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AUGUST 31 - SEPTEMBER 03, 2021 • BOLOGNA, ITALY
EUROPEAN CONFERENCE ON CHEMISTRY OF TWO-DIMENSIONAL MATERIALS

Layer-Oriented 2D Conjugated Metal-Organic Framework Films Enabling Directional Charge Transport

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Layered two-dimensional conjugated metal-organic frameworks (2D *c*-MOFs), with strong in-plane conjugation and weak out-plane van der Waals force, have emerged as promising electrically conductive materials for organic electronics such as field-effect transistors (FETs) and spintronic devices. One of the key challenges faced by the scientific community is to synthesize 2D *c*-MOFs with control of layer orientation to dial-in desired electronic properties/devices. Here, we report the novel synthesis of unprecedented edge-on layer-orientated *p*-type semiconducting 2D *c*-MOF films by combining supramolecular chemistry and interface-assisted polymerization. We figure out the crystal structure and orientations of the 2D *c*-MOFs films ($Cu_2[PcM\text{-}O_8]$, M=Cu, Fe) with molecular precision, and demonstrate that the edge-on structure formation is guided by the pre-organization of metal-phthalocyanine ligands, whose basal plane is perpendicular to the water surface due to their π-π interaction and hydrophobicity. The synthetic $Cu_2[PcCu\text{-}O_8]$ film features with a thickness of ~20 nm and ~600 nm-sized crystal domains, and exhibits a room temperature conductivity of ~5.6×10^{-4} S cm^{-1} and a Hall mobility of ~4.4 cm^2 V^{-1} s^{-1} based on macroscopic van der Pauw pattern. Lateral and vertical measurements further reveal the directional charge transport feature in this edge-on 2D *c*-MOF film, i.e., the lateral conductivity is 2~3 orders of magnitude higher than the vertical one. The directional conductivity studies combined with theoretical calculation identify that the intrinsic conductivity of $Cu_2[PcCu\text{-}O_8]$ is dominated by charge transfer along the interlayer pathway. This work provides a state-of-art insight into the controlled synthesis of layer-orientated semiconductive 2D *c*-MOF films, allowing to dial-in lateral/vertical electronic devices and determine the intrinsic transport mechanism.

References

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Figures