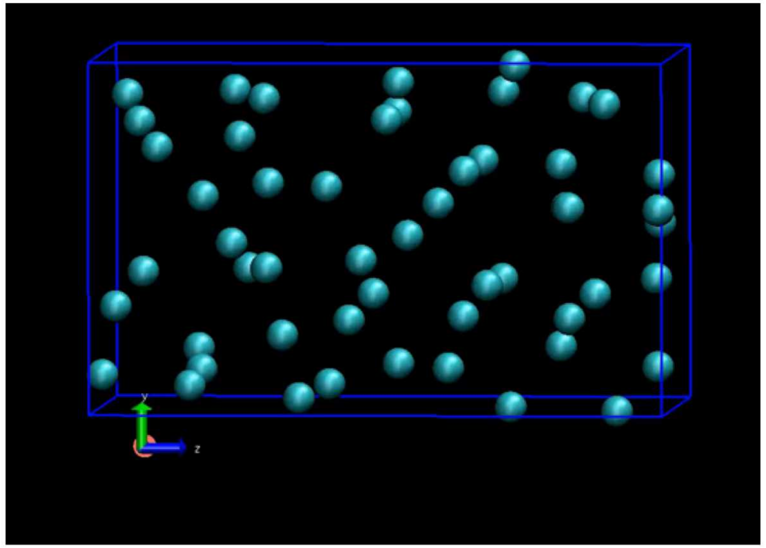
SAND2019-6841C

Dynamic Freezing of Liquid Cerium Under Shock-Ramp Compression



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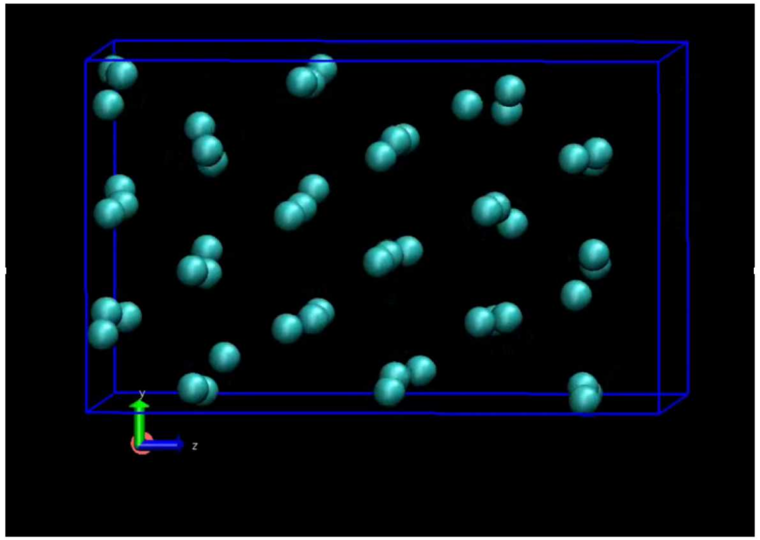

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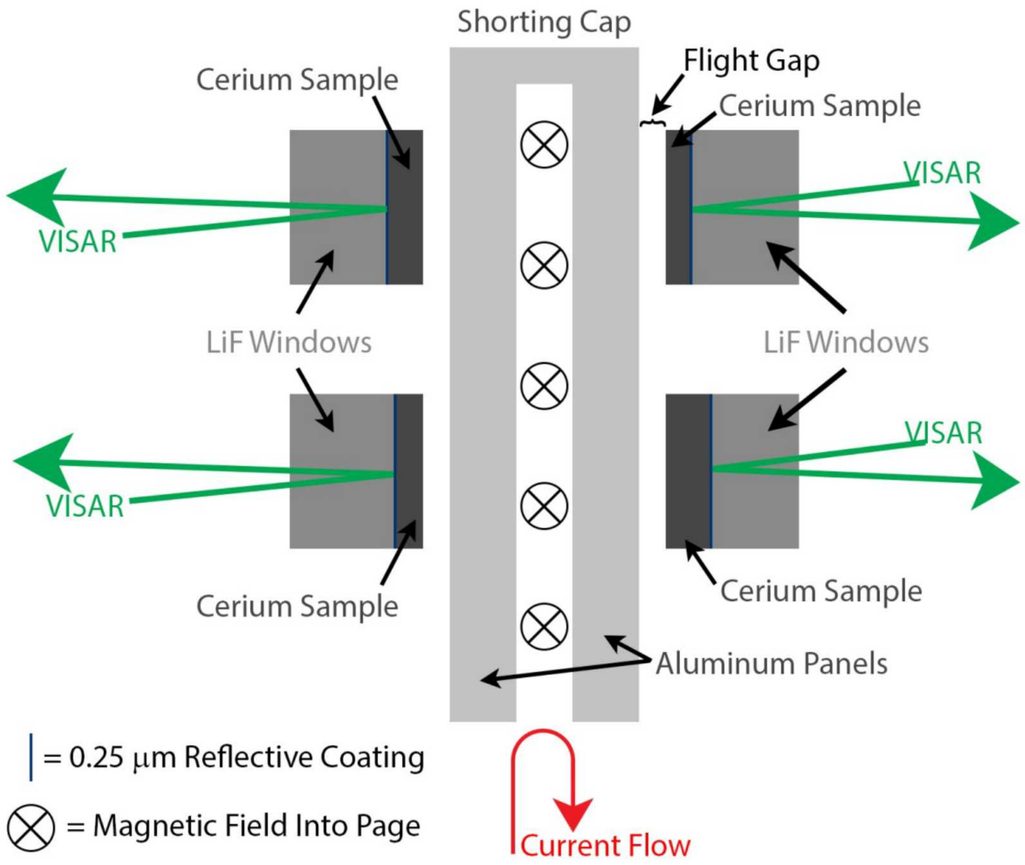
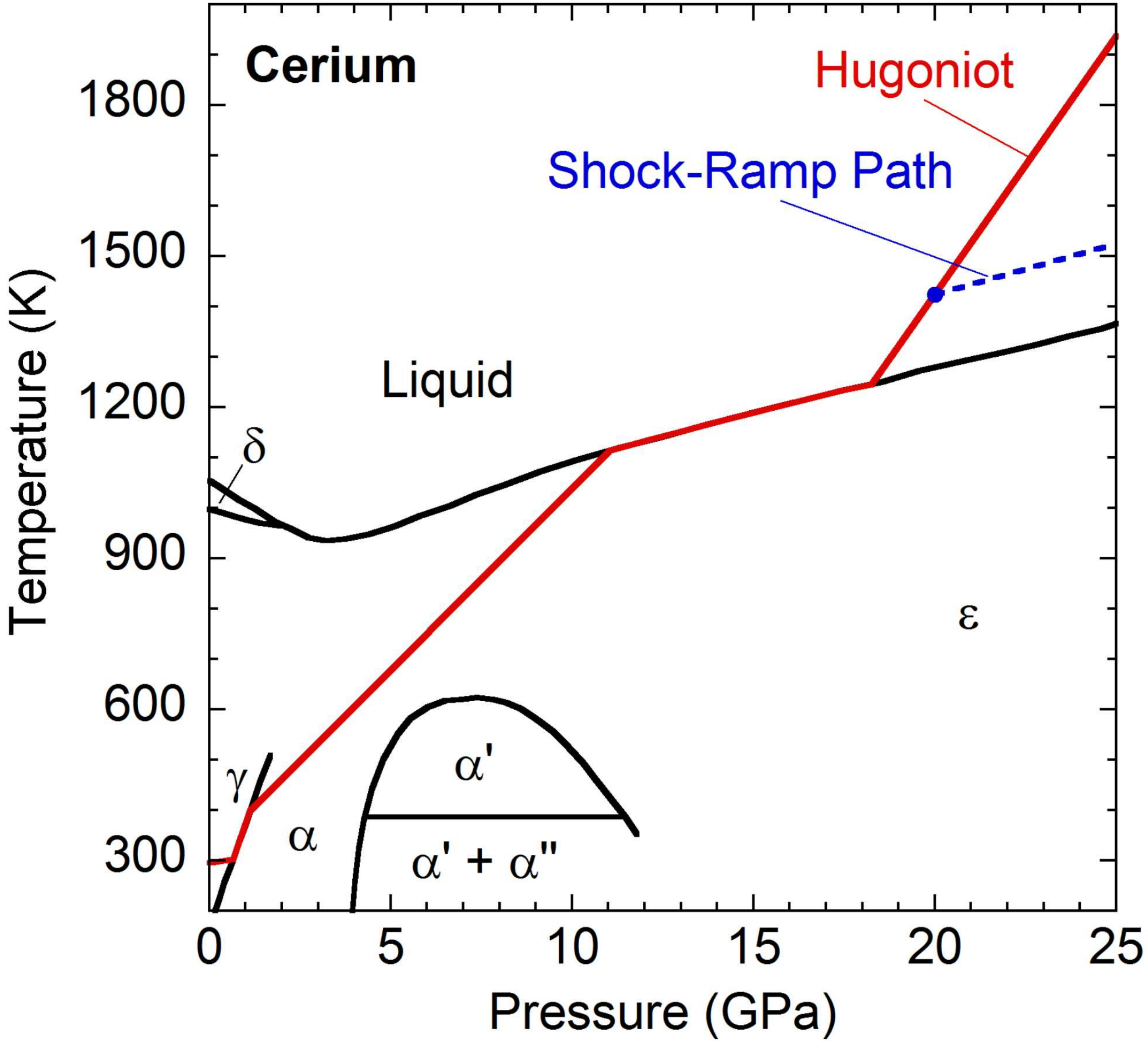
Cerium shock melts at ~10-18 GPa on the Hugoniot





