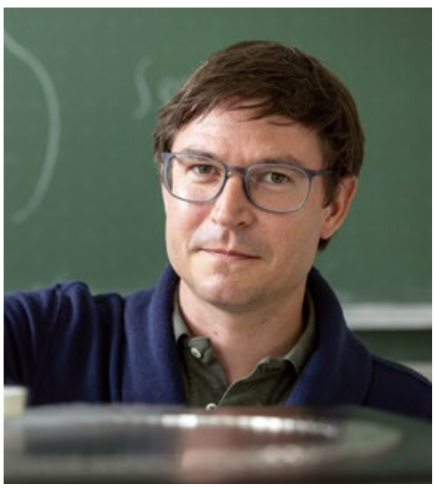
Christoph Heil

In-Silico Design of Novel Superconductors

Superconductors, which exhibit zero electrical resistivity, are extremely desirable for numerous applications. They necessitate cooling below their so-called critical temperature, generally extremely low, thus incurring high cooling expenses. Our research employs advanced ab-initio approaches to engineer superconductors capable of operating at significantly higher temperatures to enable broad applicability.



Christoph Heil

holds an assistant professorship position at the Institute of Theoretical and Computational Physics.

He started working in the field of superconductivity during his PhD studies more than 10 years ago. Today, his research primarily revolves around the accurate ab-initio modeling and innovative design of conventional superconductors.

Source: Lunghammer – TU Graz

The history of superconductivity can be traced back over more than a century to the discovery of this phase of matter in mercury cooled by liquid helium. Since then, scientists have been intrigued by this exotic phenomenon, and this has led to a quest for materials with ever higher critical temperatures (T_c's). While early investigations were primarily driven by academic curiosity, certain superconductors have found niche applications in technology and industry. Yet their practical application remains limited due to the necessity of expensive cooling systems. Presently, cuprates represent the highest-T_c materials under ambient pressure, with T_c values exceeding the boiling point of nitrogen (77 K). However, their poor processability poses challenges for widespread applications.

The prospect of ambient temperature superconductors has spurred intensive research efforts due to their transformative potential in energy, medicine, and transportation. High-performance computing and first-principles methods have revolutionized superconductivity research, enabling theoretical predictions to guide experimentation towards higher T_c materials at an unprecedented pace. Recent breakthroughs have brought about the realization of near-room-temperature superconductivity in hydrogen-rich materials under extreme pressures, exemplified by LaH_{10}. While achieving higher T_c values remains a tantalizing prospect, the emphasis has shifted towards stabilizing materials at lower pressures while maintaining T_c's above the boiling point of nitrogen – a critical focus of ongoing research efforts.

In silico modeling of a new conventional high T_c superconductor is a multi-stage process and the starting point is to identify promising candidate structures that are stable at lower pressures. To determine which compositions and structures are stable, one needs to find, as a function of composition, those structures whose enthalpy of formation is lower than that of any other structure or linear combination of structures resulting in the proper composition. The convex hull is a set of lines connecting these stable structures, as all other structures have enthalpies lying above these lines and combining convex hulls for various pressures then allows to construct a phase diagram, providing extensive information which structures are thermodynamically stable at which pressures.

These calculations are performed using density functional theory and encompass the computation of total energies for a few thousand (in the case of a binary system) to several hundred thousand (for ternary or higher systems) different structures. To make such crystal structure predictions

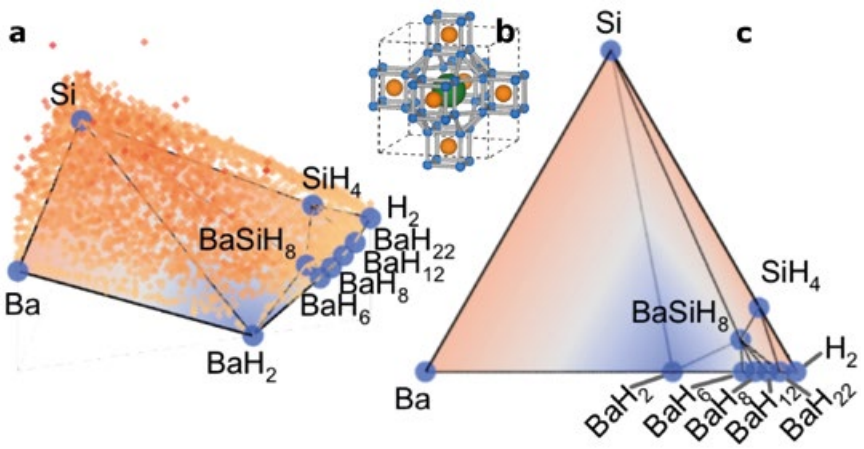


Figure 1: Results of a crystal structure prediction calculation for the Ba-Si-H system at a pressure of 100 GPa. (a) Side view of the convex hull with markers indicating individual structures taken into account. (c) Top view of the convex hull, showing only stable phases for clarity. (b) Crystal structure of high-T_c $BaSiH_8$.

