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# Investigation of the solid–liquid phase transition of carbon at 150 GPa with spectrally resolved X-ray scattering

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ABSTRACT

We have resolved the solid–liquid phase transition of carbon at pressures around 150 GPa. High-pressure samples of different temperatures were created by laser-driven shock compression of graphite and varying the initial density from 1.30 g/cm$^3$ to 2.25 g/cm$^3$. In this way, temperatures from 5700 K to 14,500 K could be achieved for relatively constant pressure according to hydrodynamic simulations. From measuring the elastic X-ray scattering intensity of vanadium K-alpha radiation at 4.95 keV at a scattering angle of 126°, which is very sensitive to the solid–liquid transition, we can determine whether the sample had transitioned to the fluid phase. We find that samples of initial density 1.3 g/cm$^3$ and 1.85 g/cm$^3$ are liquid in the compressed states, whereas samples close to the ideal graphite crystal density of 2.25 g/cm$^3$ remain solid, probably in a diamond-like state.



## 1. Introduction

A deeper understanding of carbon under extreme conditions is highly relevant to various fields of science [1]. Prominent examples are the internal structure and evolution of the carbon-bearing ice giants in our solar system [2–4], extrasolar carbon planets [5] or the outer layers of white dwarfs [6]. In these systems, warm dense carbon, i.e. carbon with temperatures from 5000 K to 100,000 K at solid density or above, is supposed to be highly abundant and models remain challenging since the complex underlying physics of warm dense matter (WDM) is still poorly understood [7]. In the low-temperature regime of WDM, the available energy from pressure and temperature are on the order of chemical bonding and thus, remaining bonds can strongly influence the microscopic structure [8]. In the laboratory, WDM states are traversed in every process where a solid density sample is rapidly heated to a plasma state. Within the Inertial Confinement Fusion experiments presently ongoing at the National Ignition Facility [9], carbon is one possible ablator material which is currently being investigated [10]. Here, the high-pressure solid–liquid transition of carbon is of special interest, since re-freezing of the ablator layer can occur after the first shock, leading to density fluctuations which can seed hydrodynamic instabilities. Therefore, an improved knowledge of the microscopic structure and the ion–ion potential close to the melting line is needed for accurate models of these experiments. Classical measurements of the shock Hugoniot, giving the macroscopic quantities pressure and density, can only give hints of the underlying microscopic properties [11,12]. Whereas for high-pressure solid–solid transitions, fast X-ray diffraction has usually been the method of choice [13], liquid structure can hardly be resolved in a single event. This is especially true for low-Z materials, which have a small elastic scattering amplitude. Spectrally resolved X-ray scattering, however, has been proven to be capable of directly determining the microscopic structure of short-lived warm dense matter samples [14,15], recently revealing the importance of short-time chemical bonds in liquid carbon around 100 GPa pressure [8]. Here we show a further development of these experiments using graphite samples of different initial density. In this way, a broader variety of final states can be achieved, in particular creating states

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of very similar pressure at different temperatures for a given drive [16]. Our results show that this method is capable of resolving the solid–liquid transition of carbon at relatively constant pressure.

## 2. Experiment setup

The experiments were performed at Target Area West of the Central Laser Facility based at STFC Rutherford Appleton Laboratory, UK. A sketch of the experiment setup is shown in Fig. 1. Four beams of the VULCAN laser system were focused onto graphite samples with a spot diameter of 700 μm, an accumulated energy of 250 J at a wavelength of 527 nm and 6–7 ns pulse duration. These laser parameters were used for all runs. Random phase plates were implemented to ensure planar and homogeneous shock waves were driven into the ~ 100 μm thick samples. Three graphite types varying in initial density and micro-structure were used during the experiments: Flexible Graphite (FG), 1.3 g/cm$^3$, which is produced by breaking bonds between the graphene layers several times and compressing the exfoliated graphite into form [17]. Rigid Graphite (RG), 1.84 g/cm$^3$, is made out of graphite powder under isostatic pressure and heat. Highly Ordered Pyrolytic Graphite (HOPG), 2.25 g/cm$^3$, has a high degree of preferred crystallite orientation, usually with the c-axis perpendicular to the sample surface and an angular variation of the crystallite orientation of less than one degree. For estimation of the pressure reached in the shock-compressed graphite samples, the time of the shock release at the rear surface of the carbon sample was measured by an optical streak camera [18].

