Probing Structural Changes in the Properties of Matter Using Extreme Conditions

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The study of materials with novel structures or properties is of increasing importance for our society. One approach to synthesize novel compounds is by using a combination of high pressures and high temperatures. These extreme conditions can be achieved using diamond anvil cells and laser-heating, respectively. X-ray powder diffraction is a powerful probe to study the crystal structure of newly synthesized compounds or structural phase transitions. These measurements can be conducted *in situ* under extreme conditions with pressures up to more than 100 GPa and temperatures exceeding thousands of Kelvin. However, these conditions limit the data quality and a structure solution from these experiments alone is often impossible. One way to overcome this problem is by supplementing the experimental studies with theoretical predictions for energetically favorable crystal structures. The evolutionary algorithm USPEX[1], as well as additional calculations in the framework of density functional theory, has been used in this project to guide the structure solution of novel compounds or unknown high-pressure phases observed after a structural phase transition.

Here we present two projects using this combination of theoretical predictions and experiments. The first project concerns nitrogen-rich binary materials, where the group of pernitrides, that have a composition of MN_2 (with M being a metal), is known to have an ultrahigh bulk modulus. This has for instance been observed in IrN_2 with K_0 = 428(12) GPa[2]. Here, we discuss novel nitrides with rare-earth metals which have been realized experimentally using high pressures and high temperatures.

The second project illustrates an example of a structural phase transition at high pressure in the $CrSb_2$ compound, which has been extensively studied for its thermoelectric properties[3]. The crystal structure at ambient pressure is the orthorhombic marcasite structure (space group *Pnnm*). However, during compression we observe a structural phase transition around 12 GPa to a yet unknown structure. In addition, we present the results from laser-heating experiments of $CrSb_2$ starting from either the marcasite phase or from the unknown high-pressure phase.

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