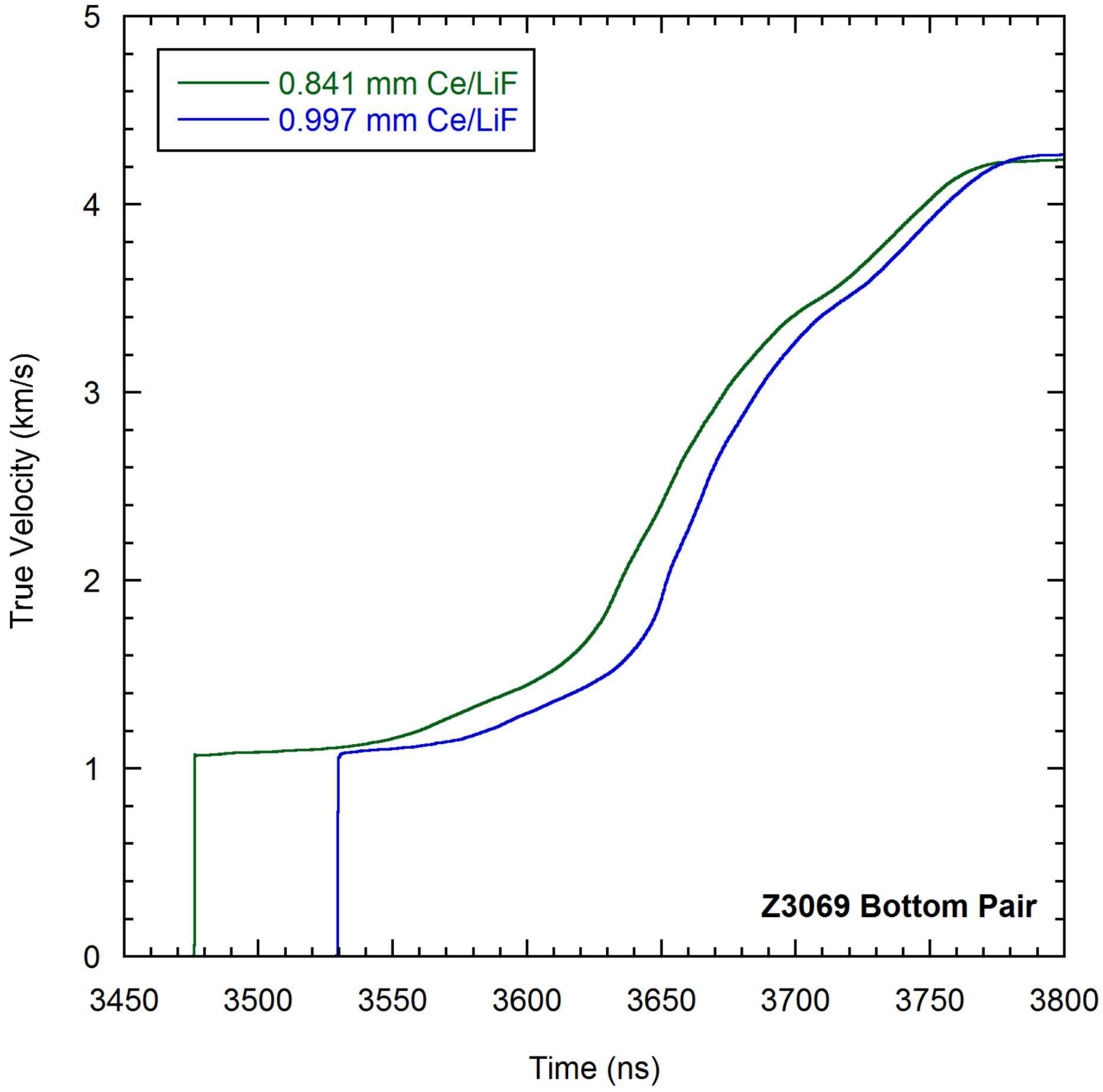
We performed shock-ramp experiments on Z to explore the freezing of liquid cerium under ramp compression

We have completed four “modern” experiments* starting with Z3005. (Older experiments reverberated very early). Smooth ramp compression from ~20 GPa initial shock.

Each experiment utilized 3-4 sample pairs. Some of the thickest samples from the last two experiments (Z3145 and Z3248) shocked up during the ramp. Those data were discarded from this analysis, which left 10 sample pairs representing shockless compression from an initial ~20 GPa shock.

Sound speed data from those 10 pairs are statistically consistent with one another (typically within mutual error).

Velocimetry shows no obvious “kink” in raw velocities.



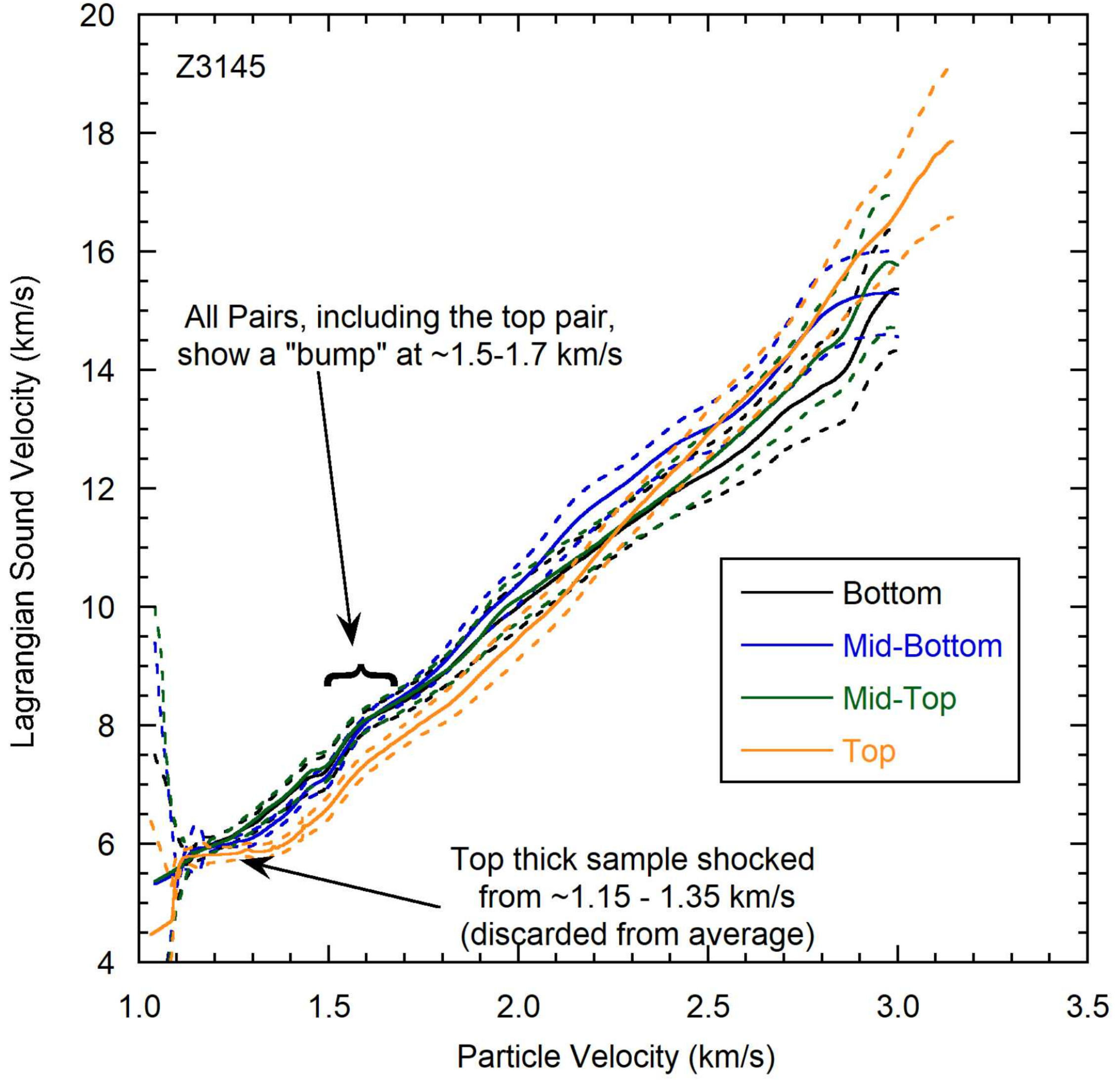
*Chris Seagle is the PI for these cerium experiments

Individual sample pairs exhibit a suggestive bump in the sound speed around 1.5 km/s --- but this feature is not statistically significant for single pairs

A total of four experiments have been executed, each with 3-4 sample pairs.

