High Energy Density Physics 8 (2012) 46–49

Contents lists available at SciVerse ScienceDirect

High Energy Density Physics

journal homepage: www.elsevier.com/locate/hedp

X-ray Thomson scattering on shocked graphite

D. Kraus [a,*], A. Otten [a], A. Frank [a], V. Bagnoud [b], A. Blažević [b], D.O. Gericke [c], G. Gregori [d], A. Ortner [a,b], G. Schaumann [a], D. Schumacher [c], J. Vorberger [c], F. Wagner [a], K. Wünsch [c], M. Roth [a]

[a] Institut für Kernphysik, Technische Universität Darmstadt, Schlossgartenstrasse 9, 64289 Darmstadt, Germany
[b] GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstrasse 1, 64291 Darmstadt, Germany
[c] Centre for Fusion, Space and Astrophysics, Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom
[d] Department of Physics, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom

ARTICLE INFO

Article history:
Received 20 November 2011
Accepted 21 November 2011
Available online 30 November 2011

Keywords:
Warm dense matter
Liquid carbon
Hexagonal diamond
X-ray Thomson scattering
Laser-driven shock wave

ABSTRACT

We present measurements of the changes in the microscopic structure of graphite in a laser-driven shock experiment with X-ray scattering. Laser radiation with intensities of $\sim 2 \times 10^{13}$ W/cm^2 compressed the carbon samples by a factor of two reaching pressures of ~90 GPa. Due to the change of the crystalline structure the scattered signals of the probe radiation were modified significantly in intensity and spectral composition compared to the scattering on cold samples. It is shown that the elastic scattering on tightly bound electrons increases strongly due to the phase transition whereas the inelastic scattering on weakly bound electrons remains nearly unchanged for the chosen geometry.

1. Introduction

Understanding the properties of warm dense matter, that is matter with temperatures of 0.5–10 eV and densities around solid density, is a highly evolving field of research because of its importance for the physics of giant planets [1,2] and inertial confinement fusion [3]. In particular, carbon is an interesting material for warm dense matter studies, as it is accessible experimentally since carbon samples can easily be manufactured and handled in the laboratory. Due to its low number of electrons, a number of theoretical and numerical techniques, including ab initio simulations, allow for the description of its properties within the computational resources available today. Additionally, the solid–liquid phase transition of carbon is in the warm dense matter regime and may play a major role in the physics of ice giants like Neptune and Uranus [4,5] and white dwarfs [6]. This transition is poorly understood so far and further investigation is needed [7].

Shock experiments are a common tool to explore the properties of matter at high pressures. Carbon is a very useful material for EOS studies with shocks because its initial density can be varied by using different types of amorphous, polycrystalline and monocrystalline carbon. There are many shock experiments using high explosives which observed a phase transition on different graphite Hugoniots starting at ~20 GPa [8,9]. Hexagonal diamond [10] is mostly considered to be the final state of this phase transition which is expected to happen on a very fast timescale of few ns [11] or faster [12]. Following the graphite Hugoniot to larger pressures, there are no reliable data of the subsequent transition to the liquid and corresponding theories are not in agreement [13]. Pressures from ~80 [8] to ≥300 GPa [14] have been proposed for the melting threshold on the Hugoniot of graphite with ideal crystal density of 2.26 g/cm^3. Starting with polycrystalline samples of lower density this threshold is expected to be at lower pressures as lower density Hugoniots result in higher temperatures for a given pressure. The only experimental data of the high pressure carbon solid–liquid phase transition come from shock experiments on diamond [15–17] which monitor the transition at ~600 GPa on the diamond Hugoniot. The transparency of diamond for visible light is used to dynamically follow the propagation of the shock inside samples. This gives a very accurate measurement of the Hugoniot curve. However, the solid–liquid phase transition only results in a very small change of the Hugoniot's slope, which is very difficult to resolve, even for very high precision measurements of shock and particle velocity [16]. Furthermore, the temperature of the propagating shock front is determined with pyrometry and reflectivity measurements [17]. However, these methods are not applicable for optically opaque materials like graphite. In fact, the solid–liquid phase transition on the graphite Hugoniot has not been measured reliably so far. An alternative is the use of X-rays which are able to

* Corresponding author. Tel.: +49 6159 71 1443; fax: +49 6159 71 2992.
E-mail address: d.kraus@gsi.de (D. Kraus).

