

## ***Electronic Supplementary Information***

# **Influence of Step Faceting on the Enantiospecific Decomposition of Aspartic Acid on Naturally Chiral Surfaces Vicinal to Cu{111}**

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### **Experimental Methods**

The TPRS and XPS experiments using the Cu{111}-S<sup>4</sup>C were conducted at Carnegie Mellon University were conducted in a ThetaProbe system equipped with a preparation chamber and an analysis chamber, both evacuated to UHV. Cu{111}-S<sup>4</sup>C cleaning, annealing and the adsorption of Asp were conducted in the preparation chamber. The Cu{111}-S<sup>4</sup>C was cleaned by cycles of heating to 650 K and sputtered with 2 keV Ar<sup>+</sup> ions with a sputtering current of 10 μA for 40 minutes followed by annealing in UHV for 10 minutes at 800 K. Deposition of Asp was performed using an evaporation source consisting of a resistively heated glass vial containing the solid Asp powder. The temperature of the source was measured with a thermocouple bonded to the glass vial and controlled using a computer.

The quench TPRS experimental procedure for *L*-Asp on the Cu{111}-S<sup>4</sup>C consisted of seven steps:

1. Sputter and anneal Cu Cu{111}-S<sup>4</sup>C for two cycles.
2. Perform XPS at 5 points on the Cu{111}-S<sup>4</sup>C surface to determine whether it is clean.
3. Adsorb a multilayer of *L*-Asp on the Cu{111}-S<sup>4</sup>C with the surface at < 330 K and a doser temperature of ~440 K.
4. Perform XPS of multilayer of *L*-Asp at 5 points on the Cu{111}-S<sup>4</sup>C surface
5. Heat *L*-Asp/ Cu{111}-S<sup>4</sup>C at 405 K for > 10 minutes to desorb multilayer and leave the adsorbed monolayer.
6. Anneal *L*-Asp/ Cu{111}-S<sup>4</sup>C at 470 K while monitoring CO<sub>2</sub> desorption with a mass spectrometer and quench the temperature during isothermal CO<sub>2</sub> desorption.
7. Obtained O 1s, C 1s, N 1s, and Cu 2p<sub>3/2</sub> x-ray photoemission spectra using a 400 μm diameter x-ray spot at 225 points within 3.2 mm of the Cu{111}-S<sup>4</sup>C center.

All STM imaging was conducted at Tufts University using an UHV variable temperature scanning tunneling microscope (VT-STM, Omicron Nanotechnology). The Cu{111}-S<sup>4</sup>C surface was prepared by many cycles of Ar<sup>+</sup> sputtering (1.5 keV, 15 μA) and annealing at 750 K. *L*-Asp was deposited onto the Cu{111}-S<sup>4</sup>C surface at room temperature from a homemade sublimation source in a preparation chamber ( $P < 1 \times 10^{-9}$  mbar). The *L*-Asp/ Cu{111}-S<sup>4</sup>C sample was then transferred under UHV to the STM chamber ( $P < 1 \times 10^{-10}$  mbar) and the annealed at 475 K for 1 minute using a pyrolytic boron nitride (PBN) heater embedded behind the sample. Data was recorded with Veeco and Omicron etched W and 85:15 Pt-Ir tips.

## Decomposition of D-Asp on Cu(111)-S<sup>4</sup>C

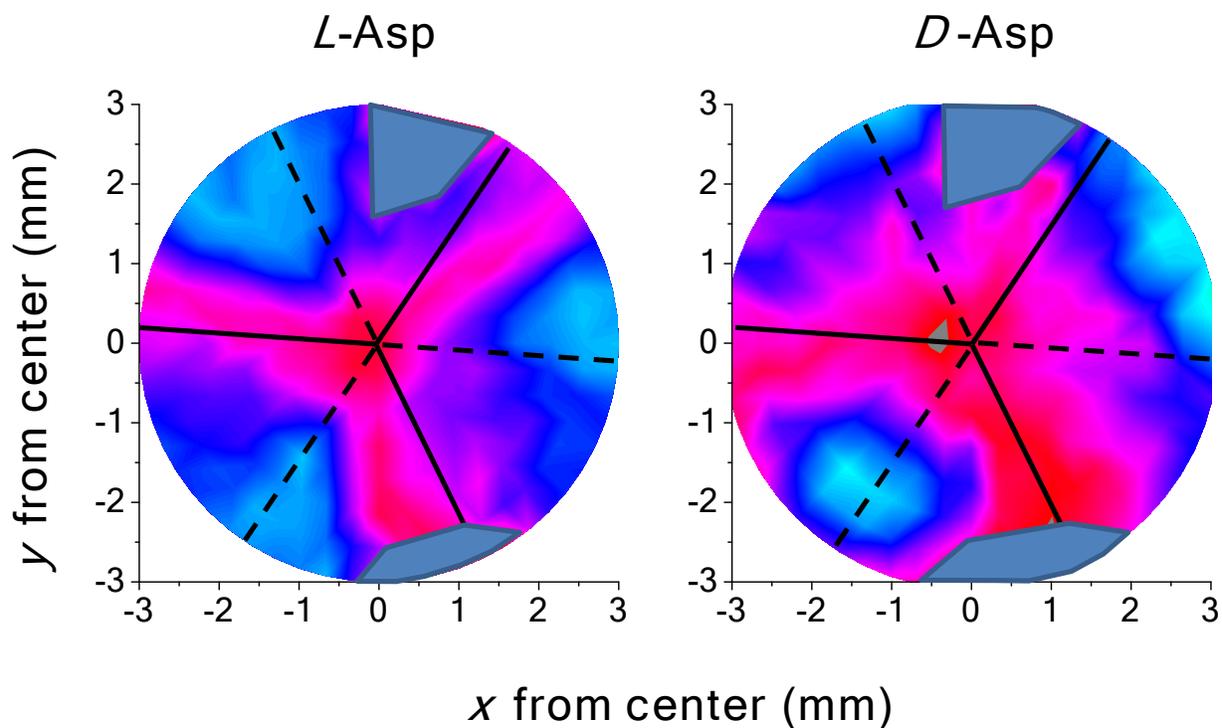


Figure S11. Comparison of the maps of the *L*-Asp (left) and *D*-Asp (right) local coverage across the Cu(111)-S<sup>4</sup>C after annealing and quenching to decompose ~60% of the initially uniform Asp monolayer. The Asp coverage remaining has been quantified by the O 1s XPS signals collected with a 400  $\mu$ m diameter x-ray spot focussed at discrete spots across the surface. Both maps are oriented within the laboratory frame of references for direct comparison with one another. Note that the regions with lowest extent of Asp decomposition (red) are rotated with respect to the high symmetry directions marked with black lines.