Crystal-Chemical Origins of the Ultrahigh Conductivity of Metallic Delafossites

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The delafossites are a class of complex oxides with general formula ABO_2 that have been available synthetically since 1971 [1]. Some of these delafossites are metallic, where conductive triangular sheets of A^{1+} ions are interspersed with insulating $B^{3+}O_6$ edge-sharing octahedral layers, generating a remarkably simple electronic structure at the Fermi level [1,2]. Only in the last 10-20 years, however, was it understood that despite their highly anisotropic complex-oxidic nature, metallic delafossites (particularly $PdCoO_2$ and $PtCoO_2$) are the *most conductive oxides known*, for reasons that remain poorly understood [1]. In particular, their room-temperature resistivity is lower than Au and their low-temperature resistivity falls as low as 8 nΩ cm, implying mean-free-paths of ~20 μm [1]. These extraordinary values have led to a slew of recent advances [1]. To reach such low-temperature values, it is widely accepted that these materials must be ultrapure and ultraperfect, although the methods for their bulk growth (which produce only small crystals) are not typically capable of such [1].

In this presentation, we first report a new approach to $PdCoO_2$ crystal growth, using a novel chemical vapor transport method to achieve order-of-magnitude gains in size and mass, the highest structural qualities yet reported, and record residual resistivity ratios ($440 < RRR < 760$) [3]. Nevertheless, the first detailed mass spectrometry measurements on these materials reveals that they are *not* ultrapure, typically harboring 100s-of-part-per-million impurity levels [3]. Through detailed crystal-chemical analyses, we resolve this apparent dichotomy, showing that the vast majority of impurities are forced to reside in the insulating Co-O octahedral layers, leaving the conductive Pd sheets highly pure (~1 ppm impurity concentrations). These purities are shown to be in quantitative agreement with measured residual resistivites [3]. We thus conclude that a previously unconsidered "sublattice purification" mechanism is essential to the ultrahigh low-temperature conductivity and mean-free-path of metallic delafossites [3], opening up many exciting device possibilities.

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