1574-1818/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.hedp.2011.11.011

access the processes inside the sample. In comparison to radiography X-ray scattering cannot only measure the propagation of the shock but also the microscopic structure inside the sample [18–20]. Thus, strong changes in the structure due to the phase transitions induced by shock can be measured directly. In this paper we present the measurement of the microscopic structure change of graphite in a laser-driven shock with X-ray scattering.

2. X-ray scattering on crystalline and liquid carbon

Thomson scattering is the scattering of electromagnetic radiation on electrons where the energy of the incident photons is much smaller than the rest mass of the electron. For unpolarized radiation the scattered power dP per solid angle dΩ and frequency dω_s is then [18]

$$\frac{\mathrm{d}^2P}{\mathrm{d}\Omega \mathrm{d}\omega_s} = I_0 r_0^2 \frac{1}{2}\left(1+\cos^2\theta\right) S(k,\omega) \tag{1}$$

I_0 is the intensity of the radiation source on the sample, $r_0 = (e^2/4\pi\varepsilon_0 m_e c^2)$ the classical electron radius, θ the scattering angle. $S(k, \omega)$ is the spectral density function or dynamic structure factor of the electrons. $\hbar k$ and $\hbar\omega$ are the momentum and the energy transferred to the electron during the scattering process, respectively. $S(k, w)$ is defined as the Fourier transform of the electron density–density correlation function in space and time. Thus, the dynamic structure factor contains all the microscopic information of the sample material. Following the approach of Chihara [21,22] the structure factor can be decomposed in three parts: elastic scattering on tightly bound electrons, inelastic scattering on weakly bound electrons and, if a plasma is present, inelastic scattering on free electrons. For a cold crystal, like graphite, scattering is dominated by the elastic (or coherent) part if a Bragg condition is fulfilled. If the scattering angle is chosen the way that no Bragg condition can be implemented, the scattering signal will be dominated by scattering on weakly bound electrons. In general, the static electron structure factor $S(k) = \int S(k,\omega)\mathrm{d}\omega$ for this case can be written as [23,24]:

$$S(k) = |f(k)|^2 S_{\mathrm{ii}}(k) + Z_{\mathrm{wb}}\left[1-\left(\frac{f(k)}{Z}\right)^2\right]. \tag{2}$$

$f(k)$ is the atomic form factor, $S_{\mathrm{ii}}(k)$ the static ion–ion structure factor which contains the ion correlations. Z ist the number of the bound and Z_{wb} the number of, compared to $\hbar\omega$, weakly bound electrons per atom. During the solid–liquid phase transition $S_{\mathrm{ii}}(k)$ changes dramatically from a function with narrow spikes at the position of the Bragg peaks to a more continuous function in the liquid (see Fig. 1). Because the contribution of the weakly bound electrons can be estimated to be nearly equal for solid and liquid, X-ray scattering is able to monitor the phase transition if the chosen scattering angle results in a large difference of $S_{\mathrm{ii}}(k)$ for the two phases. This method was first applied for the carbon solid–liquid phase transition at lower pressure in a proof-of-principle experiment where the heating was realized by laser-accelerated protons [25]. However, in this experiment only the relative increase of the total scattering signal was measured. It was not possible to distinguish between the elastic and inelastic features. In the experiment presented in this paper, we have obtained frequency-resolved scattering spectra which allow to study the evolution of both the elastic and the inelastic features separately.

