Determining Melt Curves using Liquid X-Ray Diffraction

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Introduction

The phase boundary between solid and liquid is a key material property, influencing, for example, planetary structures. At pressures relevant to supergiant planets, dynamic processes such as laser compression are generally necessary to reach the conditions of interest.

Since the melting temperature is dependent on the heating rate of the material, care must be taken when interpreting dynamically determined melt curves.

LAMMPS (an MD simulation) has been used to model dynamic melting during compression and release of samples.

From this, the effect of melting kinetics on observed melt onset can be investigated. This method opens up an experimental approach to dynamic determination of melt curves, and of measuring melting kinetics.

Z Method

When heating a material, the temperature-time profile follows a Z shape and the temperature at the trough of this profile is taken as the dynamic melt temperature. This is dependent on heating rate.

The static melt temperature can be determined from the exponential relationship between the melt temperature and heating rate. Kinetic information is also obtainable.

This was carried out in the simplest situation of heating a cube of Cu until it melted. Cooling from liquid to solid had the same effect, tending towards the same temperature in the static case.

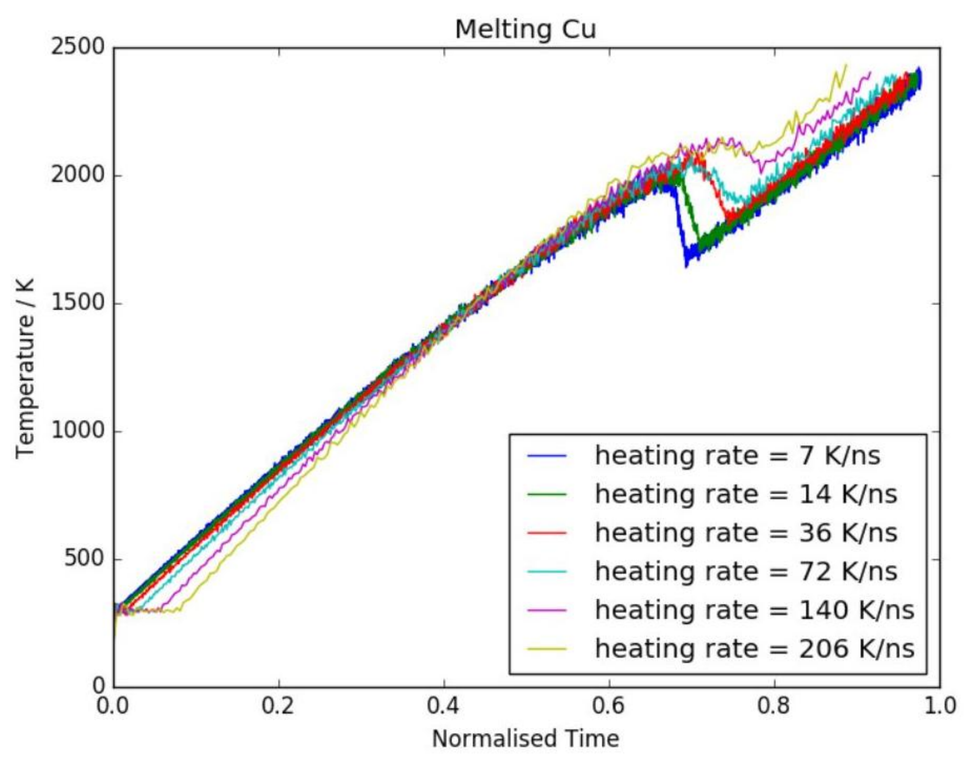

Figure 1
The standard z profile on heating.
With increased heating rate the melt temperature increases and the z-profile flattens.