Most pairs were shockless beyond the initial shock. (Samples that shocked up during the ramp were discarded from this analysis.)

None of the sample pairs exhibit elastic behavior on initial loading from the starting shock state – the cerium is shock melted.



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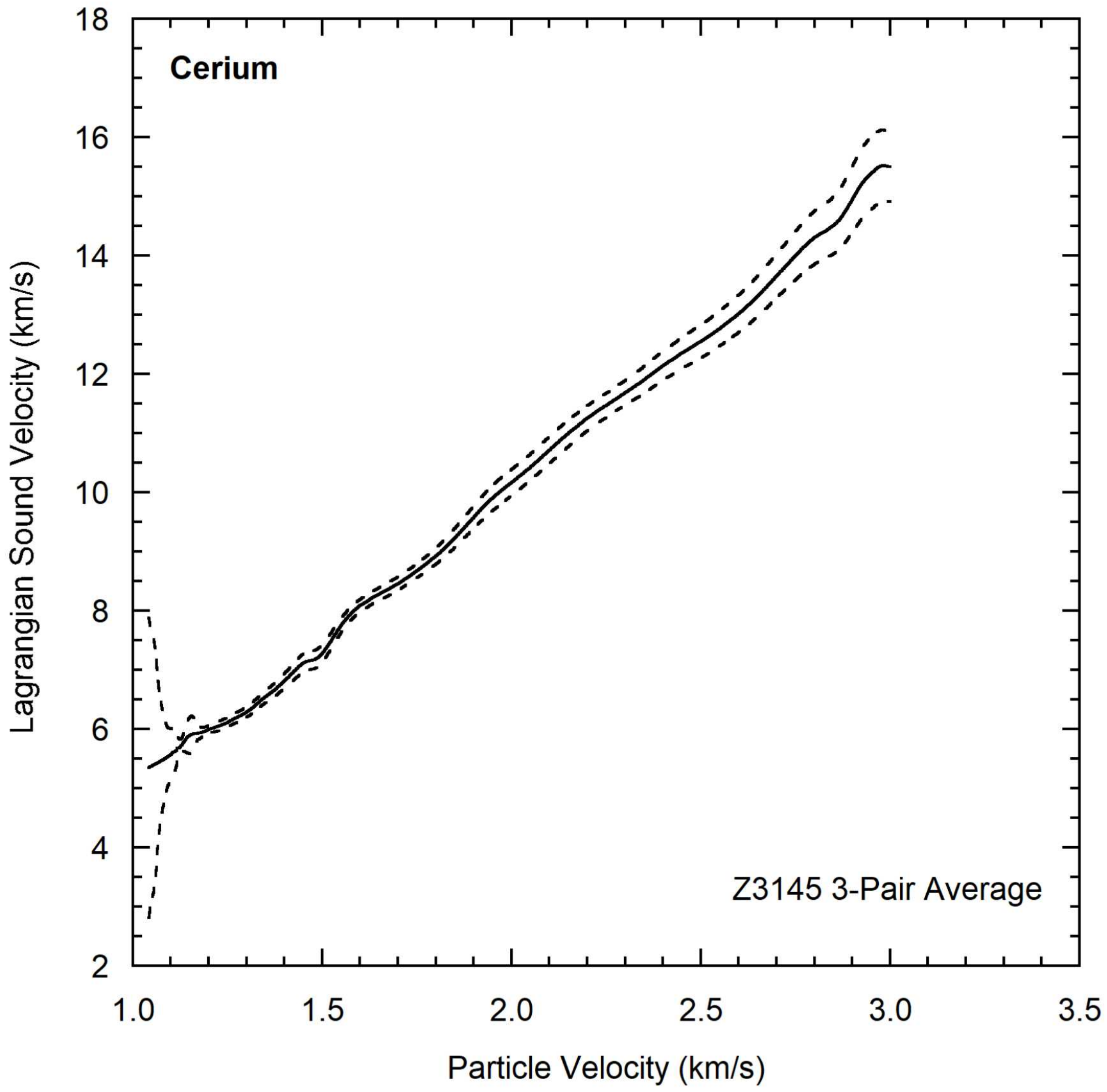
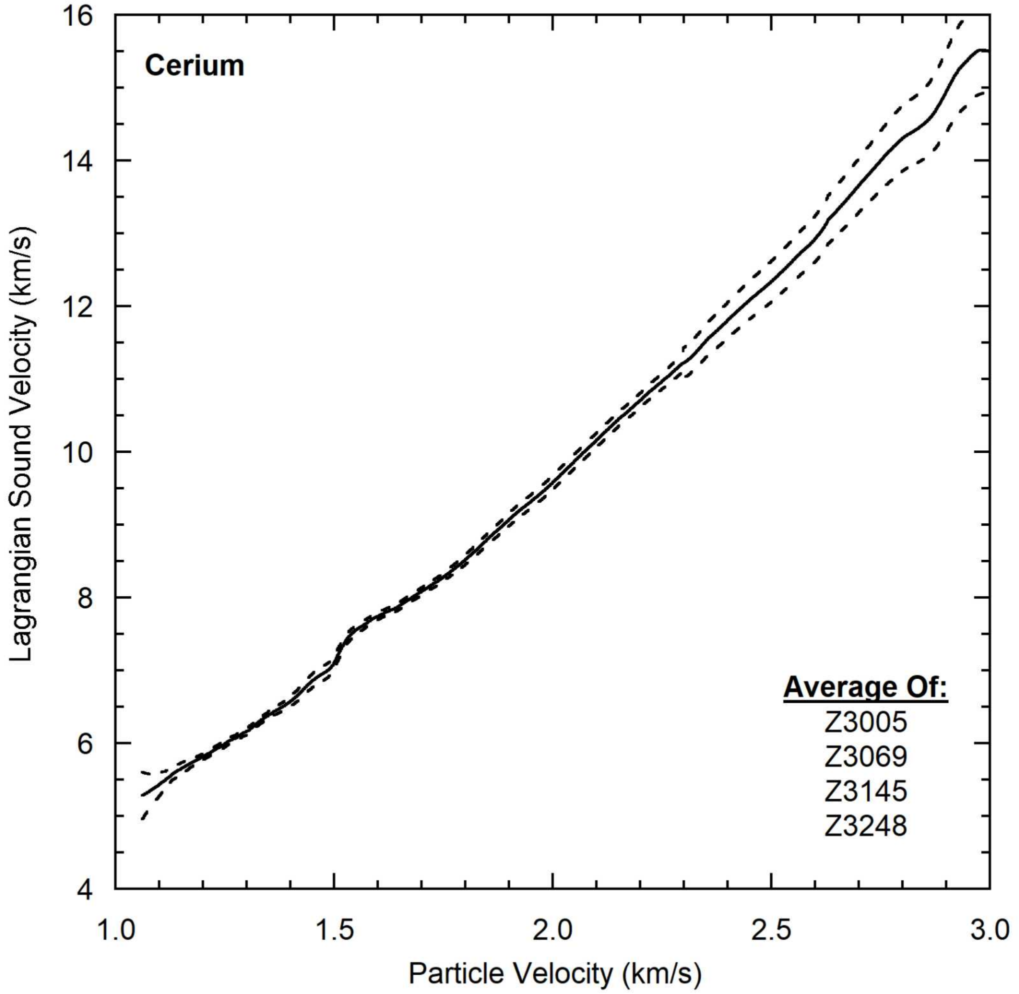
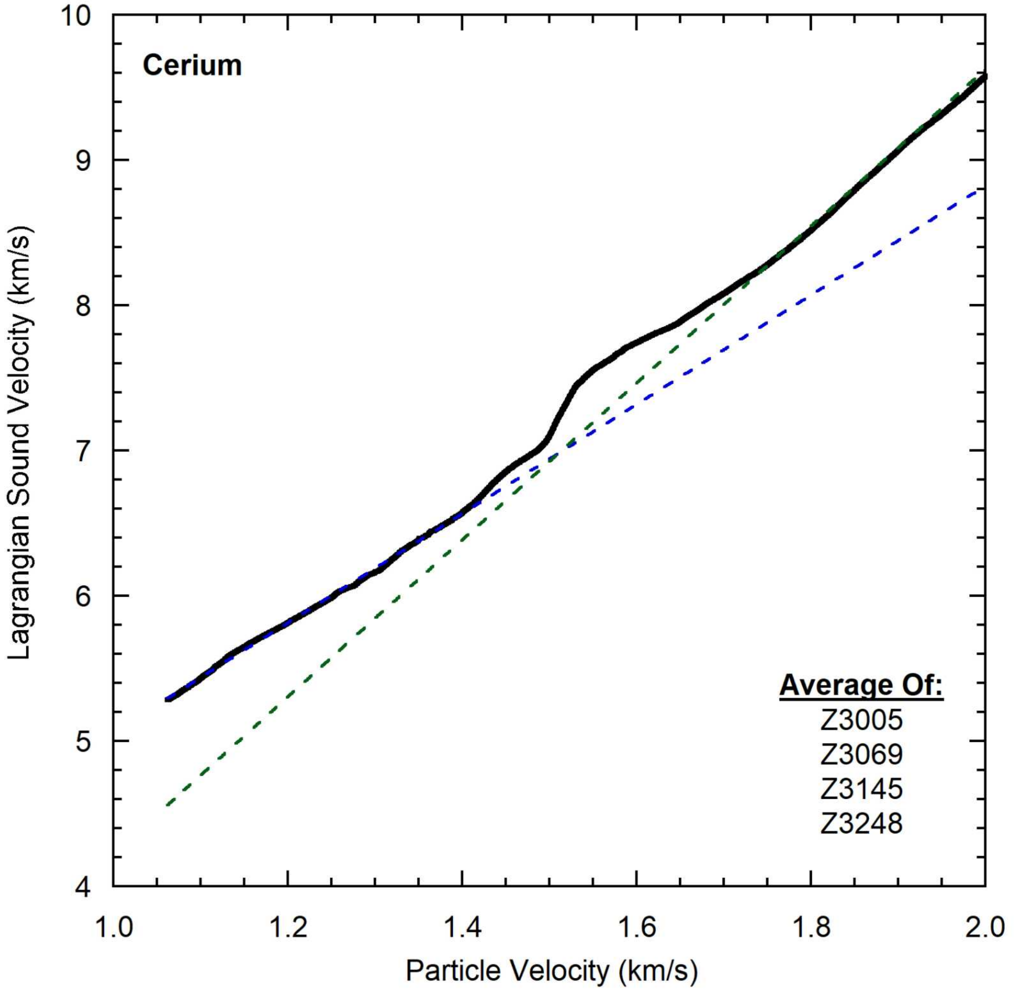
Averaging multiple sample pairs suggests a statistically significant bump in the sound velocity



