Supporting Information

Digitized Charge Transfer Magnitude Determined by Metal-Organic Coordination Number

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STS experiments are performed on various structures with different tips. Thereby the position of features in spectra depends only on the underlying structure (relevant for the main article.) whereas the fine structure depends on the specific tip. This is demonstrated here.

- **Complete curves of molecule A-D with different tips**

  1. **Tip#1**

     Fig. S1 shows STS curves measured by tip#1 on molecules A, B, B', B", D, D', and Au(111) surface. STS on molecule A shows the peak come from Au(111) substrate at around -0.4 V and the frontier orbital at 1.2 V. STS curves show similar results on molecules B, B', and B". The STS curve on molecule D is similar to D'. Clear Au(111) surface state is obtained by tip#1.

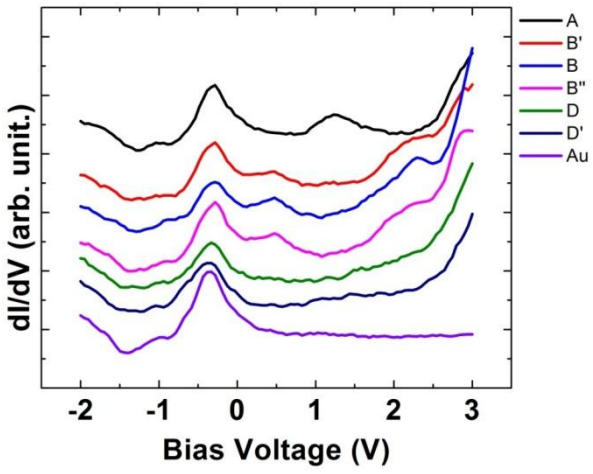

Figure S1

2. **Tip#2**

Fig. 2 shows STS curves measured by tip#2 on molecules C, D', D", and Au(111) surface. STS on molecule C shows the peak come from Au(111) substrate at around -0.4 V and the frontier orbital at 0.2 V. The STS curve on molecule D' is similar to D". Clear Au(111) surface state is obtained by tip#2. The variation of the detail line shapes of Au(111) surface state obtained by two different tips is contributed from the different tip states.

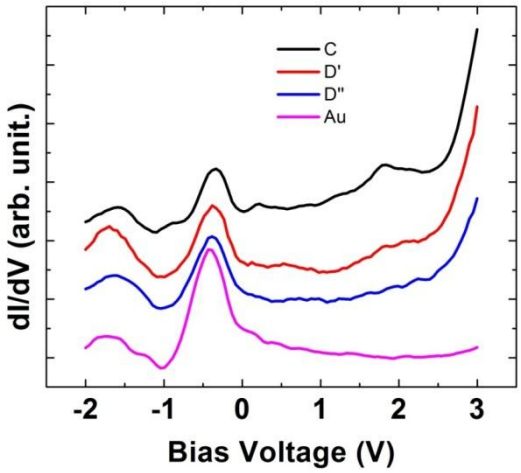

Figure S2

The STS curves presented in the manuscript are extracted from complete two-dimensional data sets with spectra recorded at *every* single image point. For demonstration, Fig. S3 presents a deeper insight into the original data as presented in Fig. 4 of the main article for the two molecules denoted as C and D". Each curve of Fig. 4 represents the average of ~50 individual STS spectra over a surface area of ~3 $Å^2$ at the central position denoted as 5 in Fig. S3. The curves taken within molecule C at different positions (from position 1 to 9) reveal similar features, the peak at -0.4 V represents the Au(111) surface state and at +0.2 V reflects the frontier orbital. As the frontier orbital is localized within specific positions of the molecule, STS curves at different positions vary respectively. The LUMO+1 (+1.8 V) appears in the spectra of molecule C but not in D", it is because of the stronger hybridization that broadening the LUMO+1 in molecule D".

- **STS curves taken at different atomic position within the same molecule.**

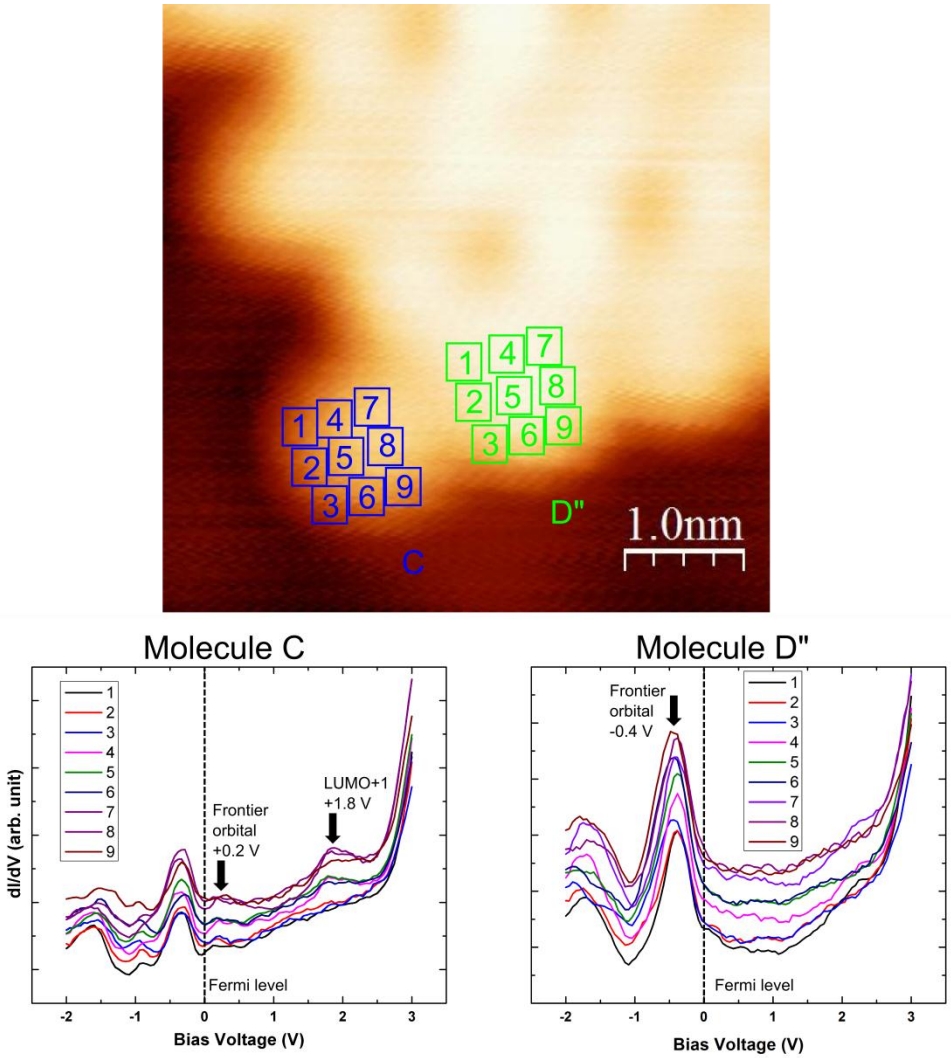

Figure S3