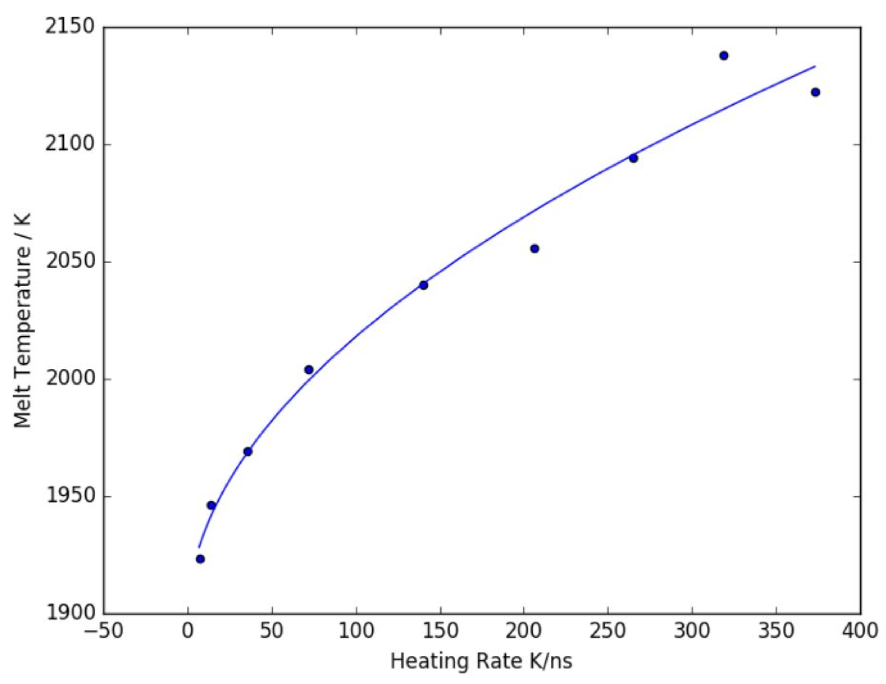

Figure 2
The logarithmic relationship between melt temperature and the rate of temperature change.

Tamper

A tamper can be used in order to change the path of release after a shock.

By calculating the particle velocity in the tamper using impedance matching and shock data from Rusbank, a thin layer which acts as a tamper can be used which makes the simulations computationally cheaper.

Changing tamper material changes the temperature at which melt occurs, as would be expected due to the different release rate.

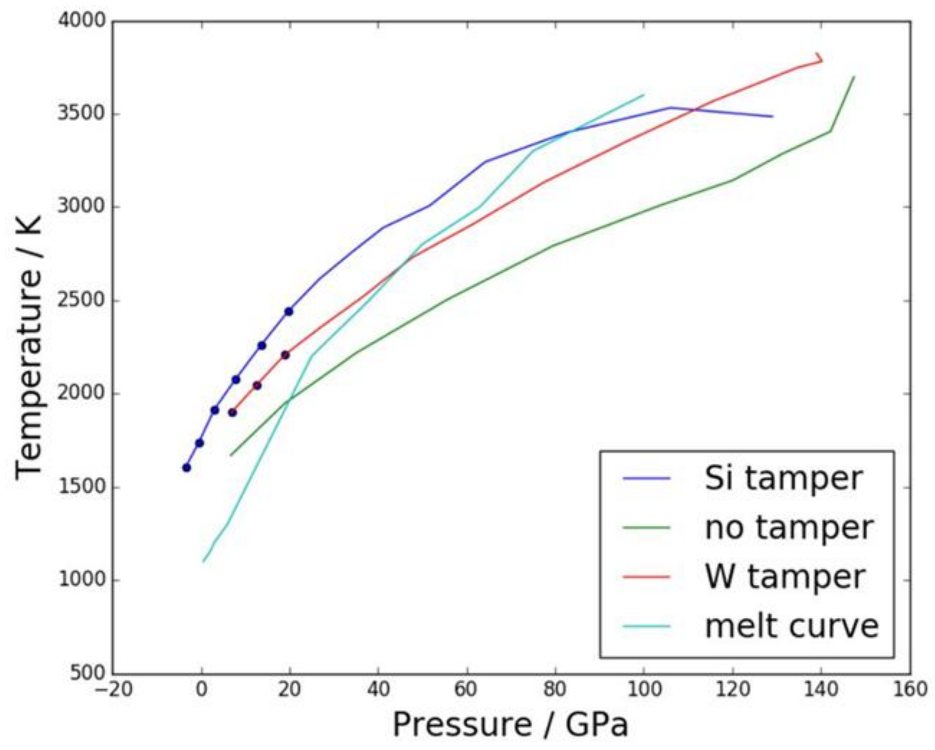

Figure 3

The pressure-temperature release paths of different tamper materials are shown.

The cyan line is the melt curve.

The data points are points at which melt has occurred.

Melt begins after crossing the melt curves, with the delay dependant on the different release rates.

Behaviour on Release

Release after shock is a dynamic system so overshoot of melt temperature is expected.

By using this dynamic method with different release rates, the static case can be calculated.

Kinetic parameters and the activation energy can be obtained from this fit.

This method could also be used for solid-solid phase transitions, and the z-profile has been observed to happen during the change of deformation mode shown in Figure 4.

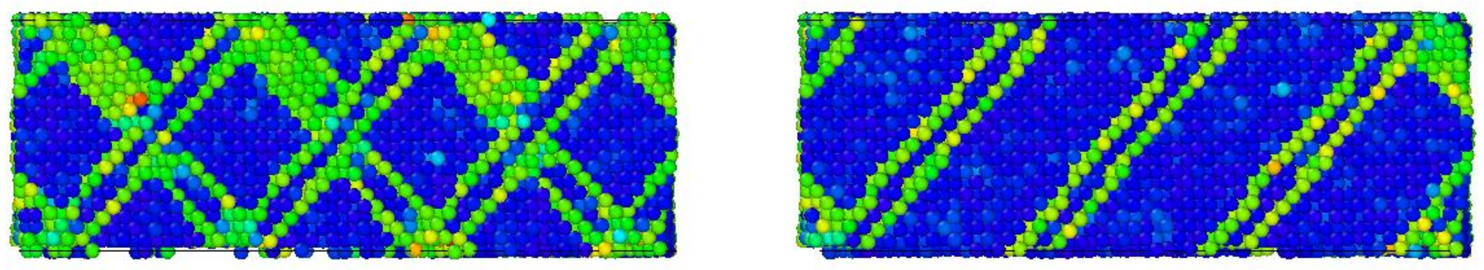

Figure 4 Different deformation modes in solid Cu on release.
The colour is based on the centro symmetry parameter.