The emergent feature is reminiscent of elastic-plastic behavior typically observed on initial loading in a pure ramp experiment (or sub-solidus shock-ramp experiment)

Averaging a total of 10 sample pairs reveals a statistically significant feature





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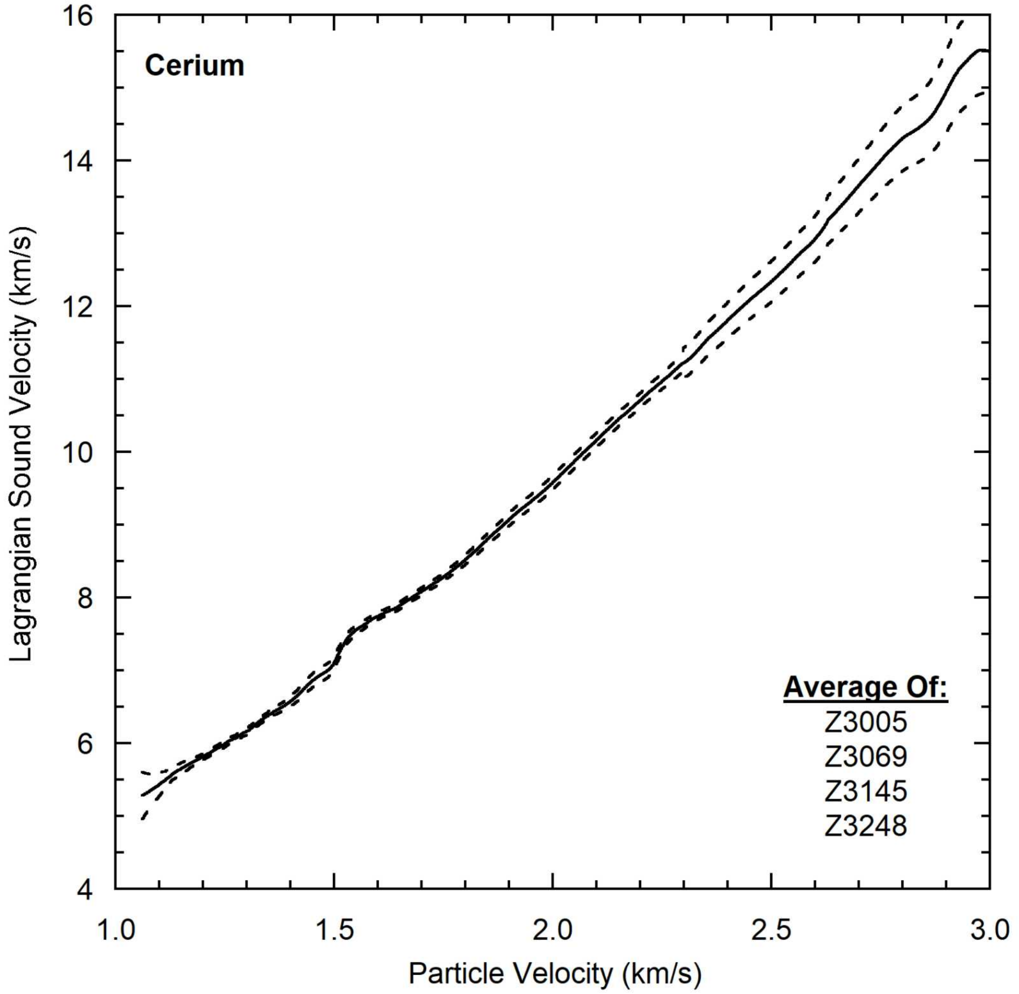
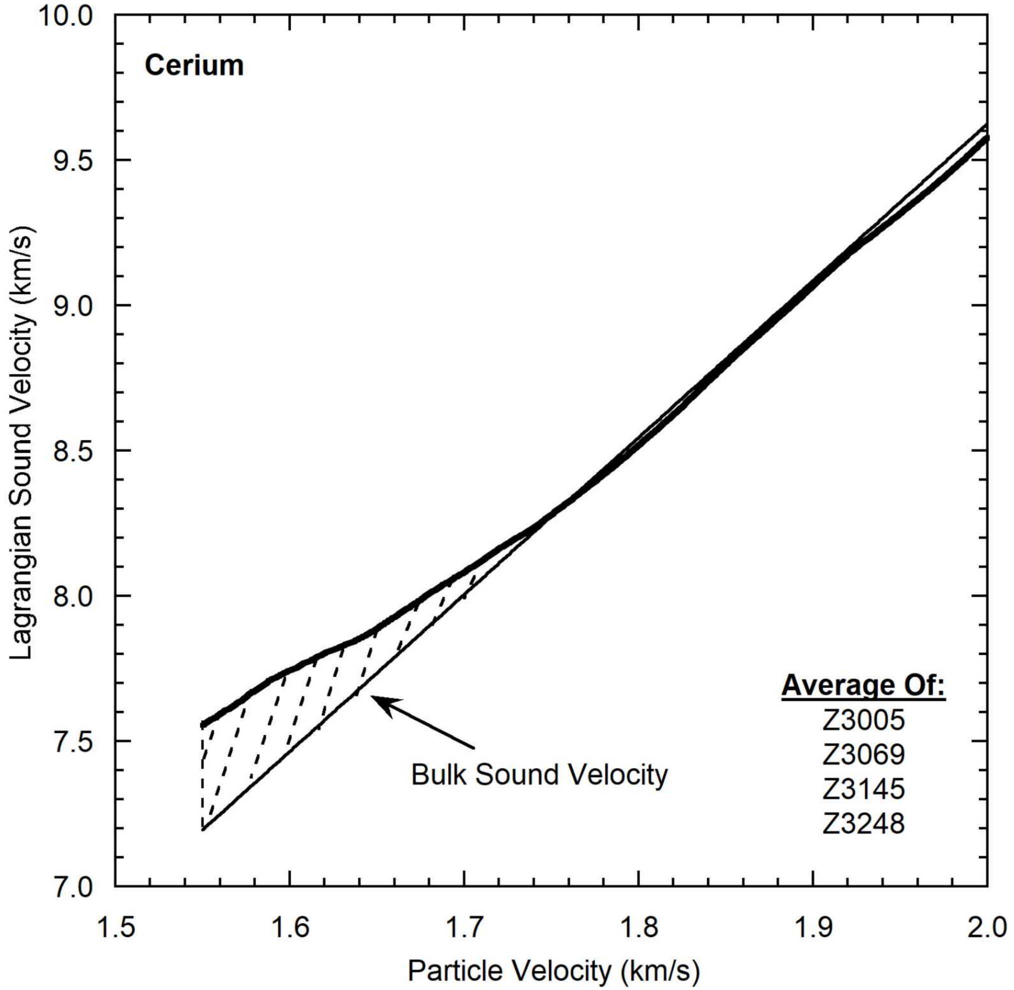




