Organic 2D Crystals and van der Waals Heterostructures

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Organic two-dimensional (O2DCs) crystals have emerged as a unique class of synthetic van der Waals layered materials built from π-conjugated molecular units. They exhibit extended in-plane π-conjugation and/or interlayer electronic couplings, and comprise a broad class of 2D polymers/supramolecular polymers, 2D conjugated polymers or covalent-organic frameworks, and 2D conjugated metal-organic frameworks. Their structural programmability and chemical diversity provide opportunities to engineer electronic and transport properties at the molecular level, offering design freedoms beyond those of inorganic 2D crystal materials.

In the first part of this talk, I will present our recent advances in the bottom-up synthesis of highly crystalline 2D polymer crystals, particularly through on-water/on-liquid surface synthesis strategies. The confined water surface enables controlled 2D polymerization, leading to single- and few-layer 2D polymer crystals with long-range in-plane order and well-defined lattice structures. These materials exhibit extended 2D π-conjugation, tunable band structures, and high intrinsic charge-carrier mobilities. Beyond charge transport, their ordered porous architectures also enable ion transport and interfacial electrochemical functionality. In the second part, I will focus on our recent realization of organic 2D van der Waals heterostructures. Using sequential on-water surface assembly, we achieve programmable layer-by-layer stacking of chemically distinct 2D polymers with defined lattice registry, stacking sequence, and thickness. This strategy enables both lattice-matched and controlled lattice-mismatched heterostructures, allowing systematic tuning of interlayer coupling. We will reveal emergent interfacial phenomena—including built-in electric fields, efficient charge separation, and diode-like rectification—that are governed by lattice matching and interfacial dipole alignment. These results establish O2DCs and their van der Waals heterostructures as a chemically programmable platform for exploring interlayer-coupled electronic, ionic, and quantum phenomena, opening new directions for functional electronics and beyond.