TPa Ramp Compression and *In Situ* X-ray Diffraction of Forsterite (Mg_2SiO_4)

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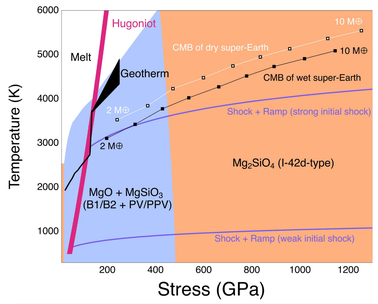
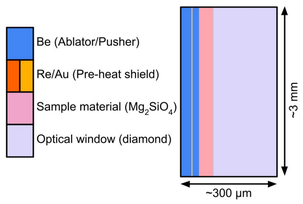
I. Introduction and Motivation

Forsterite (Mg_2SiO_4) is representative of the compositions expected in large, rocky, extra-solar planets. Under mantle conditions expected for smaller planets (~<4 ME) Mg_2SiO_4 will decompose to $MgSiO_3$ + MgO. Theoretical calculations predict that above ~500 GPa the two phases recombine into an as-yet unobserved Mg_2SiO_4 phase which could be a major constituent of super-Earth mantles for >4 ME planets.

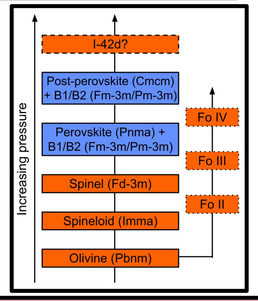


Left: theoretical phase diagram of Mg_2SiO_4 (modified from [1]). Above ~500 GPa, periclase and post-perovskite are predicted to recombine into an I-42d-type structure. CMB-conditions of wet and dry super-Earths (rocky planets with Earth-like core-to-mantle ratios) are marked in black and white respectively. Shock + ramp paths and the Hugoniot are marked in color. Blue represents $MgSiO_3$ + MgO stoichiometry, while orange represents Mg_2SiO_4.



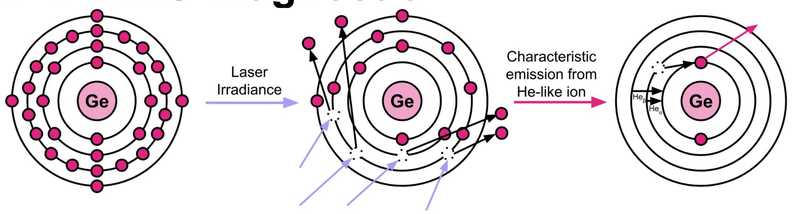
Left: sample target used in these experiments. The heat shield also protects image plates from X-rays from the ablation plume.

Right: structural progression of Mg_2SiO_4. Metastable phases = dotted, predicted = dashed. MgO + $MgSiO_3$ assemblages in blue.

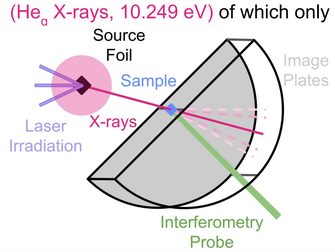


Traditional high-pressure techniques are not able to determine the structure of geologic materials at the extreme conditions of super-Earth mantles. We use the National Ignition Facility (NIF) to ramp-compress forsterite and pair *in-situ* diffraction data with theoretical calculations to characterize the high-pressure polymorphism of this material.

II. TARDIS Diagnostic



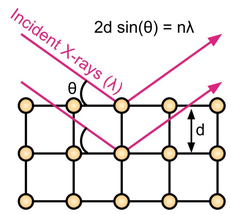
Above: X-rays are generated by irradiation of a Ge foil. 12-24 high-powered lasers strip electrons from the Ge atom until it reaches a He-like state (two remaining electrons). Electronic transitions to the ground state then isotropically release quantized photons (He_α X-rays, 10.249 eV) of which only a small fraction (~0.0001 steradians) are used [2].