The microscopic structure of the shock-compressed material was investigated by spectrally resolved X-ray scattering [19]. The short pulse configuration of the VULCAN laser system (150 J on target, 10–20 ps, 1053 nm, 50–100 μm focal spot) was used to illuminate a 10 μm vanadium foil mounted at a distance of 1.7 mm from the rear surface of the carbon sample. The high-intensity laser-plasma interaction produces hot electrons, which penetrate the cold vanadium and create K-shell holes resulting in emission of strong K-alpha line radiation at 4.95 keV [20]. The temporal duration of the emitted X-ray pulse is of the same order as the laser pulse. The emitted radiation spectrum was measured by two HOPG spectrometers: one monitoring the radiation source directly and another recording the X-ray spectrum after transmission through the carbon sample. The vanadium foil was placed inside a gold cone with a 500 μm pinhole towards the graphite target for collimating the X-rays and thus, defining the scattering geometry. The gold cone also serves for shielding the direct line-of-sight towards the X-ray scattering spectrometer, which consists of a cylindrically curved HOPG crystal in von Hamos geometry and image plates as detector. The scattering angle was chosen to be 126°, which is very sensitive to the solid–liquid transition for the applied photon energy of 4.95 keV [8]. Moreover, at this scattering angle regime, there are no strong Bragg reflections expected, which could interfere with the elastic scattering. This is true for cold graphite (see Fig. 2) as well as for possible solid compressed diamond states [8].

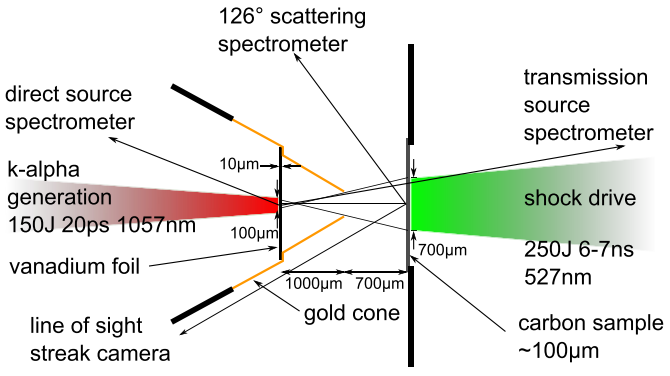


**Fig. 1.** Sketch of the experimental setup: The four shock-driving lasers incident on the graphite sample. A laser pulse with 20 ps pulse length illuminates a vanadium foil to generate the K-alpha radiation at 4.952 keV. The radiation is collimated by a pinhole to restrict the measurement to the shocked area. The X-ray source, transmission and scattered radiation are monitored and spectrally resolved by HOPG spectrometers.

## 3. Spectrally resolved X-ray scattering

The scattering of X-ray radiation from matter is generally dominated by the electrons. For small momentum transfers, where the energy of the scattered photon is nearly unchanged, the magnitude of the scattering vector $\mathbf{k} = \mathbf{k_i} - \mathbf{k_s}$, which is the difference of the incident wave vector $\mathbf{k_i}$ and the scattered wave vector $\mathbf{k_s}$, can be approximated as [21].

$$k = |\mathbf{k}| = \frac{2\omega_i}{c}\sin(\theta/2), \tag{1}$$

where $\omega_i$ is the frequency of the incident light and $\theta$ denotes the scattering angle. For our experiment, the magnitude of the scattering vector is $k = 4.47$ Å$^{-1}$. The power spectrum of the scattered radiation per sample atom is given by the double-differential scattering cross section [22]:

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = r_0^2\frac{\omega_s}{\omega_i}\frac{1}{2}\left[1+\cos^2\theta\right]S(k,\omega). \tag{2}$$

Here, $r_0$ is the classical electron radius, $\omega_s$ the frequency of the scattered radiation, $\omega = \omega_i - \omega_s$ is the frequency shift and $S(k,\omega)$ denotes the total electron structure factor which contains all the microscopic information of the system and can be expressed in terms of the electron density fluctuations [22]:

$$S(k,\omega) = \frac{1}{2\pi N}\int dt' \langle n_e(k,t) n_e^*(k,t+t')\rangle e^{i\omega t'}, \tag{3}$$

with $n_e(k,t)$ denoting the Fourier transform of the electron number density $n_e(r,t)$ in space and $N$ the number of sample atoms. For warm dense matter conditions, which usually involve partial ionization, the different contributions of bound and free electrons need

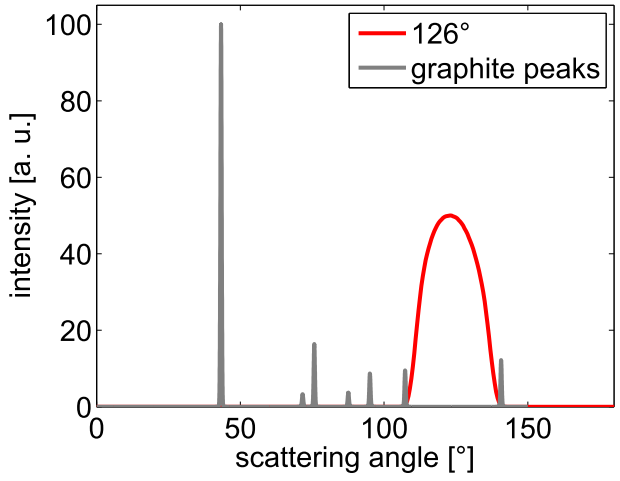


**Fig. 2.** Positions of the Bragg-reflections from polycrystalline graphite and the angle range covered by the scattering spectrometer at 126°.