

# High-Pressure Synthesis of Novel Binary Intermetallics

J. P. S. Walsh<sup>1\*</sup>, S. M. Clarke<sup>1</sup>, K. M. Powderly<sup>1</sup>, A. D. Tamerius<sup>1</sup>, Y. Meng<sup>2</sup>, S. D. Jacobsen<sup>3</sup> and D. E. Freedman<sup>1</sup>

<sup>1</sup>Department of Chemistry, Northwestern University, Evanston, Illinois 60208, United States

<sup>2</sup>HPCAT, Geophysical Laboratory, Carnegie Institution of Washington, Argonne, Illinois 60439, United States

<sup>3</sup>Department of Earth and Planetary Sciences, Northwestern University, Evanston, Illinois 60208, United States

Keywords: solid-state, synthesis, laser heating

\*e-mail: james.walsh@northwestern.edu

The pressure scale of the universe spans roughly fifty orders of magnitude, and yet most chemistry is carried out under atmospheric pressures. We already know that under the colossal pressures found in the center of stars, chemistry as we know it makes way for nuclear fusion—but what happens at intermediate pressures? Do chemical syntheses follow the same rules under planetary core pressures that they do under standard pressures? And in what ways do electronic and magnetic structure respond as atoms are forced closer to each other? We can begin to answer such questions using handheld devices known as diamond anvil cells (DACs), which are able to provide access to static pressures on the order of millions of atmospheres—the same pressures found deep within our planet.

This talk will discuss the use of the DAC as a reaction vessel for the synthesis of novel binary intermetallic compounds that are unattainable under ambient pressures, with a focus on *in situ* XRD experiments performed at the HPCAT beamlines at the Advanced Photon Source. The combination of laser heating methods with highly focused and high-brilliance radiation is ideal for the real-time detection of new intermetallic phases forming under extreme conditions of temperature and pressure, and has led to the discovery and characterization of a number of new compounds that cannot be synthesized by traditional solid-state techniques [1–5]. The unprecedented bonding and structure exhibited by the compounds we have synthesized hint at the enormous potential of high-pressure synthesis as a route toward materials with exotic emergent properties.

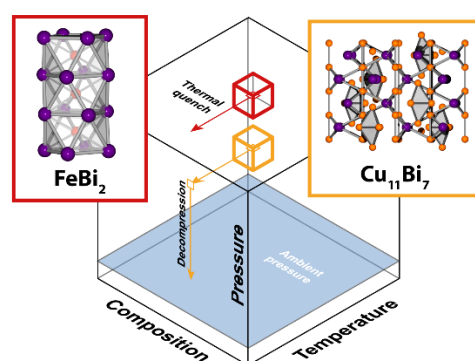


Figure 1. Schematic representation of the synthetic phase space opened up by the use of high pressure as an additional thermodynamic variable, allowing for the discovery of novel intermetallic compounds such as FeBi<sub>2</sub> and Cu<sub>11</sub>Bi<sub>7</sub>.

**Acknowledgments:** This research was initiated with funding from DARPA (W911NF15100069) and executed with funding from AFOSR (FA9550-14-1-0358), AFOSR-MURI (FA9550-17-1-0247), and DOE (DE-SC0018092). J.P.S.W. is supported by the Innovative Initiatives Incubator (I3) at Northwestern University. This work used resources of the APS (DOE: DE-AC02-06CH11357), with additional beamtime support from the Carnegie/DOE Alliance Center (CDAC). HPCAT operations are supported by DOE-NNSA (DENA0001974) and DOE-BES (DEFG0299ER45775), with partial instrumentation funding by NSF.

- [1] Clarke, S. M., Walsh, J. P. S., Amsler, M., Malliakas, C. D., Yu, T.; Goedecker, S., Wang, Y., Wolverton, C., Freedman, D. E., *Angew. Chem. Int. Ed.* **2016**, *55*, 13446–13449.
- [2] Clarke, S. M., Amsler, M., Walsh, J. P. S., Yu, T., Wang, Y., Meng, Y., Jacobsen, S. D., Wolverton, C., Freedman, D. E., *Chem. Mater.* **2017**, *29*, 5276–5285.
- [3] Powderly, K. M., Clarke, S. M., Amsler, M., Wolverton, C., Malliakas, C. D., Meng, Y., Jacobsen, S. D., Freedman, D. E., *Chem. Commun.* **2017**, *53*, 11241–11244.
- [4] Walsh, J. P. S., Clarke, S. M.; Meng, Y.; Jacobsen, S. D.; Freedman, D. E. *ACS Cent. Sci.* **2016**, *2*, 867–871.
- [5] Walsh, J. P. S., Freedman, D. E., *Acc. Chem. Res.* **2018**.