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Polymorphism in Low-dimensional Molecule-based Synthetic Control of Magnetic and Electronic Materials: Structure¹ ADORA BALDWIN, JAMIE MANSON, Eastern Washington University, JOHN SCHLUETER, Argonne National Laboratory — Competition for hydrogen bonds in molecule-based materials frequently leads to the stabilization of polymorphic structures with similar energies. Herein, we report that among magnetic coordination polymers selective crystallization of the $[Cu(HF_2)]$ $(\text{pyrazine})_2$ SbF₆ and $[Cu_2F(\text{HF})(\text{HF}_2) (\text{pyrazine})_4](\text{SbF}_6)_2$ polymorphs can be achieved through control of pressure and temperature during the crystallization process. Through this manner, orbital order and magnetic dimensionality (2D vs. 1D) can be controlled in this system. Similarly, in the analogous $[Co(HF_2) (pyrazine)_2]SbF_6$ system, selective crystallization of the $[CoF_2 (pyrazine)_2]SbF_6(H_2O)_3(H_3O)$ polymorph has been achieved. While these cobalt materials exhibit a 2D magnetic structure, the interlayer coupling is diminished in the hydrated material. Among cation radical salts, five polymorphic phases in the [bis(ethylenedithio)tetrathiafulvalene] $_2$ Ag(CF₃)₄(1,1,2trichloroethane) system have been crystallized, four of which have superconducting ground states. Among the superconducting salts, the ones with intercalated charge ordered layers exhibit a five-fold enhancement of T_c .

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