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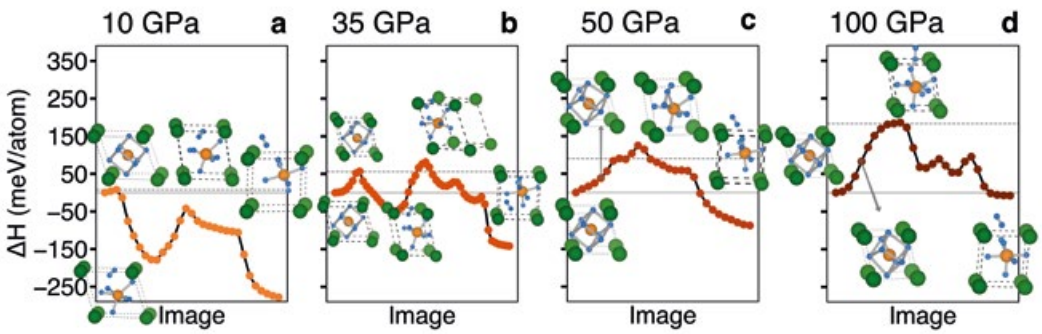

Figure 2: Assessment of kinetic energy barrier protecting metastable high-T_c $BaSiH_8$ from decomposing at various pressures.

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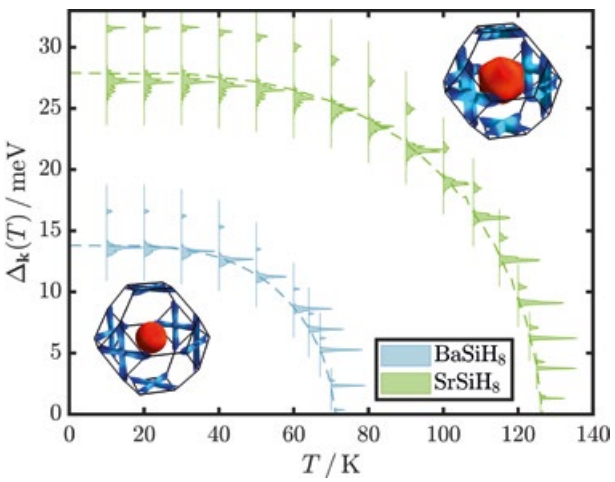

Figure 3: Superconducting properties of $BaSiH_8$ and $SrSiH_8$ at 5 and 30GPa, respectively, as obtained from the solution of the Migdal-Eliashberg theory. T_c is defined as the temperature for which the superconducting gap function Δ vanishes.

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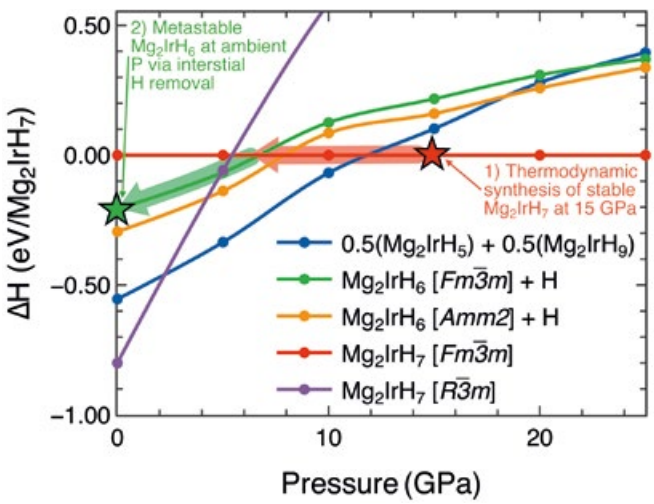

Figure 4: Proposed synthesis route to obtain high-T_c Mg_2IrH_6 at ambient pressure, starting from Mg_2IrH_7 at 15 GPa.

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feasible, clever methods are employed that use concepts found in nature like evolutionary survival-of-the-fittest and swarm algorithms, enabling researchers to scan through a vast compositional phase space efficiently.

Recently, the community also started to try and identify promising metastable structures, i.e., structures that are not on the convex hull but still experimentally feasible. In fact, many functional materials are achieved through carefully designed synthesis paths, resulting in metastable states and identifying viable pathways when proposing new metastable structures will be an important topic in the ensuing years.

The subsequent step involves accurate determination of superconducting properties, particularly for conventional superconductors enabled by electron-phonon coupling. The Migdal-Eliashberg theory provides a theoretical framework for understanding the microscopic mechanisms underlying superconductivity and modern high-performance computing clusters allow to overcome the difficult problem of solving these Migdal-Eliashberg equations numerically to determine properties such as T_c.

To provide examples from our own research: Motivated by the discovery of near-room-temperature SC in the clathrate hydride LaH_{10}, we investigated ternary systems containing La and H, leading to the prediction of a new ternary hydride, $LaBH_8$, which was the first example of a conventional superconductor with a T_c = 126 K, i.e. exceeding the boiling point of N_2, that should remain dynamically stable down to 35 GPa. Using the XYH_8 template as the next starting point, we further identified two silicides that are an improvement on LaBH8: $SrSiH_8$ with a dynamical stability pressure p_c of 27 GPa and a T_c of 127 K, and even more remarkable, $BaSiH_8$ that is dynamically stable down to pressures of 3 GPa with a critical temperature of 71 K. The first experimental validations of these ternary superhydrides have been carried out, and successful synthetizations of $LaBeH_8$, $LaBH_8$, and $BaSiH_8$ reported.

Continuing on this trajectory, our recent collaborative efforts have led to the discovery of a metastable cubic phase of Mg_2IrH_6 with a T_c exceeding 100 K at ambient pressures. This brings us closer to the ultimate goal of uncovering ambient-pressure superconductors with T_c values surpassing the boiling point of nitrogen, thus paving the way for practical applications across various technological domains.