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研究代表者
Ｈａｒｔｌｅｙ　Ｎｉｃｈｏｌａｓ（hartley, Nicholas）

大阪大学・先導的学際研究機構・特任助教（常勤）

研究者番号：１０７７０９０１

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研究成果の概要（和文）：高温・高圧の物質についてX線回折を用いた研究である。最近まで、X線回折を低原子番号の液状物質に適用するには、物質の散乱断面積が小さすぎて不可能であったが、X線自由電子レーザーの十分な輝度を利用し、世界で初めて低原子番号の原子からなる物質系からのX線回折の観測に成功した。また、極端に非平衡な物質系を可視光や赤外レーザーおよびX線レーザーによる加熱を用い高温・高圧の物質状態について研究を行った。

研究成果の概要（英文）：Matter at high temperatures and pressures has been widely studied, often using X-ray diffraction, which can directly probe the ion structure of systems. Until recently, this diagnostic was not feasible with low-Z liquid materials, due to the weak scattering cross-section. However, with the increased brilliance available at X-ray Free Electron Lasers, we have succeeded in observing light elements in systems without strong structural order using diffraction for the first time. We also consider novel approaches to studying highly non-equilibrium systems at XFELs, using heating with optical lasers and the X-rays themselves, with SACLA's two-color mode.

研究分野：High Energy Density Physics

キーワード：XFEL,　Carbon,　SACLA

１．研究開始当初の背景

Materials containing low-Z elements at high pressures and temperatures, both as elements or parts of compounds, are relevant to a variety of scientific fields, including planetary interiors, geophysics and inertial confinement fusion research. These often include hydrocarbons which, being formed from some of the most abundant elements in the universe, are commonly found as a major constituent of `icy giant' planets. In the form of plastics, hydrocarbons are also used as ablator materials in high energy density (HED) research, and to drive the compression in inertial confinement fusion implosions.

At high pressures and temperatures the behavior of materials is generally complex, because long range order is lost as the thermal energy is comparable to the binding energy. It may then be energetically favorable for the mixture of various atom types to demix into separate phases with different atomic ratios, as with hydrogen and helium in giant planet interiors, or the formation of diamonds within icy giants. This means that describing the behavior of mixtures requires detailed knowledge of how both the mixture and the individual components will behave.

Within the gas giants – Jupiter and Saturn – demixing of hydrogen and helium, believed to be driven by the metallization of hydrogen, leads to reduced heat transport with significant effects on the planetary evolution. Similar processes inside the icy giants - Uranus and Neptune, along with increasing numbers of exoplanets - would again have strong ramifications for models of their formation and evolution. Experimental results suggest that solid carbon precipitates within the icy mantle, leading to a carbon layer around the core. Being able to confirm whether this layer is solid or liquid, by refining measurements for diamond melting, would also be important for planetary modeling.

Inside planets, matter is gravitationally contained; to recreate such conditions here on Earth, other approaches must be used. Although static compression techniques are able to cover an increasingly large region of pressure-temperature space, the highest pressures can only be reached through dynamic compression techniques. These can include magnetically driven flier plates or laser driven shock compression. These techniques create transient high pressure and temperature states, and so the sample must be studied within the confinement time, which is on the order of ns for laser-driven shock compression

２．研究の目的

The aim of this research was to study the behavior of carbon and carbon-containing compounds at conditions of high temperature and pressure.

In the case of pure carbon, the aim was to study the high-pressure melting line of diamond, which remains relatively uncertain, and looking for new crystalline phases by heating the electrons to cause additional ionization. For mixtures, primarily hydrocarbons, this was to look for the separation of the material into carbon-enriched and carbon-depleted regions – a process known as demixing.

From a technical point of view, observing liquid carbon and hydrocarbons using X-ray diffractions would also represent a breakthrough for HED research. Due to the low atomic number, the scattering cross-section of the atoms is very low, meaning that the diffraction signal is correspondingly weak. While signal can be obtained for such low-Z samples in solid phases, such as diamond and graphite, and for higher-Z elements in phases without long range order, such as the results on warm dense aluminum, low-Z materials without long range order have not previously been studied with single X-ray shots.

３．研究の方法

The experiments described here were carried out at the X-ray Free Electron Facilities of SACLA (SPring-8 Angstrom Compact free electron Laser), in Harima, Japan, and LCLS (Linac Coherent Light Source), in Stanford, USA. At both facilities, the XFEL beam was used to probe samples at high pressures and/or temperatures, using X-ray diffraction.

