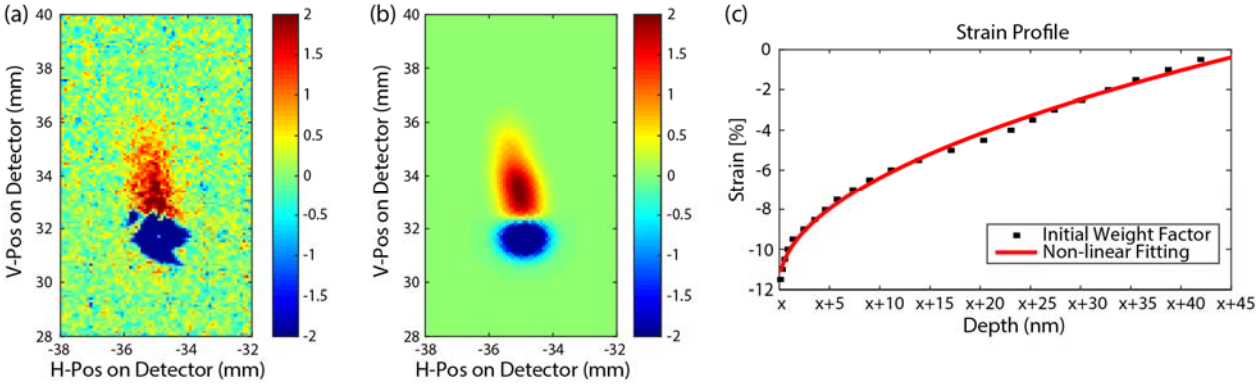

Figure 2 (a) Experimental difference image of laser on and laser off at 100 ps time delay for fluence 16.3 J/cm^2. (b) Simulation difference image from non-coplanar diffraction code. (c) Strain profile used in the simulations

The lack of dymic data on the graphite-to-diamond transition may be due to the fact that the laser conditions at ESRF differed from our pre-exposed sample or that the flux required is even higher than what could be achieved in 4-bunch mode at ID09.

The the data is currently being analyzed further. In particular we are carrying out simulations which may explain the high compressive strain. Theoretical modelling using the ESTHER code is ongoing to interpret the strain profile[5]. Our initial analysis will concentrate on the strain evolution and the fluence dependence of the strain profile. We believe this will help us to understand the mechanism of strain generation and propagation in the crystal. Plans for publication of this work are in progress.

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