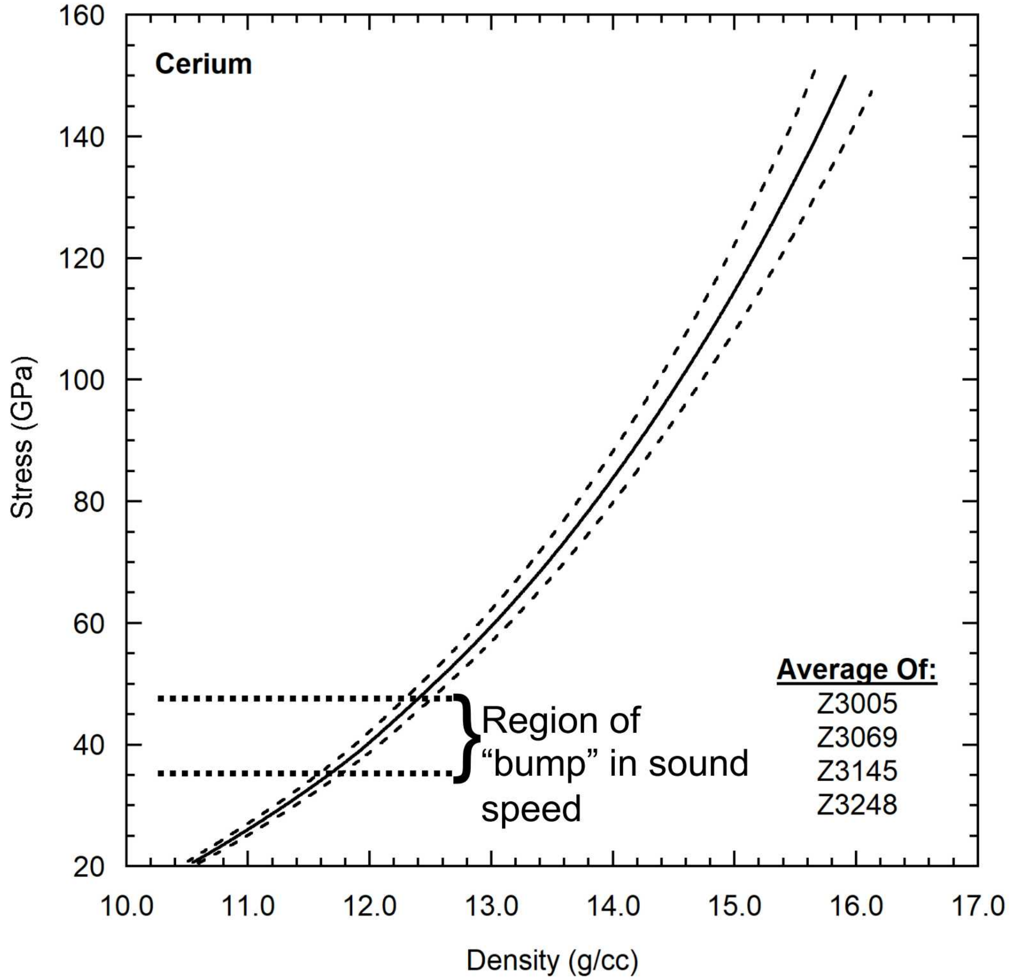
The averaged data provides a good constraint for the equation of state

Average of four experiments places tight constraints on the cerium EOS

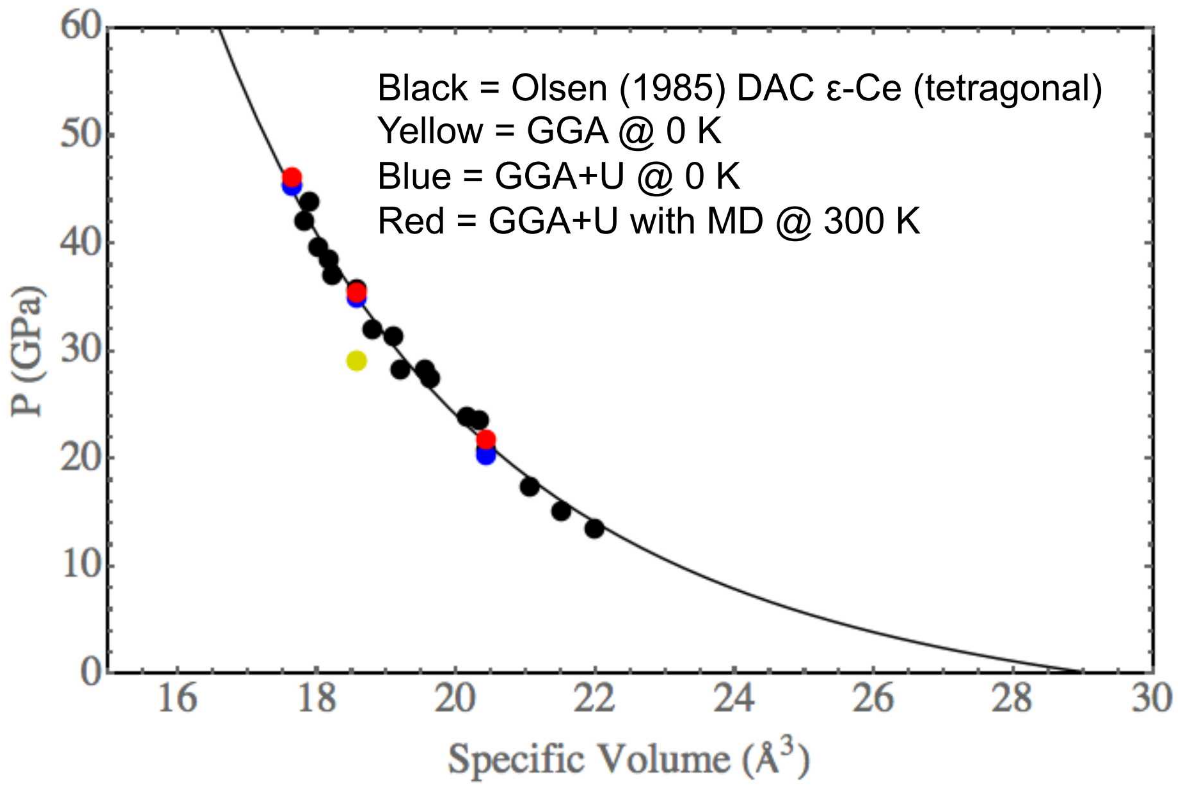
Low error on stress-density, 1.7% and 1.4% respectively at peak stress of ~150 GPa

Other than the Hugoniot and shock-release, this is the only data constraining the dynamic cerium response above ~40 GPa

No obvious kink in stress-density



We performed DFT simulations of cerium to compare the experimental observations with theoretical calculations of the cerium response

