Negative-Pressure Polymorphs via Heterostructural Alloying

Scientific Achievement
For some heterostructural alloys, a metastable low-density polymorph can be the preferred structure for non-phase-separated intermediate compositions (top panel). For the MnSe-MnTe system, where this effect was predicted theoretically (middle panel), the low-density metastable wurtzite (WZ) phase was successfully synthesized using non-equilibrium thin-film growth (bottom panel).

Significance and Impact
Although high pressure can often be used to synthesize high-density phases not stable at ambient pressure, the analogous low-density (negative-pressure) phases are not readily synthesizable. Here, we have shown that heterostructural alloying is a viable synthesis method to realize such materials. This opens a new design space for targeting functional materials such as piezoelectric WZ-Mn(Se,Te).

Research Details
Theory: For MnSe$_{1-x}$Te$_x$ alloys, the mixing enthalpy for the low-density WZ structure has smaller bowing than for high-density nickeline (NC) and rock salt (RS) structure (middle panel).

Experiment: Mixing high-density forms of RS-MnSe and NC-MnTe at low temperature using non-equilibrium synthesis stabilizes a MnSe$_{1-x}$Te$_x$ alloy with a low-density WZ structure (bottom panel).