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## Crystal Engineering of Low-Dimensional Metal Halide Hybrids for In-Memory Computing Technologies

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Low-dimensional metal halide hybrids have recently emerged as promising materials for adaptive electronic and neuromorphic devices due to their structural versatility and defect-mediated transport properties. In this research journey, a crystal engineering approach was employed to systematically control the dimensionality and electronic structure of halide complexes through modification of the organic component. Pyridinium-based cations were shown to direct the assembly of metal–halide units from isolated 0D clusters to extended 1D chains through hydrogen bonding and electrostatic interactions, enabling cation-dependent band-gap modulation and influencing resistive switching behaviour in metal–insulator–metal devices [1-4]. Low-dimensional systems exhibited enhanced resistive switching characteristics associated with defect-assisted charge transport. The design strategy was further extended to aliphatic cations and organic dications, introducing additional structural stabilization and tunable connectivity within the inorganic framework. These results highlight the potential of structurally engineered metal halide hybrids as a versatile platform for low-power neuromorphic and in-memory computing technologies, encompassing both lead-free and transition-metal-based systems [1-4].

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