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Assembling Quantum Materials from Molecular Building Blocks: Metallic Transport and Bloch–Grüneisen Behavior in a SURMOF Thin Film

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The deliberate assembly of "designer solids" from molecular building blocks is a central objective of reticular chemistry and has stimulated enormous interest in metal–organic frameworks (MOFs). Beyond their structural diversity and porosity, conductive MOFs have recently emerged as promising candidates for low-dimensional quantum materials. However, compelling experimental evidence for metallic transport in these systems has remained elusive.

This lecture will describe recent progress achieved using highly ordered MOF thin films prepared by a layer-by-layer (lbl) deposition method. These films, termed SURMOFs,[1,2] are grown on functionalized conducting substrates and combine exceptional structural quality [3] with straightforward electrical contacting [4] and access to band-structure effects.[5] Previous measurements on Cu_3HHTP_2 SURMOF thin films using lithographically defined electrodes with dimensions down to 100 nm revealed clear non-activated, metal-like transport down to 100 K.[6]

In conventional metals, the low-temperature conductivity follows Bloch–Grüneisen behavior characteristic of electron–acoustic-phonon scattering. Extending transport measurements on Cu_3HHTP_2 thin films down to 7 K, we observe a pronounced increase in conductivity and signatures of Bloch–Grüneisen behavior, providing strong evidence for transport via delocalized electronic states rather than thermally activated hopping. Together with theoretical predictions of Dirac-cone band structures in Kagome lattices, these findings establish reticular materials as a new platform for quantum materials assembled from molecular building blocks.

The progress achieved was greatly facilitated by autonomous experimentation. Owing to their modular layer-by-layer synthesis, SURMOFs are ideally suited for robotic preparation and machine-learning-guided optimization, enabling efficient optimization of film orientation, crystallinity, and electrical conductivity.

References:

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