The pressure variable opens the door towards the synthesis of materials with unique properties, i.e. superconductivity, hydrogen storage media, high-energy density and superhard materials, to name a few. Under pressure elements that would not normally combine may form stable compounds or may mix in novel proportions. As a result, we cannot use our chemical intuition developed at 1 atm to predict phases that become stable when they are compressed. To enable our search for novel BCS-type superconductors that can be synthesized under pressure we have developed XtalOpt, an open-source evolutionary algorithm for crystal structure prediction. XtalOpt has been employed to find the most stable structures of polar intermetallics and hydrides with unique stoichiometries under pressure. Herein, we describe our predictions of novel superconducting systems: BaGe3 phases that can be quenched to 1 atm, as well as iodine polyhydrides (IHn, n > 1) and phosphorous hydrides (PHn, n=1-5). The electronic structure and bonding of the predicted phases is analyzed by detailed first-principles calculations. The results of our computational experiments are helping to build chemical and physical intuition for compressed solids.