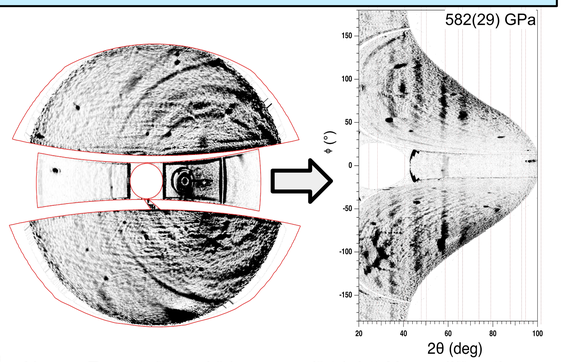


Left: TARget Diffraction *In-Situ* (TARDIS) diagnostic at the NIF. Two flat, semi-circular and one curved, rectangular image plates form a taco-shape. Mg_2SiO_4 sample (synthetic, ~0% porosity, untextured polycrystals synthesized at U. Tokyo) is affixed opposite the curved rectangular plate and compressed. Incident X-rays record diffraction data on the image plates and an interferometry probe shines through an aperture.

The TARDIS diagnostic with Ge backlighters is used to characterize the *in situ* structure of the dynamically compressed synthetic forsterite samples by recording X-ray diffraction on three photo-sensitive image plates.

Below: Schematic of X-ray diffraction. Incident X-rays interact with repeated planes of atom in the crystal lattice, diffracting at a specific angle based on the known incident wavelength.

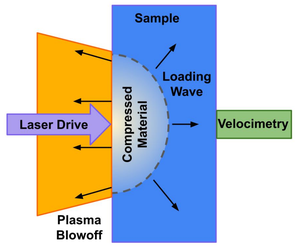


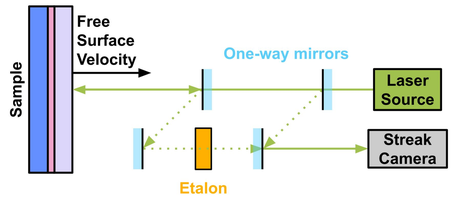


Above: Raw (left) and "de-warped" (right) X-ray diffraction image plates. De-warping is a mathematical transformation constrained by known geometry and refined by the positions of Laue spots resulting from the single-crystal diamond window.

III. Compression and Stress Determination

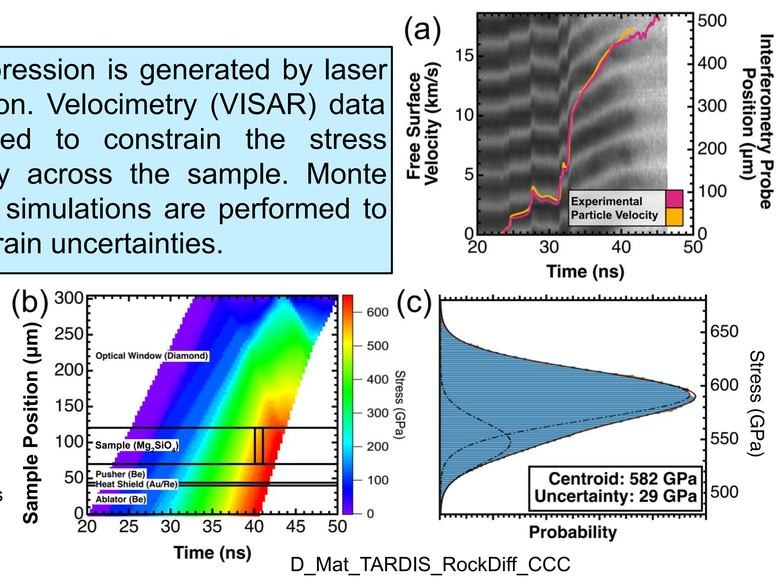
Right: the front surface of the target package is ablated by focusing 16 high-power laser beams of 3.545 kJ each onto it. This generates a plasma, whose blowback, by conservation of momentum, introduces a loading wave forwards, into the sample.Laser interferometry is used to measure the wave profile of the rear surface of the target package.





Above: a laser shines off of the back surface of the target. A series of two-way mirrors splits the reflected beam into two. One beam passes through an etalon, which slows it slightly. When the light is recombined, this results in a phase difference, which is recorded by a streak camera.

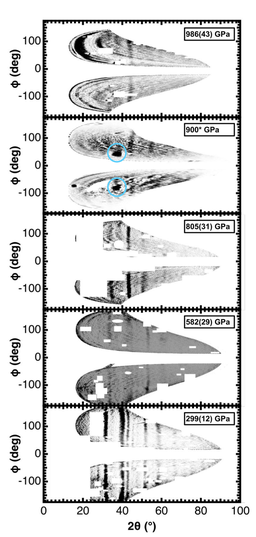
Compression is generated by laser ablation. Velocimetry (VISAR) data is used to constrain the stress history across the sample. Monte Carlo simulations are performed to constrain uncertainties.