The use of lasers to drive shocks into samples is well established, and works by the laser rapidly ablating the surface and creating an expanding plasma, which correspondingly compresses the sample due to the rocket effect. The conditions reached by the shock are governed by the Rankine-Hugoniot relations, such that a shock at a given pressure will give a consistent density and temperature rise in a material. In general, a single shock will give too large a temperature rise to reach conditions relative to planets; this can be avoided by using successive shocks, giving the same total pressure increase but with a lower associated temperature. In the experiment at LCLS, for instance, a double shock scheme was used to give a temperature of 5000 K at 150 GPa. Reaching the same pressure with a single shock would give T>10,000 K.

The experiments at SACLA had different aims and sought to use the unique capabilities available at SACLA to reach more exotic conditions. In the first, a shock-compressed sample was subsequently irradiated by a short-pulse, high-intensity beam. Since the beam primarily couples to the electrons rather than the ions, by a factor of ~1000, this gives a much higher electron temperature, without affecting the ions. With such temperatures, the average ionization state is much higher, increasing the forces between the ions, and simulating the effect of higher pressure. If the system is then probed before the electron and ion temperatures have equilibrated, something that previous work has suggested can take up to ns, the highly-ionized system can be observed.

In the latter, samples were instead heated directly by the X-ray beam, and the ion structure evolution observed by diffraction. Such an experiment is not possible at LCLS, as they cannot focus the beam to a small enough spot, but X-ray heating has many advantages over laser heating, primarily that the sample can be heated uniformly throughout its depth. As X-ray lasers become more widespread and increase in power, such techniques will only become more common, so preliminary research in this area is very valuable.

４．研究成果

Three main experiments have been carried out, supported wholly or in part by this funding:

SACLA experiment 2016B8044: *New Phases in High Energy Density Carbon*

LCLS experiment LP34: *In situ investigation of nanodiamonds formed in shock-compressed plastics*.

SACLA experiment 2017B8075: *Crystal Mismatch Heating as a Mechanism for Ultrafast Melting*

One experiment proposal has been accepted, and will carried out after the end of the funding period, but is supported by project funding:

SACLA experiment 2018A8056: *Thermal and Nonthermal Melting Driven by X-Ray Heating*

Additionally, equipment bought with the grant money continues to be used in to analyze data from other beamtimes at SACLA.

The LCLS experiment used the new pulse-shaping capabilities of the long pulse laser at the MEC (Matter at Extreme Conditions) beamline to reach planetary interior conditions with two shocks in combination. A variety of different targets were used, including graphite, diamond and a variety of plastics. In polystyrene, nanometer sized diamonds were observed to form, confirming the results from the previous beamtime (Kraus et al. 2017). Other plastics were primarily observed in liquid structures, and agreed very well with first principles simulations of such materials; the results were compared to DFT-MD simulations carried out by colleagues in Germany, and the results are currently under review with Physical Review Letters (Hartley et al. 2018). A comparison on the experimental and computational data, for the case of CH (polystyrene) at 190 GPa and 10,000 K, is shown in Figure 1.

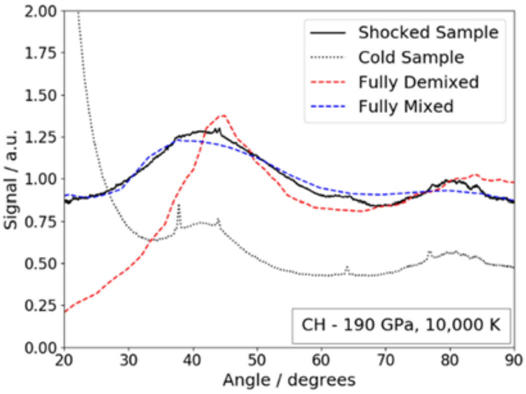

Figure 1: Example of the analysis of experiment LP34, showing the comparison of experimental data (black solid line) with DFT-MD results.

The first SACLA experiment aimed to use both long and short pulse optical beams in combination with the XFEL, as shown in Figure 2, in order to create a highly-ionized, transient state, with the strong forces between the ions giving an analogous effect to that of additional compression. Some predictions suggested that this could cause the formation of a purely ionic crystal (Coulomb crystallization) from an initially covalent system.

