Computational challenges in the search for better superconducting hydrides

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Structure prediction, and the theoretical computation of reliable superconducting transition temperatures, have undoubtedly played a major role in the discovery of novel high temperature superconductivity in dense hydrides.[1] While the field has delivered room temperature superconductivity,[2] the technological relevance will be limited while the phenomenon is restricted to extremely high pressures. Furthermore, the number of experimental research groups that can study the properties of these compounds at megabar pressures is limited, restricting the potential scientific impact. Rightly, the field is focusing on identifying compounds that superconduct at high temperatures, but much lower pressures. But there are considerable obstacles to progress. The number of theoretical candidates far exceed those experimentally confirmed, suggesting more attention should be paid to predicting synthesisability. It is becoming clear that metastability favours high temperature superconductivity, but how should we choose from the multitude of metastable candidates? Experimentally determined structures are frequently not found to be dynamically stable in static calculations, but full dynamics is computationally expensive, and difficult to account for in high throughput searches. At the same time, it is not clear how to compute superconducting transition temperatures in highly dynamic systems. So far, most attention has been paid to perfect crystals. Doping and deviation from perfect stoichiometry, and well as defects (both point, and extended, such as grain boundaries and interfaces) are likely to be important to the detailed properties of these materials. Finally, as we turn to exploring a broader range of compounds, in the ternaries and beyond, structure prediction becomes more challenging, not least in terms of the management of the quantities of data generated, and the computation of large numbers of superconducting transition temperatures. I will show some recent results which go some way to addressing this.

[1] Pickard, Chris J., Ion Errea, and Mikhail I. Eremets. "Superconducting hydrides under pressure." *Annual Review of Condensed Matter Physics* 11 (2020): 57-76.

[2] Snider, Elliot, Nathan Dasenbrock-Gammon, Raymond McBride, Mathew Debessai, Hiranya Vindana, Kevin Vencatasamy, Keith V. Lawler, Ashkan Salamat, and Ranga P. Dias. "Room-temperature superconductivity in a carbonaceous sulfur hydride." *Nature* 586, no. 7829 (2020): 373-377.

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