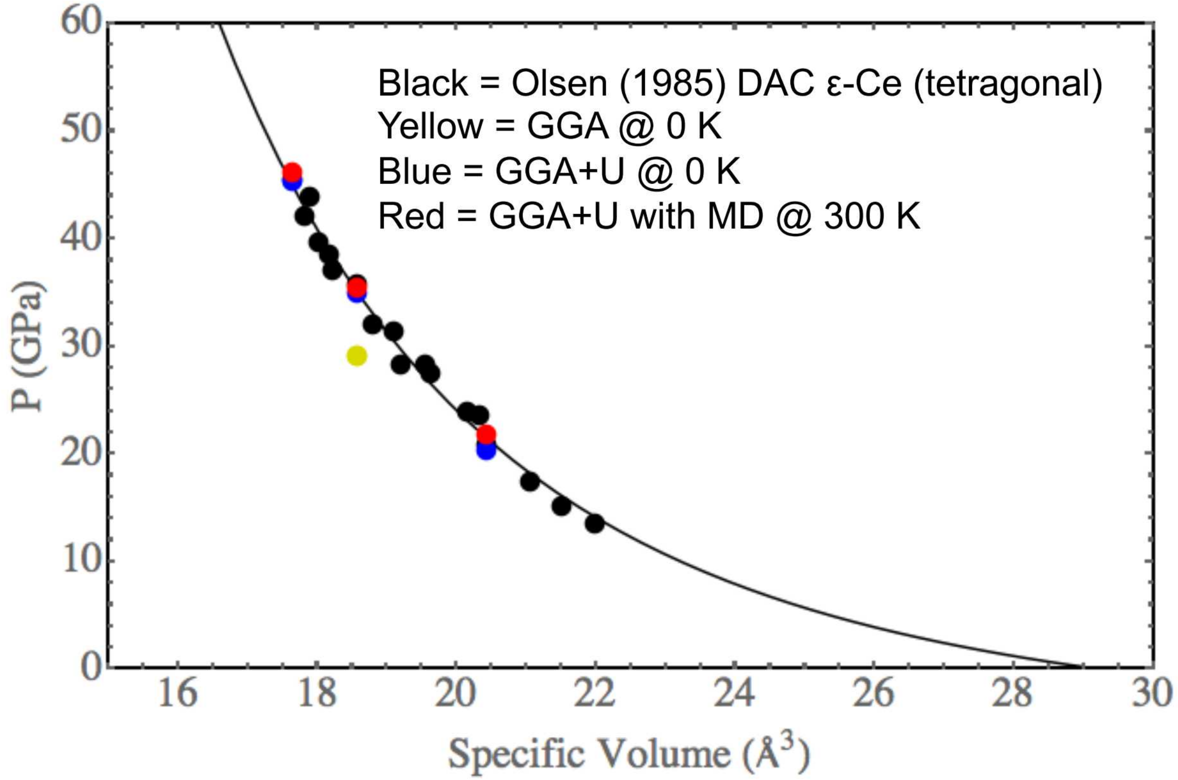


DFT-MD Simulations were first validated again static compression data

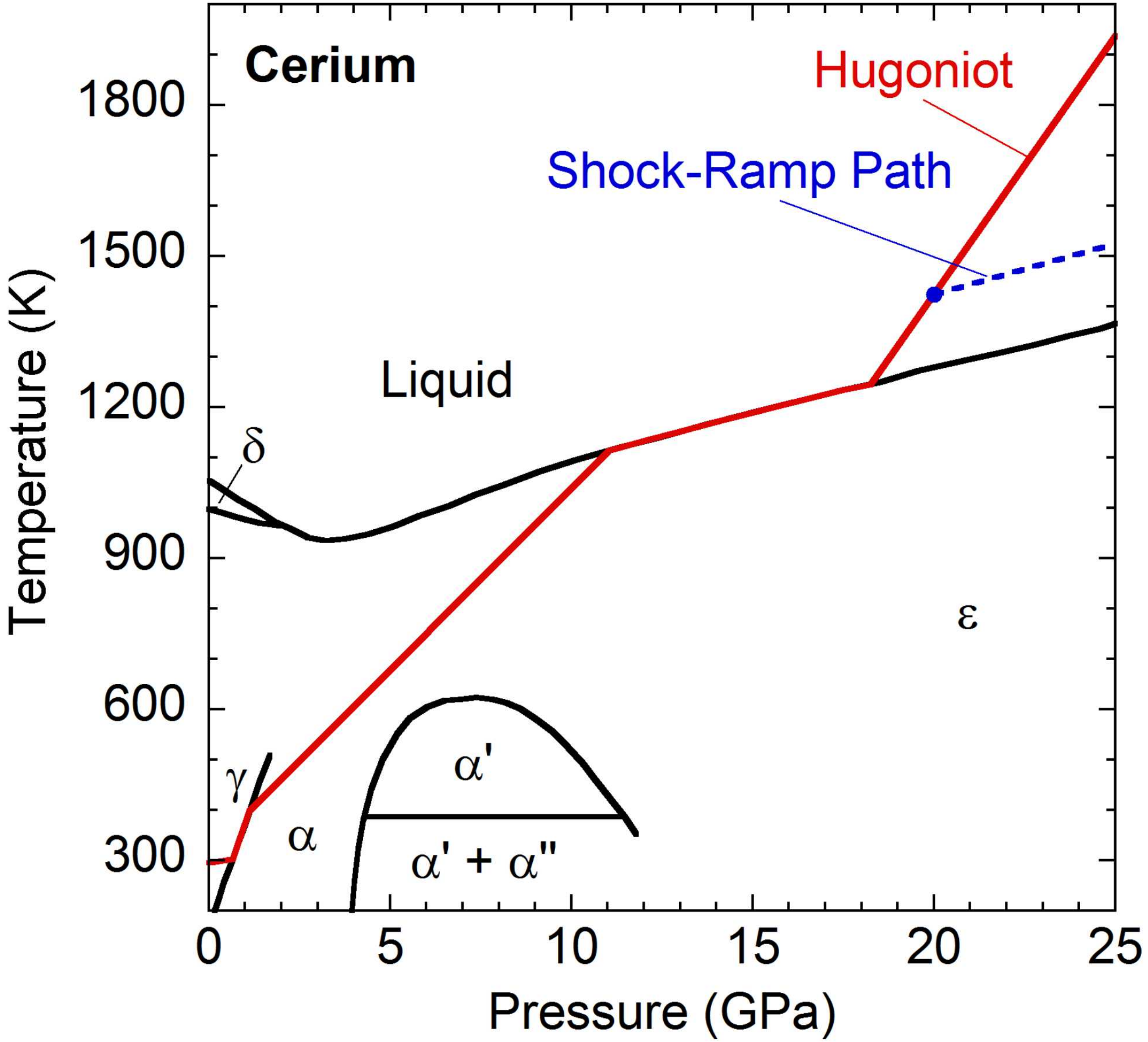
DFT with GGA+U gives good agreement with DAC data (Olsen, *et al.,* 1985) for ε-Ce

DFT GGA+U with 54 atoms, 12 electrons in the valence

DFT-MD simulations were then used to follow the compression isentrope from the experimental Hugoniot state