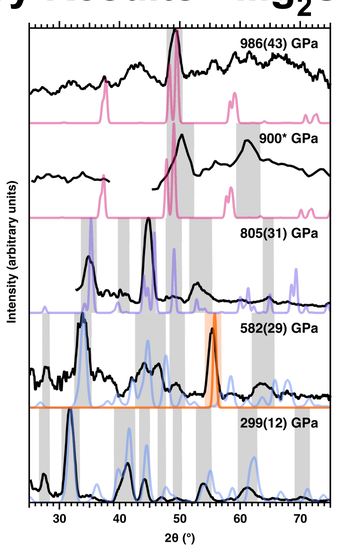


Right: (a) a VISAR interferogram (background) and the processed velocity histories (magenta, orange). (b) the method of characteristics is used to backwards propagate the velocity history through the sample, creating a stress map over the duration of the experiment. The pulse duration and sample position are marked in black. (c) a series of Monte Carlo simulations are used to constrain uncertainty in stress considering the uncertainties in experimental and equation of state parameters.

D_Mat_TARDIS_RockDiff_CCC

IV. Preliminary Results - Mg_2SiO_4



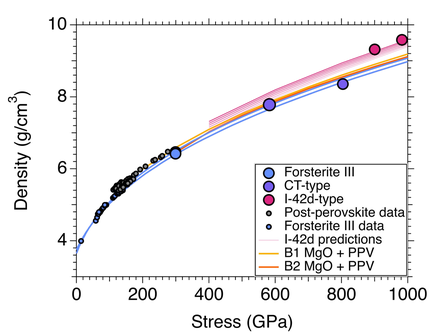


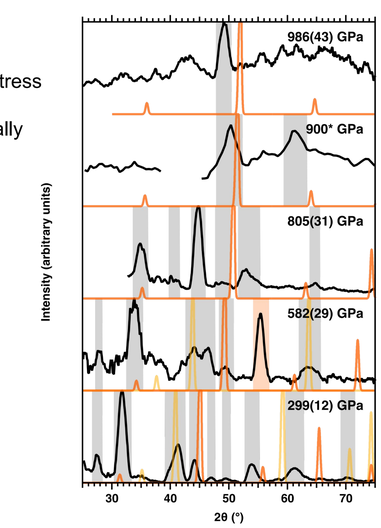
Far left: 2D (dewarped) diffraction data collected. *900 GPa is a nominal stress; complete stress analysis is ongoing. Blue circles mark suspected diamond diffraction spots.

Left: Integrated lineouts of each diffraction image. Shaded areas represent areas of high confidence in the data peaks. In color are the following phase assignments: I-42d-type, calcium-titanate type, Be (pusher/ablator), and forsterite III.

Right: Integrated lineouts of each diffraction image against potential decomposition products. In color are MgO (B1) and MgO (B2). No evidence for decomposition is found.

Below: stress-density relationship for the high-stress phase assignments in the left figure. Fo III and CT-type lattices were fit to the data or theoretically calculated. I-42d densities were theoretically calculated in [1]. Data from [3-6].





V. Conclusions

Diffraction peaks are observed in ramp-compressed Mg_2SiO_4 between 299-986 GPa, indicating extensive stability of crystalline phases (in contrast to amorphization observed at ~100 GPa along the Hugoniot.)

At 299-805 GPa, diffraction data are qualitatively consistent with formation of a metastable post-spinel phase such as Fo III.

Between 805-986 GPa, observed diffraction patterns are variable, indicating the formation of multiple phases across this stress range or large changes in texture from shot to shot. At these stresses, diffraction data cannot be assigned to the MgO B2 phase, indicating that decomposition does not occur on the ramp-loading timescale.

Diffraction data at 900 GPa are not consistent with any known post-spinel phases, indicating that it adopts one or more unknown crystal structures. At 986 GPa, preliminary analyses of the diffraction data are consistent with formation of the predicted I-42d-type phase.

VI. References + Acknowledgements

[1] Umemoto, K. et al. EPSL **478**, 40-45 (2017)
[2] Coppari, F. et al. Rev. Sci. In. **90**, 125113 (2019).
[3] Finkelstein, G. et al. Am. Min. **99**, 35-43 (2014).
[4] Sakai, T. et al. Sci. Rep. **6**, 22652 (2016).
[5] Guignot, N. et al. EPSL **256**, 162-168 (2007).
[6] Speziale, S. et al. JGR **106**, 515-528 (2001).

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