Redox-Coupled Structural Distortions in Quasi-1-Dimensional Au_2MP_2 (M=Tl, Pb, and Bi)

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Recent research in topological materials has predicted and confirmed new quasi-particles, categorized based on electronically distinct states in crystals. At the foundation of these states lies two tuning parameters, namely crystalline symmetry and Fermi-level filling. For example, in the $GdSb_xTe_{2-x-\delta}$ (x=0-1, δ=concentration of defects) system, the Sb:Te ratio governs the electron-filling of the band structure, producing a tunable system of structural distortions in its square-net layer. At specific ratios, these distortions retain certain symmetry-protected bands, such as a Dirac node on the Fermi surface, and gap out topologically trivial bands at the Fermi surface.

One interest now is to investigate tunable topological structural motifs beyond a square net of atoms. A one-dimensional (1D) chain of atoms realizes similar symmetry protected Dirac nodes as the square net. The first part of my presentation expands on previously reported Au_2MP_2 (M=Hg, Tl, Pb, and now Bi), which contains a 1D chain of M atoms. Surprisingly, the series remains isotypic when substituting M across a change of four valence electrons per chain atom. Additionally, the Bi analogue also contains a monoclinic polymorph that retains the linear chain motif despite a distortion of the atoms surrounding the chain.

The second part of my presentation demonstrates the importance of chemical workup in solid state compounds. X-ray and electron diffraction characterizations indicate soaking crystals of the Au2MP2 system in HNO_3 insights a structural distortion whose resulting symmetry and size of supercell depends on the identity of M. For example, the Pb analogue undergoes a 13-fold supercell enlargement along the chain axis. The efficiency of this distortion seems to be enhanced when triggered electrochemically. Further characterization and computations suggest the averaging of oxidation states between Au-Pb contacts as a possible driving force for the distortions. Finally, electronic transport between the modulated and unmodulated compounds is compared. As a result of these insights, metastability in various structural types may be further understood with respect to their chemical environment.

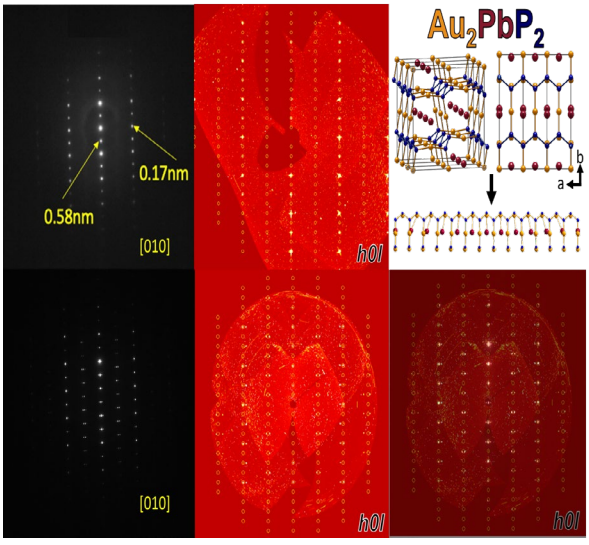

Figure 1