DFT GGA+U with 54 atoms, 12 electrons in the valence



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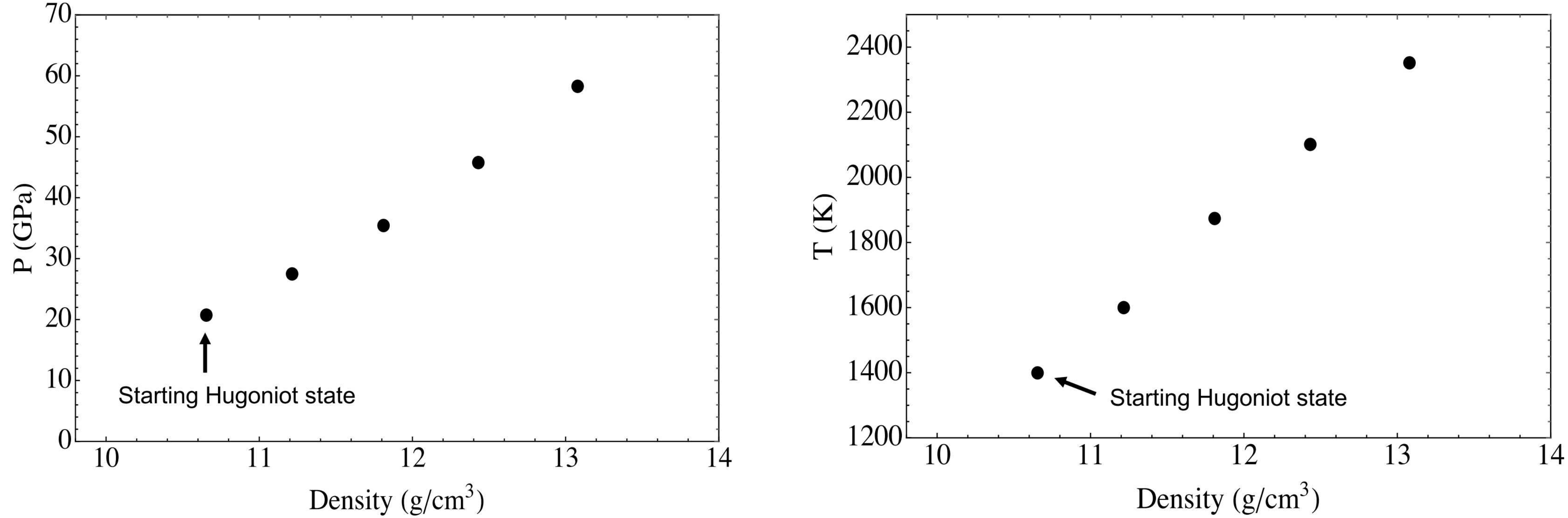
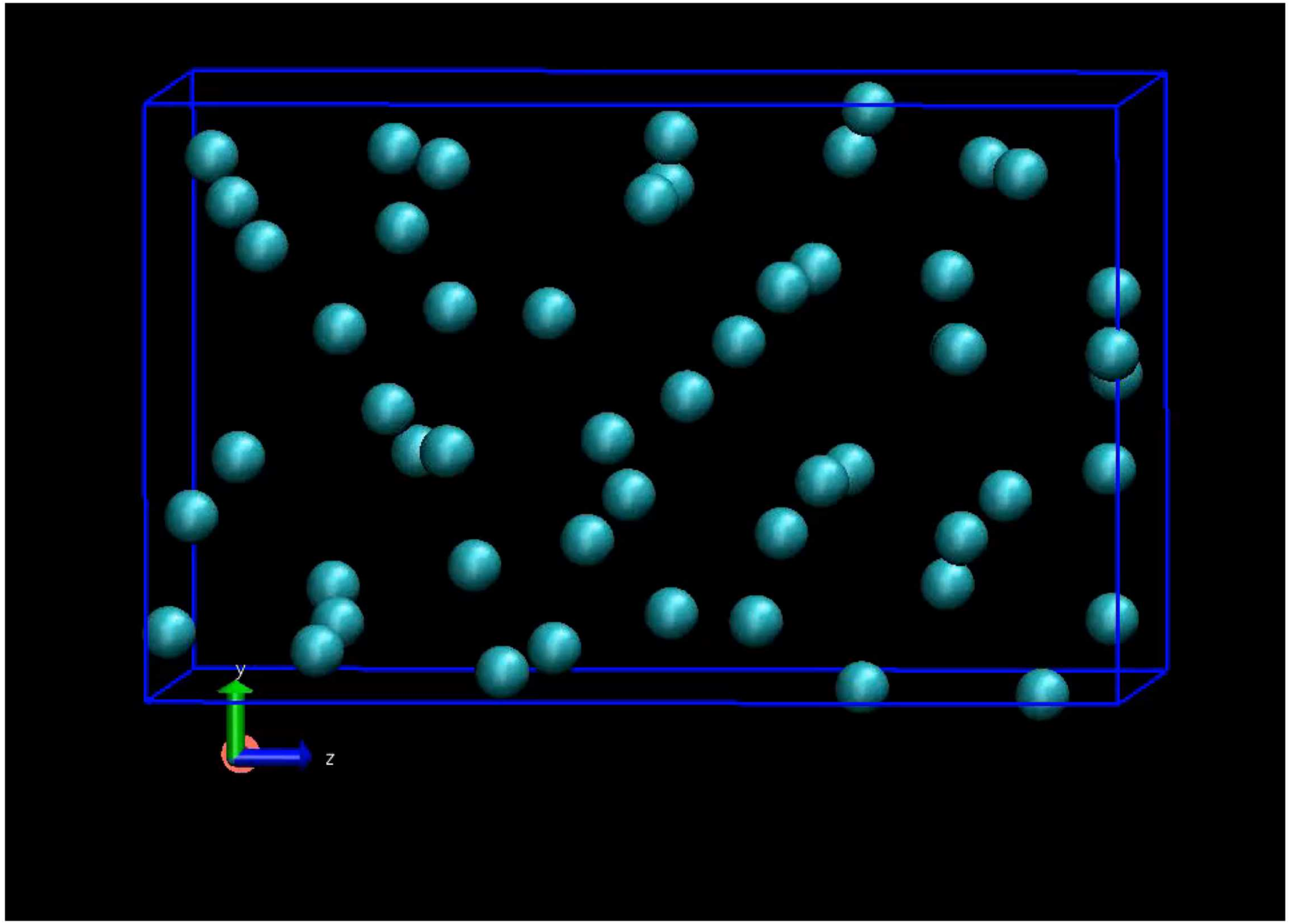


Direct entropy calculations were used at each step to follow the isentrope

Spontaneous Freezing was observed in the DFT-MD simulations at ~35 GPa



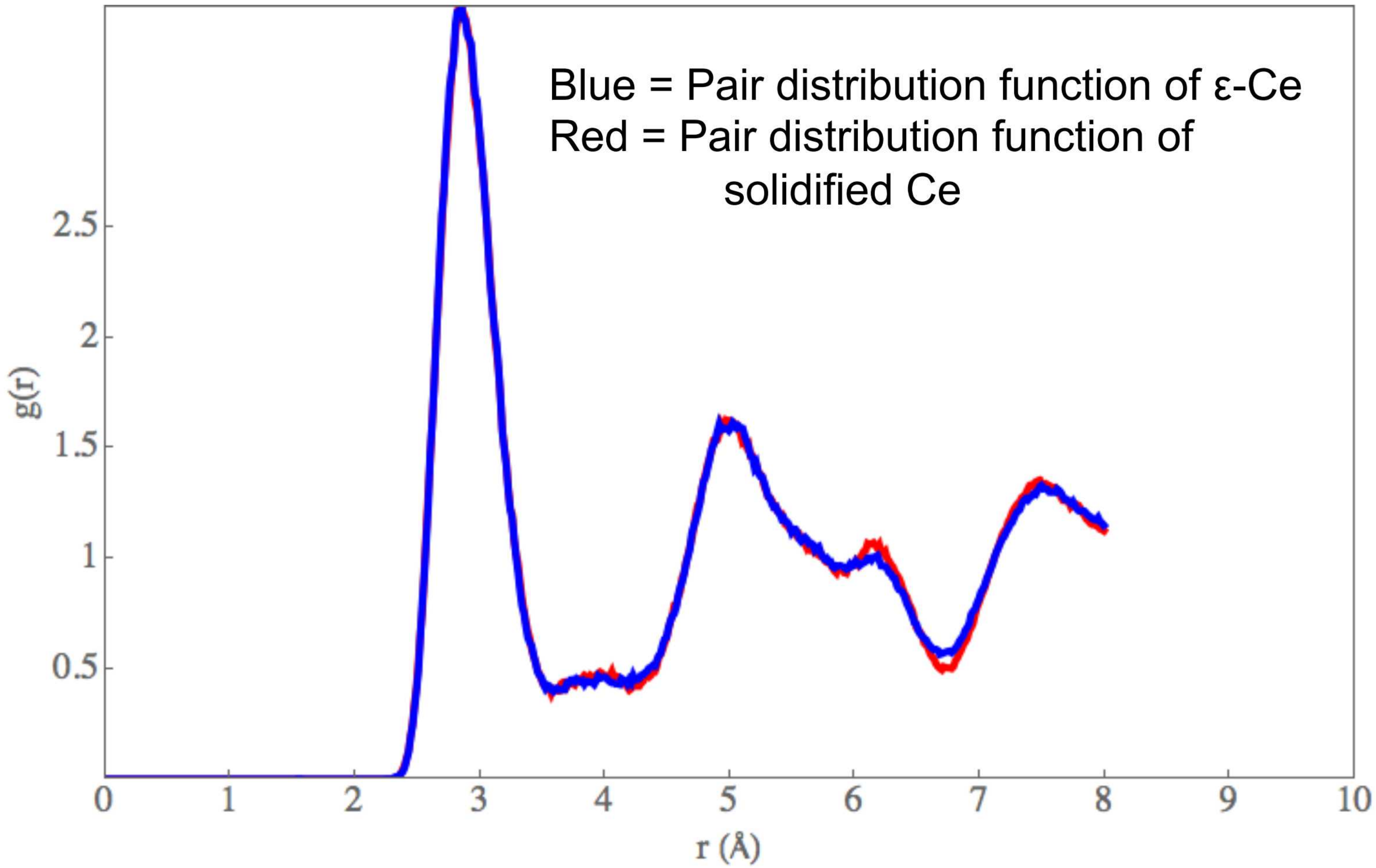
35 GPa, 11.8 g/cc, 1750K

The emergent phase from the initially liquid simulations is ε-Ce (tetragonal)

Spontaneous freezing is observed along an isentrope in DFT-MD (which originated from the experimental Hugoniot state) at ~35 GPa.

The “bump” in sound velocity observed experimentally occurs at ~35 GPa.

The body centered tetragonal phase of ε-Ce emerges from the liquid.



