Developing a Data Format and Repository for Sharing First-principles Defect Calculations
What can we learn from defect calculations?

- We can learn about the defect chemistry, which is important in applications such as: thermoelectric, photovoltaic, etc.
- First-principles defect calculations are now mature, are routinely done and provides useful insights

**npj Computational Materials**

First-principles calculation of intrinsic defect chemistry and self-doping in PbTe
Anuj Goyal\textsuperscript{1,2}, Prashun Gorai\textsuperscript{1,2}, Eric S. Toberer\textsuperscript{1,2} and Vladan Stevanović\textsuperscript{1,2}

**Chemistry of Materials**

Thermoelectric Performance and Defect Chemistry in n-Type Zintl KGaSb$_4$
Brenden R. Ortiz\textsuperscript{a}, Prashun Gorai\textsuperscript{b}, Vladan Stevanović\textsuperscript{1,4} and Eric S. Toberer\textsuperscript{a,†}

**Chemistry of Materials**

Searching for “Defect-Tolerant” Photovoltaic Materials: Combined Theoretical and Experimental Screening
Riley E. Brandt\textsuperscript{a,†}, Jeremy R. Poindexter\textsuperscript{a}, Prashun Gorai\textsuperscript{b,†}, Rachel C. Kurchin\textsuperscript{a}, Robert L. Z. Hoye\textsuperscript{a,‡}, Lea Nienhaus\textsuperscript{a}, Mark W. B. Wilson\textsuperscript{a,‡}, J. Alexander Polizzotti\textsuperscript{a}, Raimundas Žaltauskas\textsuperscript{a}, Lan C. Lee\textsuperscript{a,‡}, Judith L. MacManus-Driscoll\textsuperscript{a}, Moungi Bawendi\textsuperscript{a}, Vladan Stevanović\textsuperscript{a,‡} and Tonio Buonassisi

**ELSEVIER**

A computational framework for automation of point defect calculations
Anuj Goyal\textsuperscript{a,†}, Prashun Gorai\textsuperscript{a,‡}, Haowei Peng\textsuperscript{a}, Stephan Lany\textsuperscript{a}, Vladan Stevanović\textsuperscript{a,‡}

Editor's Choice

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Read more at www.sciencedirect.com/locate/commatsci
Why do we need a defect data format and repository?

- The problem is two fold:
  - No standard data format for sharing defect energetics, requires cumbersome plot digitization to grab data
  - Partly due to the lack of a shareable data format, a data repository of defect calculations is missing that would otherwise be immensely useful
A Simple Example of a Defect Diagram: PbTe

\[ \Delta H_{D,q}(E_F, \mu) = [E_{D,q} - E_H] + \sum_i n_i \mu_i + qE_F + E_{corr} \]

- Defect Formation Energy
- Defect Supercell
- Host Supercell
- Chemical potentials from phase stability
- Electron chemical potential
- Finite size corrections

Rocksalt PbTe

Pb-rich and Te-rich

Phase Stability

A. Goyal, P. Gorai, E. Toberer, V. Stevanović, npj Comp. Mat. 42 (2017)
What Should the Defect Data Format Look Like?

Defect Data Format

<table>
<thead>
<tr>
<th>Energy</th>
<th>Type</th>
<th>Site</th>
<th>Charge</th>
<th>Label</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.52</td>
<td>V</td>
<td>Te</td>
<td>2</td>
<td>V_Te_2</td>
</tr>
<tr>
<td>0.86</td>
<td>I</td>
<td>Pb</td>
<td>2</td>
<td>I_Pb_2</td>
</tr>
<tr>
<td>1.05</td>
<td>Pb</td>
<td>Te</td>
<td>1</td>
<td>Pb_Te_1</td>
</tr>
<tr>
<td>1.65</td>
<td>V</td>
<td>Pb</td>
<td>-2</td>
<td>V_Pb_-2</td>
</tr>
<tr>
<td>2.36</td>
<td>Te</td>
<td>Pb</td>
<td>0</td>
<td>Te_Pb_0</td>
</tr>
<tr>
<td>2.77</td>
<td>I</td>
<td>Te</td>
<td>1</td>
<td>I_Te_1</td>
</tr>
</tbody>
</table>

Band gap

Defect Diagram

Pb-rich
What Should the Defect Data Format Look Like?

Defect Diagram

**Pb-rich**

\[ \Delta H, \Delta g \text{ (eV)} \]

0

0.0

0.1

0.2

E\(_F\) (eV)

T\(_{Pb}\)

V\(_{Pb}\)

Pb\(_{Te}\)

I\(_{Pb}\)

V\(_{Te}\)
Examples of More Complicated Defect Diagrams

Defect physics of ZnSiP$_2$

Gorai et al., Energy Environ. Sci. 9, 1031 (2016)

Defect Chemistry in n-type Zintl KGaSb$_4$

Ortiz et al., Chem. Mater. 29, 4523 (2017)

**p-type doping in rocksalt ZnO**

- Definitely more complicated defect diagrams, but they can be reproduced using the same minimal defect data format. Check out: [Citrine Webpage](https://www.citrine.com/)

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### Figure 3

- **Point 3**

### Figure 4

- **Native defects**
  - $V_{\text{Zn}}$
  - $V_{\text{P}}$
  - $V_{\text{Si}}$
  - $P_{\text{Si}}$
  - $\text{Si}_{\text{Zn}}$
  - $\text{Zn}_{\text{Si}}$

- **$\Delta H_{D,q}$ (eV)**
  - $E_F$ (eV)

- **$\Delta E_{\text{acc}}$ (eV)**
  - $E_F$ (eV)

- **$\Delta E_{\text{don}}$**

- **Defect formation enthalpies** (e.g., $V_{\text{Ga}(4)}$, $V_{\text{K}}$)

- **Examples of More Complicated Defect Diagrams**

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**Notes:**

- The phase diagram (a) shows the Fermi level ($E_F$) and mid-gap which is shown as a vertical dotted line. The relevant defect states are shallow and appear as shoulders of the band edges.
- There is no evidence of mid-gap states that could trap carriers and detrimentally affect carrier transport.
- The Fermi levels in Regions 1, 2, and 3 (labeled R1, R2, and R3) with different formation enthalpies ($\Delta H_{D,q}$) of 7 different point defects (vacancies and antisites) in all possible charge states ($Zn\text{SiP}_2$, without defects, and $Zn\text{SiP}_2$).
- The Fermi level in the mid-gap region results in intrinsic p-type material until 275 $°C$.
- The Seebeck coefficient behavior ($S$) and power factor ($\alpha$) across the temperature range ($T$) are shown.
- The density of states (DOS) is shown for KGaSb$_4$.
- Representative Rietveld refinement (red) of the KGaSb$_4$ crystal structure is shown for comparison. Rietveld indicates that material is >98% phase pure.

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**References:**

- Gorai et al., Energy Environ. Sci. 9, 1031 (2016)
- Ortiz et al., Chem. Mater. 29, 4523 (2017)
- Citrine Webpage
Looking Ahead: Building a Public Repository

• **Test the parser** for more complicated defect diagrams

• Once parser is ready, **build a public repository** on Citrine for sharing defect diagrams

• Encourage the community to use the **data format for sharing** - example, supplementary data for publication

• Once the repository has a sizable number of materials, use **data informatics tools** on Citrine to tease out trends etc.

Thank you!