Experiment

An experiment at the Gemini laser at the Rutherford Appleton Laboratories, UK will be carried out in the coming year, to test this theory.

A diagram of the experimental setup is shown below in Figure 5.

Sn is the main target to used in this experiment, because the melt curve and Hugoniot coincide in the range 50-70 GPa.

Shocks and release will be repeated, with different shock forces, with pressures of up to 100 GPa.

When the material is in coexistence, temperature and pressure are the same in the solid and liquid regions, so these parameters should both be calculable using the solid and liquid diffraction.

Different tamper materials will be used to show the effect of release rate on observed melt temperature.

Static melt temperature will be inferred from these dynamic melt temperature measurements, using the technique outlined above.

A CdTe detector will be used in single photon counting mode. The Single Photon Energy Dispersive X-ray Diffraction (SPEDX) will be used due to the low x-ray flux compared to XFELs.

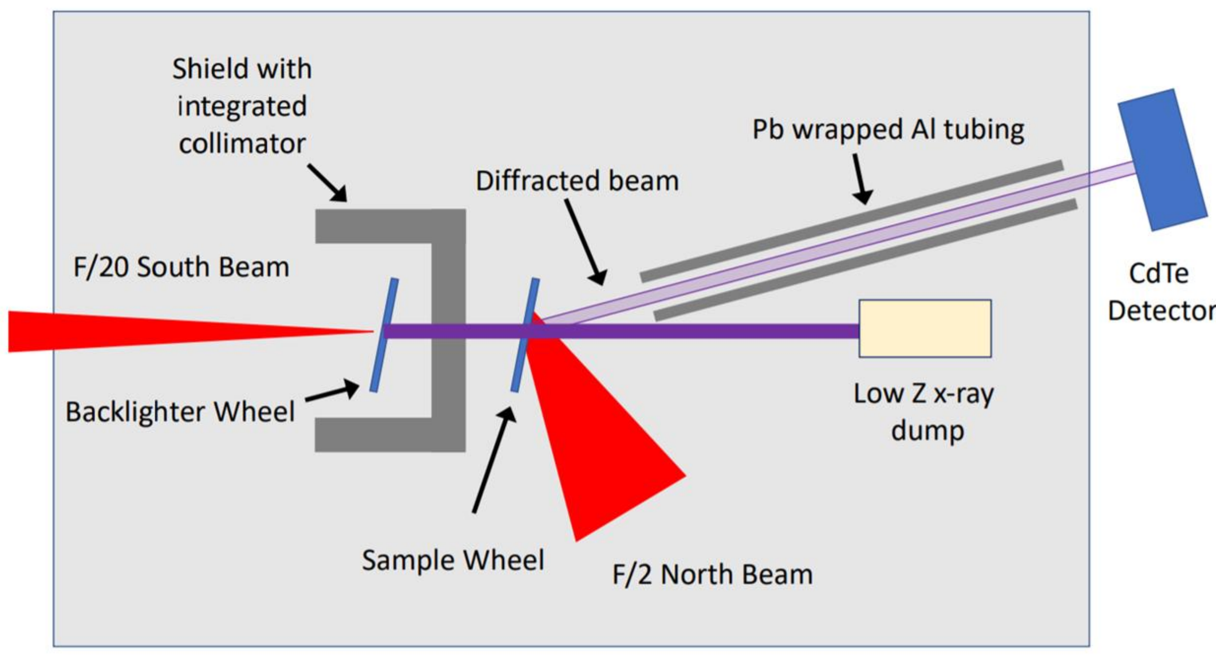

Figure 5

Design of target chamber for 2020 Gemini experiment.

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Conclusion

MD simulations show that static melt temperatures can be inferred from dynamic results.

This can be used to extract kinetic information, and may also be applicable to solid release behaviour such as deformation modes.

A future experiment aims to serve as a proof of principle that by using the solid and liquid diffraction patterns at melt, a pressure and temperature can be extracted.

This could provide a method to experimentally determine phase boundaries and verify equation of state models at the high pressures relevant to planetary cores.

References

Simulations carried out using LAMMPS http://lammps.sandia.gov

S. Plimpton. LAMMPS User's Manual, Sandia National Laboratories, 2017

Simulation visualised with OVITO http://ovito.org/

A. Stukowski. Visualization and analysis of atomistic simulation data with OVITO–the Open Visualization Tool, Modelling and Simulation in Materials Science and Engineering, 18(1):1–7, 2010

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