DFT-MD simulations with strained lattices were used to extract the second-order elastic constants

We perform the DFT molecular dynamics simulations* with the deformed lattice specified by

$$\vec{a}' = (\overleftrightarrow{I} + \overleftrightarrow{e}) \cdot \vec{a}.$$

For the body-centered tetragonal cell, we employ the two strain tensors

$$\overleftrightarrow{e}_1 = \begin{bmatrix} \delta & 0 & 0 \\ 0 & 0 & \delta/2 \\ 0 & \delta/2 & 0 \end{bmatrix} \text{ or } \overleftrightarrow{e}_2 = \begin{bmatrix} 0 & \delta/2 & 0 \\ \delta/2 & 0 & 0 \\ 0 & 0 & \delta \end{bmatrix}$$

with δ negative (contraction) or positive (dilation).

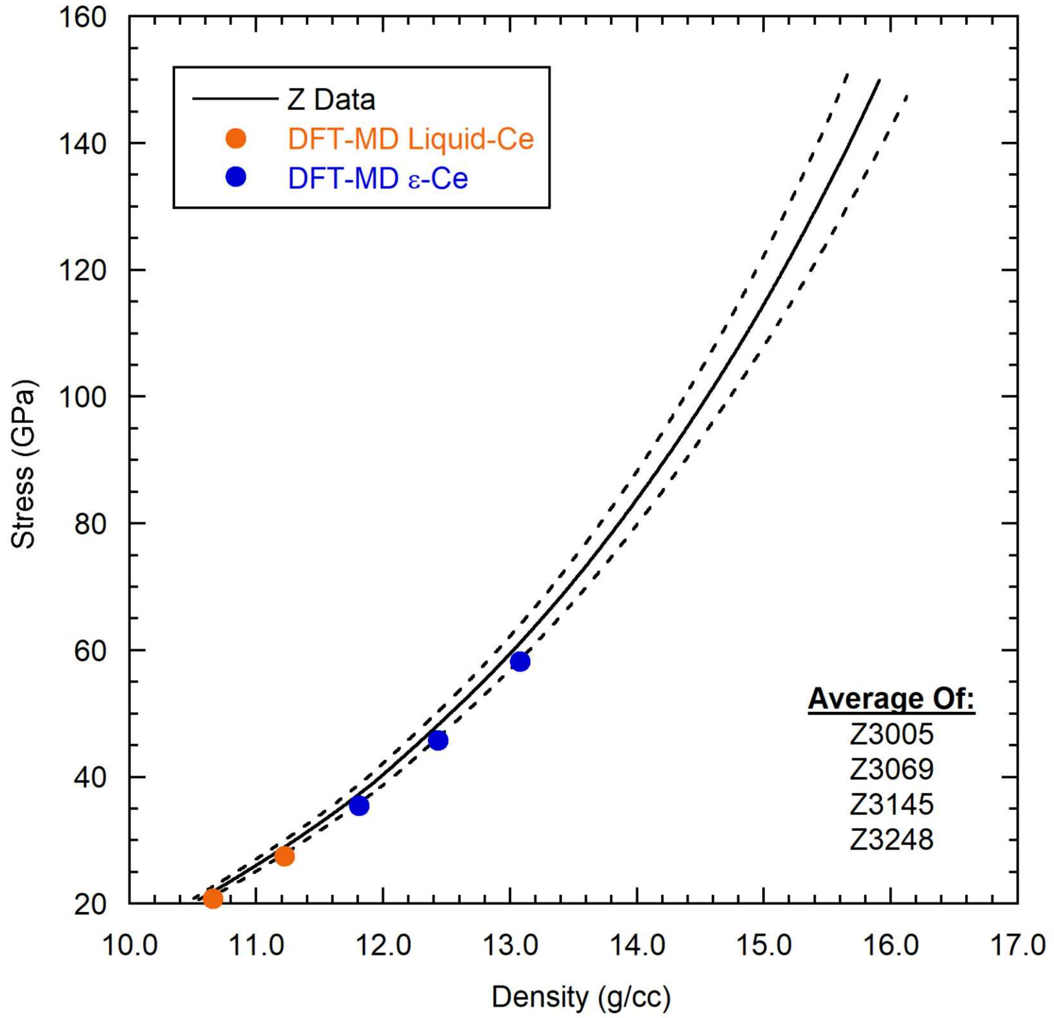
Through the generalized Hooke's law,

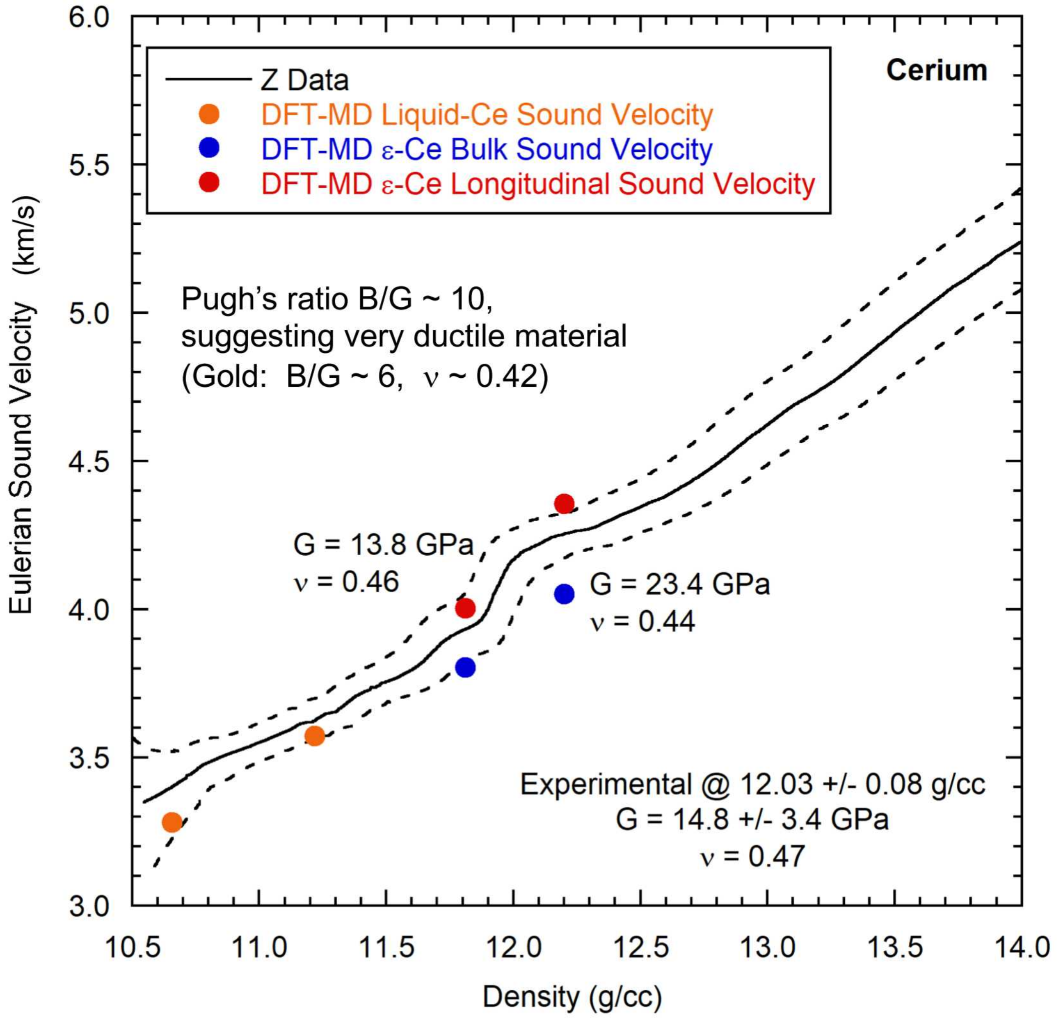
$$\sigma_{ij} = C_{ijkl} e_{kl}$$

application of the first strain tensor and differencing the stresses generated with dilation and contraction (δ = +/- 0.01) yields C_{11}, C_{12}, C_{13}, and C_{44}; the second strain tensor yields C_{13}, C_{13}, C_{33}, and C_{66}.

*DFT-MD: GGA+U with 54 atoms, 12 electrons in the valence, Γ-centered 2x2x2 Monkhorst-Pack grid (8 k-points)

DFT-MD simulations agree with experimental observations of stress-density and sound velocities





Assuming a polycrystalline solid, Voigt-Reuss-Hill averages are quoted for moduli and sound speeds

Summary

- An experimental observation of solidification has been detected through the recovery of strength on dynamic compression: elastic wave velocity observed during ramp compression of liquid.

- DFT-MD simulations exhibit spontaneous freezing of cerium on the experimental isentrope at a pressure close to the experimental observation.

- The stress, density, longitudinal and bulk sound velocities, and shear modulus and Poisson ratio are in agreement between calculations and experimental observations.

- Dynamic Solidification of Cerium occurs at or near the equilibrium melt line on the compression isentrope from an initially liquid state on nano-second time-scales.