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Emergent nanoscale fluctuations in high rock-salt PbTe[1]

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Lead Telluride is one of the most promising thermoelectric materials in the temperature range just above room temperature. It is a narrow band gap semiconductor with a high Seebeck coefficient and a low thermal conductivity. It is structurally much simpler than many other leading candidates for high performance thermoelectrics being a binary rock-salt, isostructural to NaCl. The thermoelectric figure of merit, ZT, can be markedly improved by alloying with various other elements by forming quenched nanostructures. The undoped endmember, PbTe, does not have any such quenched nanostructure, yet has a rather low intrinsic thermal conductivity. There are also a number of interesting and non-canonical behaviors that it exhibits, such as an increasing measured band-gap with increasing temperature, exactly opposite to what is normally seen due to Fermi smearing of the band edge, and an unexpected non-monotonicity of the band gap in the series PbTe - PbSe - PbS. The material is on the surface simple, but hides some interesting complexity. We have investigated in detail the PbTe endmember using x-ray and neutron diffraction and neutron inelastic scattering [1]. To our surprise, using the atomic pair distribution function (PDF) analysis of neutron powder diffraction data we found that an interesting and non-trivial local structure that appears on warming. with the Pb atoms moving off the high-symmetry rock-salt positions towards neighboring Te ions. No evidence for the off-centering of the Pb atoms is seen at low temperature. The crossover from the locally undistorted to the locally distorted state occurs on warming between 100 K and 250 K. This unexpected emergence of local symmetry broken distortions from an undistorted ground-state we have called emphanisis, from the Greek for appearing from nothing. We have also investigated the lattice dynamics of the system to search for a dynamical signature of this behavior and extended the studies to doped systems and I will also describe the results of these experiments. This work gives key insights into PbTe, the possible origin of its anomalous electronic structure properties, and why it is such an attractive parent compound for nanostructured high performance thermoelectric materials. I would like to acknowledge the excellent collaborations that occurred during this work, including Emil Bozin at Brookhaven National Laboratory, Mercouri Kanatzidis and Christos Malliakas at Northwestern University and Argonne National Laboratory, Kirsten Jensen from U. Aarhus, Steve Shapiro at Brookhaven National Laboratory, Matt Stone and Mark Lumsden at Oak Ridge National Laboratory, Nicola Spalding at ETH Zurich and Petros Souvatzis at Los Alamos National Laboratory. I would also like to acknowledge the support of the national user facilities and their staff where the work was done. [1] E.S. Bozin et al., Science v330, pp1660 (2010).

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