Supporting Information

Coverage and Enantiomeric Excess Dependent Enantiomorphism in Two-Dimensional Molecular Crystals

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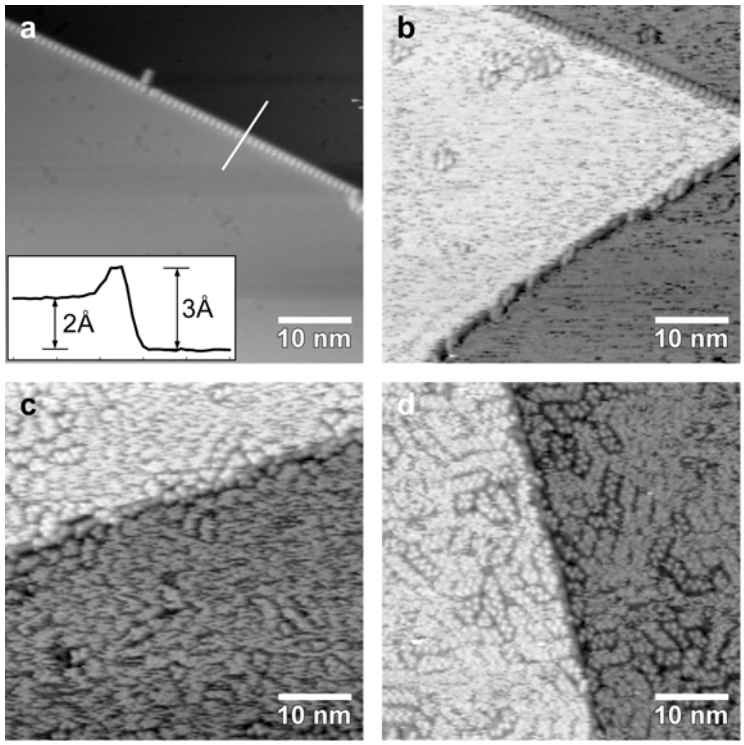

Figure S1. Series of STM images with increasing global coverage up to half of a monolayer. a) At low coverage the [7]H molecules decorate the lower part of step edges and the apparent height of a molecule (~ 3 Å) is larger than a Cu(111) single step with a height of 2.08 Å. b), c) With increasing coverage noisy patterns are formed on flat terraces due to a high mobility of molecules on the close-packed surface and a weak attractive intermolecular interaction. d) At coverages close to half of a saturated monolayer short double rows with a zigzag structure appear to condensate randomly in the mobile phase. (a: θ_{rel}^{global} = 0.05, U = +2.61 V, I = 80 pA; b: θ_{rel}^{global} = 0.17, U = +2.81 V, I = 16 pA; c: θ_{rel}^{global} = 0.44, U = +2.84 V, I = 17 pA; d: θ_{rel}^{global} = 0.51, U = +2.83 V, I = 20 pA;)

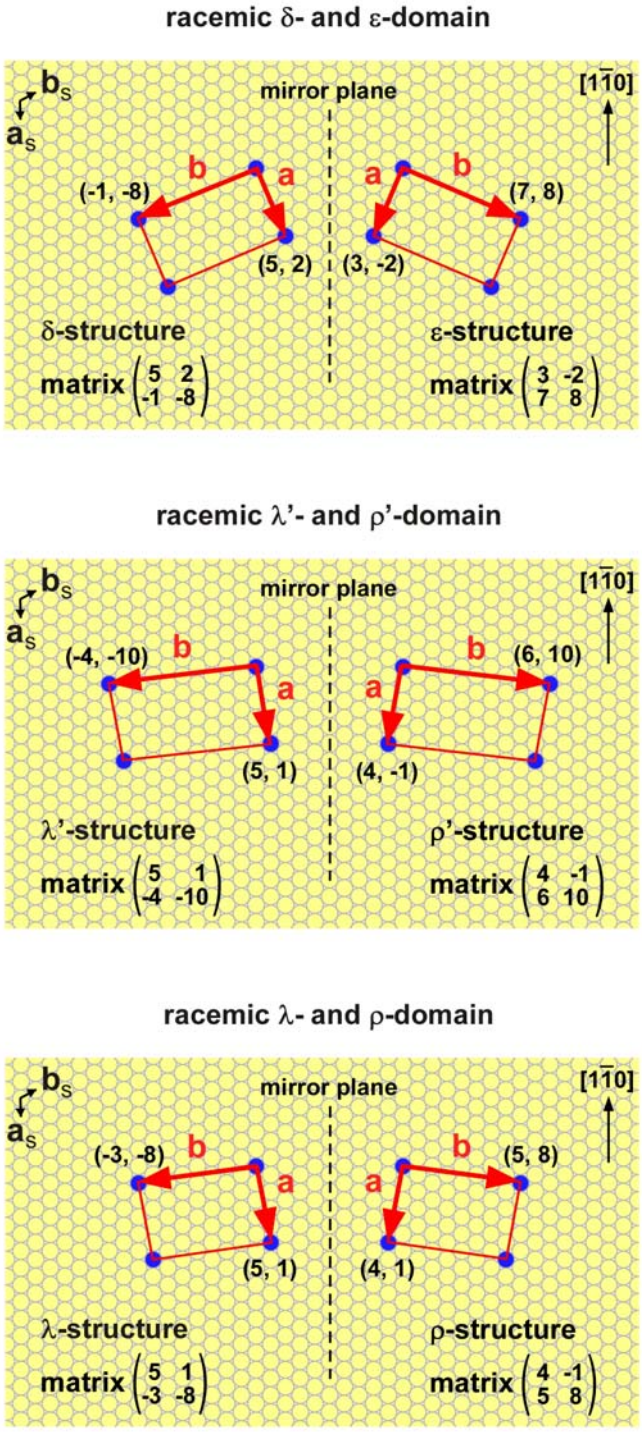

Figure S2. Unit cells for the six racemic heptahelicene lattices on Cu(111). The unit vectors have been chosen for the enantiomorphous cells on the right, respectively, according to the rules given in Ref. S[1].

