NIF_DS Proposal (P000559) on Chemistry under Extremes (XCHEM): Metallization of Ionic and Covalent Solids - NaCl

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Novel states of matter under extreme conditions

Condensation  Bonding  Packing  Ionization

Metallic H₂
Ice X  BC₈ C  Al Electride

Yoo, MRS Bul. (2017)

PΔV_{NN}

0 1 atm 1 MPa 2 3 1 GPa 1 Kbar 4 5 6 1 Mbar 7 8 9 10 10 log P 100 K 0.1 eV 1 eV 10 eV 1 keV
Significant questions on the chemistry at extreme PT

- What are novel states, structures and properties of materials at extreme PT?
- How does chemical bonding evolve and chemistry occur in dense solids at compression energy of core electron?
- What are the governing principles of chemistry in dense solids under extreme conditions? How does kinetics interplay?
- How does core-electrons contribute to the chemistry? When and how are dense solids become metallic?

CsI and Xe becomes the same hcp with “indistinguishable” Cs, I and Xe

Pressure evolution of chemical bonding in dense solids

The pressure-induced structural changes in solids can be predicted based on first principles in physics and chemistry; computational materials design.

Ice VII, Pn3m

Single-bonded “polymeric” nitrogen frameworks

Wang et al. PRL (2012)

Tomasino et al. PRL (2014)
High-PT research windows novel state, structure, bonding, and transformation of material that may occur in deep interiors of the Earth and Giant planets.

New “core-electron” chemistry at TPa

Exotic Conducting States predicted:
- Metallic H2
- Metallic diamond
- Metallic salt
- Metallic He
- Electrides
- Superionic water
- Etc. etc.
Goal: Exploit novel metallic phases with predominantly ionic structure predicted for NaCl at NIF-accessible pressure

*Chen & Ma, EPL 100, 26005 (2012).*

<table>
<thead>
<tr>
<th>Structure</th>
<th>Pressure</th>
<th>Charge transfer</th>
</tr>
</thead>
<tbody>
<tr>
<td>cF8</td>
<td>0 GPa</td>
<td>0.87</td>
</tr>
<tr>
<td>cP2</td>
<td>50 GPa</td>
<td>0.83</td>
</tr>
<tr>
<td>cP2</td>
<td>300 GPa</td>
<td>0.77</td>
</tr>
<tr>
<td>cC8</td>
<td>350 GPa</td>
<td>0.75</td>
</tr>
<tr>
<td>cP2</td>
<td>450 GPa</td>
<td>0.75</td>
</tr>
<tr>
<td>018</td>
<td>600 GPa</td>
<td>0.74</td>
</tr>
<tr>
<td>018</td>
<td>640 GPa</td>
<td>0.73</td>
</tr>
<tr>
<td>018</td>
<td>650 GPa</td>
<td>0.74</td>
</tr>
<tr>
<td>018</td>
<td>680 GPa</td>
<td>0.74</td>
</tr>
<tr>
<td>018</td>
<td>685 GPa</td>
<td>0.75</td>
</tr>
<tr>
<td>018</td>
<td>700 GPa</td>
<td>0.75</td>
</tr>
<tr>
<td>018</td>
<td>900 GPa</td>
<td>0.74</td>
</tr>
</tbody>
</table>

Chen & Ma, EPL 100, 26005 (2012).

- Crystal structure of extended ionic sublattice favors conductivity
- This unusual state is predicted for many other systems at inaccessible pressures; thus, the present results will have broad implications
- New chemistry for prototypical ionic solid NaCl will have a high impact
**Approach:** identify new NaCl phases with x-ray diffraction and metal-insulator transitions from VISAR reflectivity

10-quad direct-drive configuration

- Blue: sample drive: 4 quads
- Red: x-ray source drive: 6 quads

0 log growth: no optics damage

**TARDIS diffraction diagnostic**

- Drive: (30 ns pulse)
- X-ray source: (2 ns pulse)
- 'Catcher'
- Four SiO₂ windows (1st two shatter)

**Diagnostic configuration**

<table>
<thead>
<tr>
<th>DIM</th>
<th>diagnostic</th>
<th>priority</th>
</tr>
</thead>
<tbody>
<tr>
<td>90-315</td>
<td>VISAR</td>
<td>1</td>
</tr>
<tr>
<td>90-78</td>
<td>GXD/Supersnout</td>
<td>1</td>
</tr>
<tr>
<td>fixed</td>
<td>SPIDER, NSS, FFLEX, DANTE</td>
<td>2</td>
</tr>
</tbody>
</table>

X-ray diffraction at the National Ignition Facility, Rygg et al, RSI, submitted

Metastability of Diamond Ramp-Compressed to 2 TPa, Jenei et al, planned submission by Dec 2019

Structural Complexity in Dense Magnesium, Gorman et al., planned submission by Dec 2019
Shot plan: Ramp compression of NaCl to 1 TPa

<table>
<thead>
<tr>
<th>Shot number</th>
<th>Pressure in GPa</th>
<th>Primary phase</th>
<th>Secondary phase</th>
<th>Optical reflectivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>1(Q1)</td>
<td>400</td>
<td>cP2 (B2)</td>
<td>oC8</td>
<td>insulator</td>
</tr>
<tr>
<td>2(Q2)</td>
<td>600</td>
<td>oC8</td>
<td></td>
<td>metallic</td>
</tr>
<tr>
<td>3(Q3)</td>
<td>700</td>
<td>oI8</td>
<td>oC8, oP16</td>
<td>metallic</td>
</tr>
<tr>
<td>4(Q4)</td>
<td>900</td>
<td>oP16</td>
<td></td>
<td>metallic</td>
</tr>
</tbody>
</table>

- Pressure increase shot to shot allows incremental improvements to the diagnostic and target design to optimize signal levels.
- As we access more complex structures at higher pressure, we will likely transition to lower Z X-ray ablators like Cu (8.4 keV) or Fe (6.7 keV)
Summary: Expected results and significance

Expected Results:

• Diffraction data (via TARDIS) to show the pressure-induced phase transformations in NaCl to 1 TPa, to explore the predicted transitions $cF8 \rightarrow cP2 \rightarrow oC8 \rightarrow oI8 \rightarrow oP16$ and develop new theoretical structure models.
• Optical reflectivity data (via VISAR) to reveal the predicted insulator-metal transition in NaCl.
• DFT-based electronic structure calculations to reveal the pressure-evolution of chemical bonding of ionic NaCl to novel metallic state.

Significance of Results:

• Uncover the novel state, structure and chemistry of solids under extreme conditions of deep planetary interiors
• Examine the pressure-evolution of crystal structure and chemical bonding of solids at compression energies of core-electrons (1-100 eV)
• Gain new insights into the fundamental principles governing core-electron chemistry – not been experienced
• Experiments will stimulate further investigations on different types of solids (quantum solids, novel gas solids, molecular solids, etc.), establishing new chemistry rules at extremes