Phase boundary mapping and phase discovery in a quaternary system: carrier density control in Cu2HgGeTe4

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Abstract

Carrier density control is a persistent challenge in the optimization of thermoelectric materials. Phase boundary mapping is a promising new technique which has enabled dramatic increases in the zT of n-type Mg3Sb2 in the last few years. The application and extension of phase boundary mapping into higher dimensional spaces (e.g. ternaries, quaternaries) is no trivial task. Our prior work has shown that the quaternary diamond-like semiconductors (e.g. Cu2HgGeTe4) are expected to have zT > 1.5 at 300°C under optimized carrier densities. However, the materials present as near degenerately doped p-type materials which are notoriously difficult to dope. Furthermore, computational defect calculations are often unreliable due to the small band gaps and spin-orbit coupling present in these materials. In this work, I demonstrate a rational method to apply phase boundary mapping to complex quaternary systems. I reveal the challenges in applying phase boundary mapping in systems where the phase diagram is unknown or sufficiently complex such that computational methods are hindered. I show how the presence of previously unknown ternary phases can dramatically influence the properties and (in some cases) provide a new means to optimize thermoelectric materials through selective scattering of phonons from antisite defects. Our work culminates in the discovery of several new phases, the optimization of an existing phase, and the realization of high p-type zT at 250-300°C in a new material class.

Keywords: Phase boundary mapping, diamond like semiconductor, p, type, carrier density control, doping, material discovery, thermodynamics

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