3. Experiment

A scheme of the scattering experiment, which was performed at the experimental area Z6 of the GSI Helmholtz Zentrum für

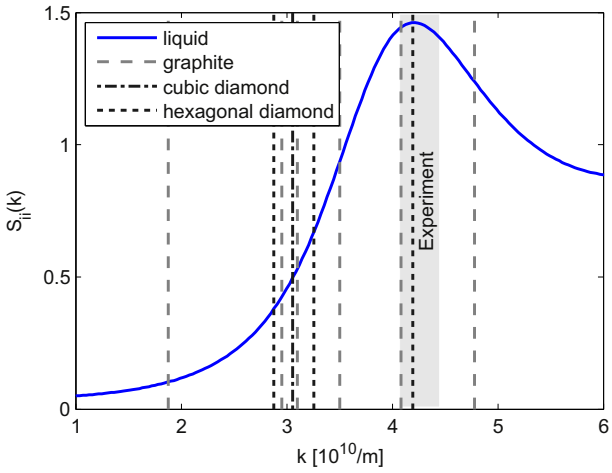

Fig. 1. Diagram of the expected ion–ion structure factor for liquid carbon with a density of 4 g/cm^3, compared with the relevant crystal phases of carbon. The positions of the strongest Bragg maxima of graphite, cubic and hexagonal diamond are represented by vertical lines. The gray shaded area shows the region of k-values covered by the scattering spectrometer in the experiment.

Schwerionenforschung GmbH in Darmstadt, Germany, can be seen in Fig. 2. The laser system nhelix [26], which delivered 65 J in 9 ns (FWHM) at a wavelength of 1064 nm was used to drive a shock into the sample. The spot size of 200 μm resulted in driving intensities of $\sim 2\times10^{13}$ W/cm^2. For X-ray generation the long pulse of the high energy laser system PHELIX [27] (150 J, 1 ns, 527 nm) was focused on a titanium foil to a spot size of 100 μm. The resulting intensities of $\sim 10^{15}$ W/cm^2 are very efficient to drive titanium helium-α radiation at 4.75 keV [18] which was used as X-ray source in the experiment. In fact, laser to X-ray conversion efficiencies of up to 5×10^{-3} were achieved with PHELIX. The scattering angle was chosen to be $(126\pm10)^\circ$ which ensures that nearly no Bragg reflection contributes to the scattering signal of cold samples. At the same time the maximum of the ion–ion structure factor $S_{\mathrm{ii}}(k)$ of liquid carbon is expected to be found around that angle for a density of 4.0 g/cm^3 [28]. As a pre-experiment the thermodynamic state in the shock wave was characterized by a classical measurement of shock velocity and particle velocity. The shock speed was measured via self emission of the shock breakout with a visible streak camera (see Fig. 3). A 500 ps laser pulse was used as timing fiducial. Additionally, the particle velocity was measured via the expansion of the shock release with a multi-frame shadowgraphy instrument [29]. The latter diagnostics takes four pictures with a constant time interval of 2 ns and a time resolution of 500 ps.

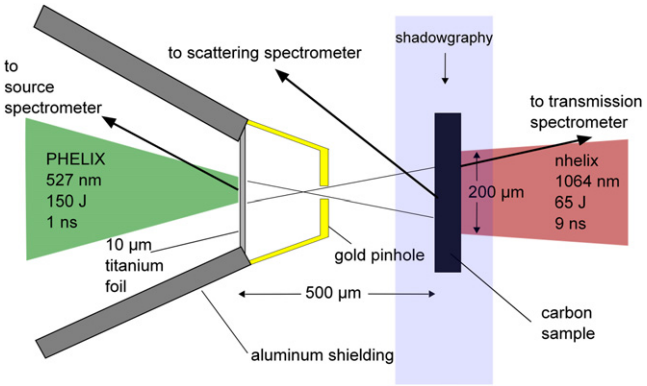

Fig. 2. Schematic of the experimental setup of the scattering experiment.