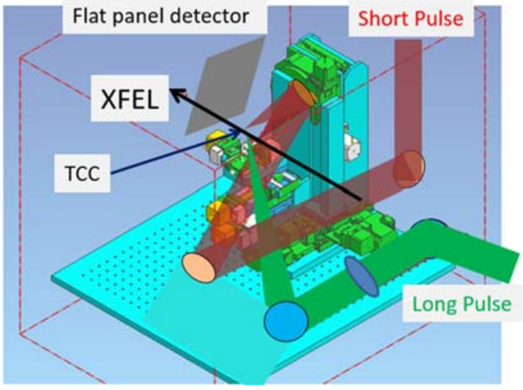

Figure 2: Setup of the lasers in 2016B8044, the first beamtime using all three available beams.

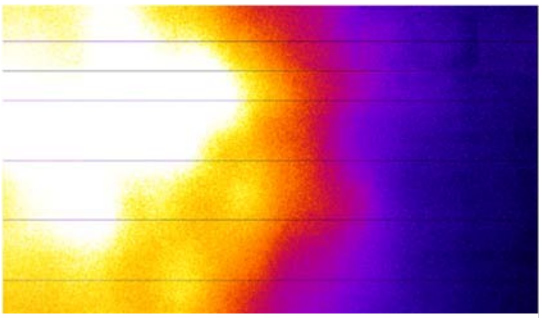

Figure 3: X-ray signal on flat panel detector, showing random emission from the coronal plasma, and no diffraction peaks. Orientation is as in Figure 2.

This beamtime was unfortunately not successful – the beamline was using a new detector which appeared to be very susceptible to noise and EMP from the laser-target interaction, even with the long-pulse laser, where much lower electron temperatures are expected. While we did attempt one shot with the short pulse laser), there were no diffraction lines from cold or new crystal structures, and it was not possible to extract useful data from this (see Figure 3.

In discussing the results from this beamtime, and ways to improve the results in the future, the idea of using the XFEL beam itself to heat the sample was proposed. This has only recently become possible, with the ability to tightly focus the XFEL beam to sufficiently small spots that the absorbed energy is enough to cause melting. As with optical lasers, the electrons are preferentially heated, so a strongly non-equilibrium system is again formed. Using SACLA's two-color mode, one pulse can heat the sample, and the second observe changes in the ionic structure. Due to how SALCA generates such a beam, this is only possible at small delays, up to 300 fs; to reduce the complexity of the experiment, optical lasers were not used.

We used thin samples, such that the entire depth is uniformly heated, but for carbon samples (diamond, graphite) this had the unfortunate effect of meaning that the crystallites were too large and the samples too textured, such that no diffraction signal was seen. Signal was seen for metallic samples, and the changes in signal for copper and aluminum are seen in Figure 3.

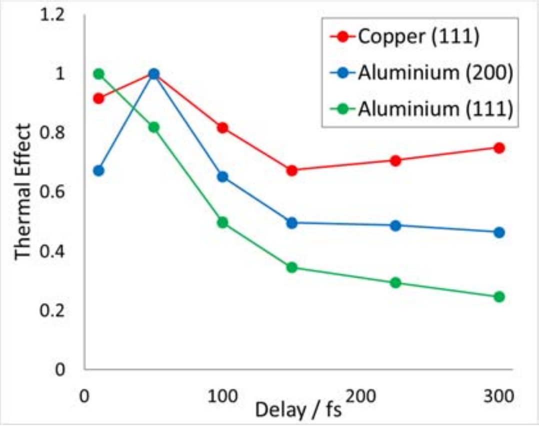

Figure 4: Relative diffraction signal as a function of delay for X-ray heated samples

In general, a drop at large delays is observed as the heated electrons exchange energy with the ions, leading to a loss of order and weakening signal, described as the Debye-Waller effect. The apparent rise at 50 fs is unexpected, but may be due to the lack of shots at the earliest delay (10 fs), and a correspondingly larger error bar (not shown).

This begins to demonstrate the capabilities of X-ray heating as a technique. The upcoming beamtime will continue to study this, focusing on non-metallic targets, including graphite, diamond and silicon. In these targets, non-thermal effects are expected to be more important, including due to induced Coulomb forces between the inefficiently packed ions, known as crystal mismatch heating. The targets are also thicker, meaning that the signal should be easier to observe, without any significant loss of homogeneity in heating. Building on these results in the future can again aim to look at additional heating and ionization in shock-compressed samples, although X-ray heating as a study in its own right is also a fruitful area of research, as some of the first work possible in this area.

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６．研究組織

(1)研究代表者

ハートリー・ニコラス (Nicholas・HARTLEY)

Specially Appointed Assistant Professor・

Open and Transdisciplinary Research Initiatives・

Osaka University

研究者番号：10770901