Abstract: The carboxylic acid (COOH) functional group contains both an OH that acts as a strong hydrogen-bond donor and a C=O that is a strong hydrogen-bond acceptor. Carboxylic acid crystals are typically built by packing dimers or linear chains, and similar bonding results in two-dimensional networks when molecules are adsorbed in a single layer on a surface. Additional groups that participate in hydrogen bonding, even if only very weakly, can modulate the interaction between carboxylic acids and create significantly more complex structure. In particular, ferrocenecarboxylic acid (FcCOOH) assembles into cyclic five-membered rings stabilized by C-H…O hydrogen bonds, and these rings precipitate out from solution alongside FcCOOH dimers. The resulting co-crystallized structure can be examined at the molecular scale with scanning tunneling microscopy, which shows that the surface has long-range orientational order. Translational order is absent: even though there are repeating patterns, repetition does not occur on fixed intervals. This is the only known case where aperiodic structure results from the non-covalent assembly of small molecules. The kinetic and thermodynamic factors responsible for this assembly will be discussed, as will extensibility to related systems.