In article number 2104645, Le Duc Anh, Masaaki Tanaka, and co-workers report the epitaxial growth of elemental topological Dirac semimetal \( \alpha \)-Sn with the highest quality thus far. The very high quantum mobilities in these samples allow a quantitative characterization of the nontrivial interfacial and bulk band structure of \( \alpha \)-Sn via quantum transport in combination with first-principles calculations. The results establish \( \alpha \)-Sn as an excellent model system to study novel topological phases and device applications.