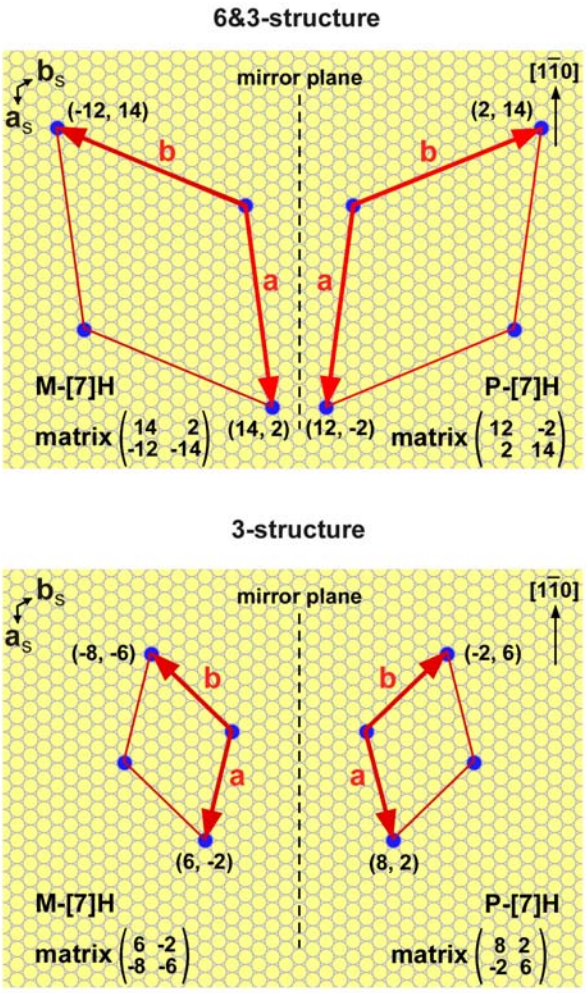

Figure S3. Unit cells for four enantiopure heptahelicene lattices on Cu(111). Top: β (6&3) structure, bottom: γ (3-) structure.

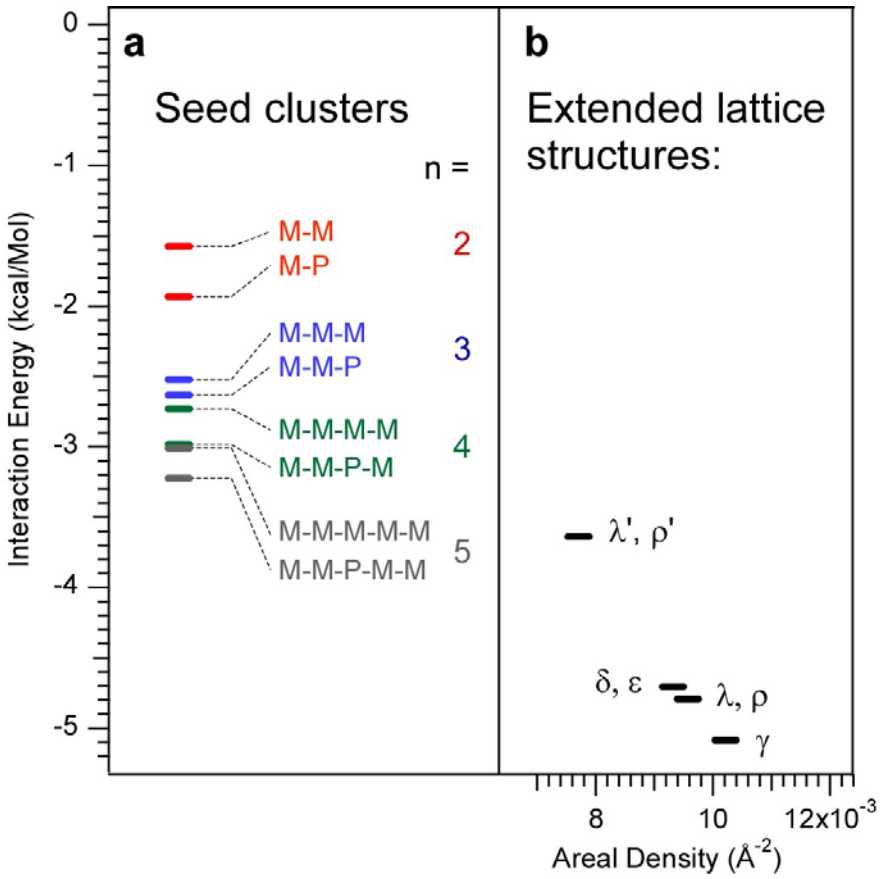

Figure S4. Interaction energies per molecule for different minimum total energy configurations as determined from molecular mechanics calculations using the AMBER force field. a) Energies of small clusters consisting of n = 2, …, 5 molecules. For each cluster size, the most favorable enantiopure and the most favorable mixed configurations are given. Mixed seed clusters are energetically favored for all sizes. b) Energies per molecule for the optimum configurations of the extended lattice structures as indicated, plotted versus the corresponding packing densities. The enantiopure γ phase is favored by 0.25 kcal/mol over the λ/ρ phase.

(S1) Unertl, W. N. Physical structure. In: Holloway S, Richardson NV (eds.) Surface crystallography, handbook of surface science 1, Elsevier, Amsterdam, chap 1, (1996).