

Engineering Artificial Spin Networks by On-Surface Metal–Organic Coordination

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Artificial spin networks on metallic substrates have emerged as a powerful platform for exploring low-dimensional magnetism. The controlled assembly of magnetic atoms or molecules on surfaces offer a versatile platform for investigating collective magnetic order, such as ferromagnetism, as well as many-body phenomena including the Kondo effect [1-3]. Metal–organic coordination nanostructures provide an effective strategy for fabricating such artificial spin networks with atomic precision. In this work, we investigate the formation of Fe-coordinated networks on the Au(111) surface with tunable pore sizes defined by the molecular precursors. Two tritopic molecules were employed, both featuring three pyridyl terminal groups connected to a central benzene core: 1,3,5-tris[4-(pyridinyl)phenyl]benzene (TPyPB) and 1,3,5-tris[4-(pyridinyl)-[1,1'-biphenyl]]benzene (TPyPPB), which differ in molecular length due to the presence of one or two phenyl spacers, respectively. Following room-temperature (RT) deposition on Au(111), scanning tunneling microscopy (STM) revealed that both molecules self-assemble into porous honeycomb networks stabilized by two-fold coordination with surface gold atoms. Subsequent Fe deposition directly induces the transformation from Au-coordinated to Fe-coordinated honeycomb lattices with threefold metal coordination at RT, a mechanism further supported by density functional theory (DFT) calculations. Thermal annealing promotes the formation of long-range ordered periodic structures with a low density of defects, with structural stability preserved up to approximately 600 K. Overall, these results demonstrate that the pore size and lattice geometry of surface-supported metal–organic networks can be systematically tuned by the choice of molecular precursor and by switching the coordination center between Au and Fe, providing a controlled approach to the on-surface engineering of artificial spin lattices.

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