

Supplementary information for

Dynamic intermolecular space for reversible CO₂ capture and release

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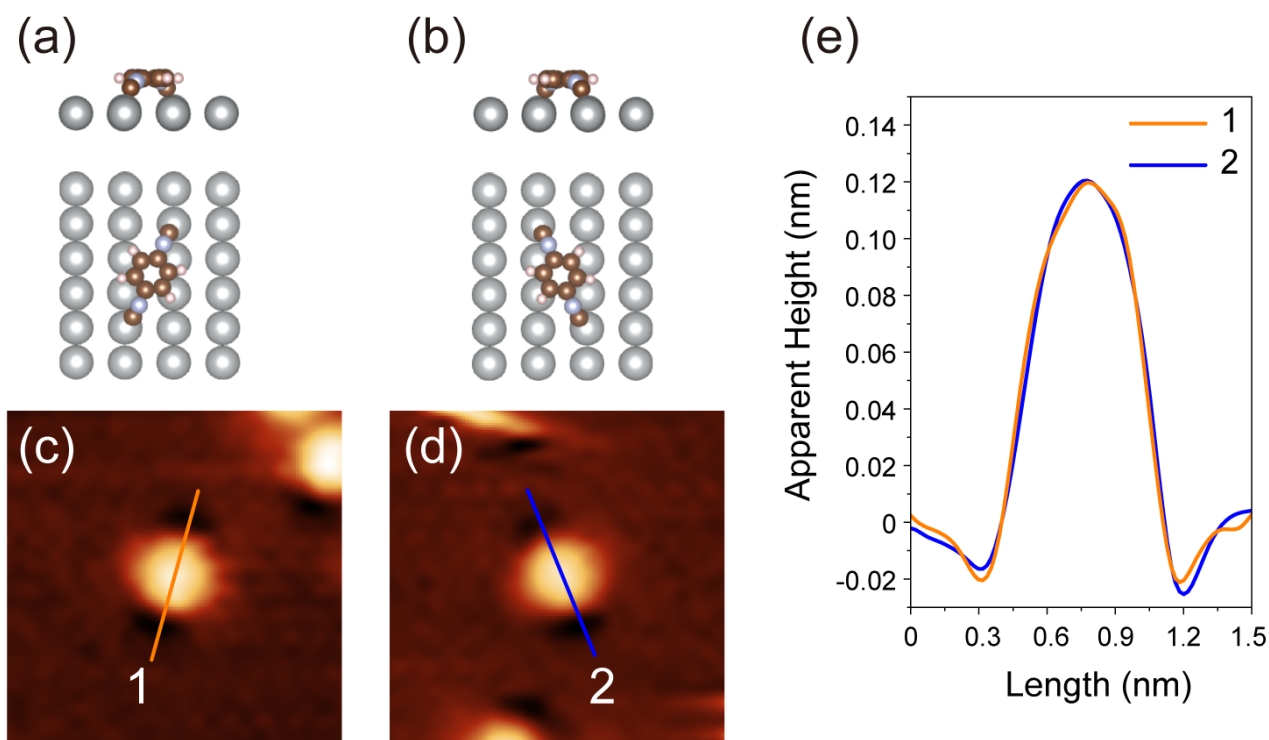


FIG S1. (a, b) The adsorption configuration of flat-lying PDI molecule on Ag(110). (c, d) STM images of individual flat-lying PDI molecules adsorbed at 5 K, with different adsorption orientations with respect to substrate Ag latticed. (e) Line profiles obtained in (c, d), showing the apparent height of ~ 120 pm.

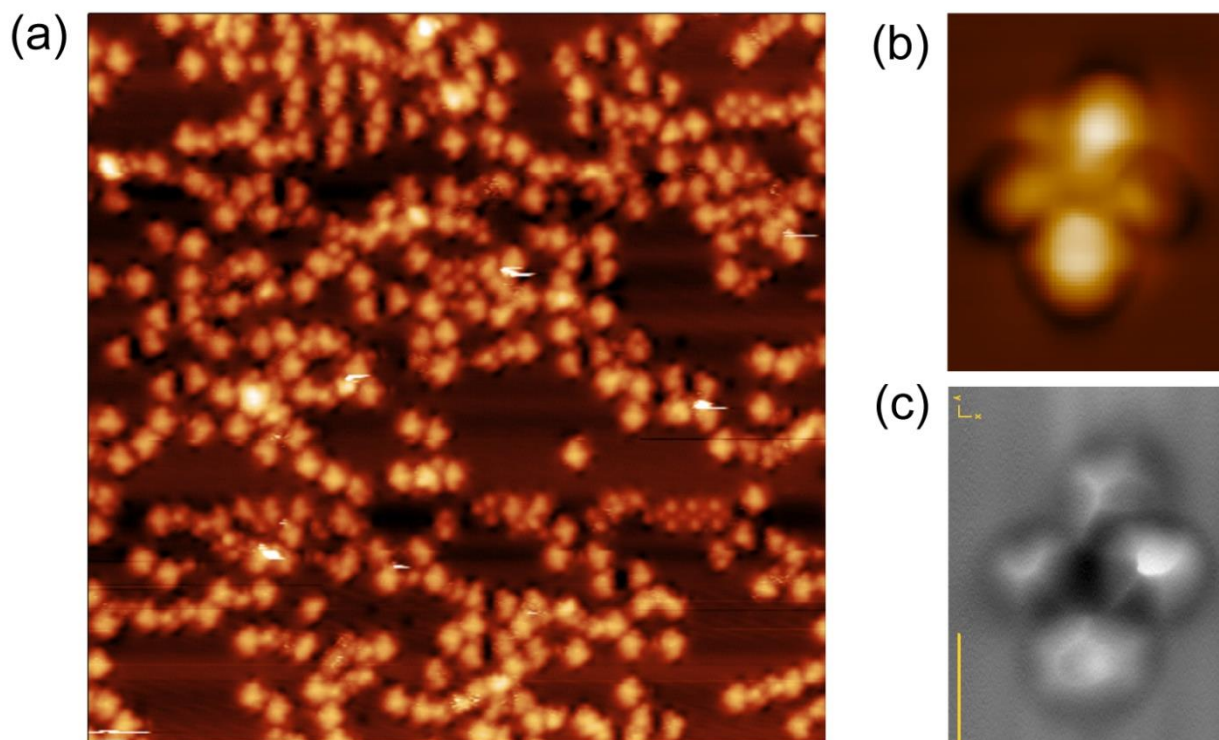


FIG S2. Formation of PDI clusters at a temperature of 250-300 K. (a) A representative STM image of Ag(110) surface after dosing PDI molecules at room temperature. (b) A zoom-in STM image of one PDI cluster, showing complicated structures. (c) The non-contact atomic force microscope (AFM) image of the cluster in (b), showing bond structures. The image in (a) was obtained at 80 K, and the images in (b,c) were obtained at 5 K with a CO-functionalized AFM tip.

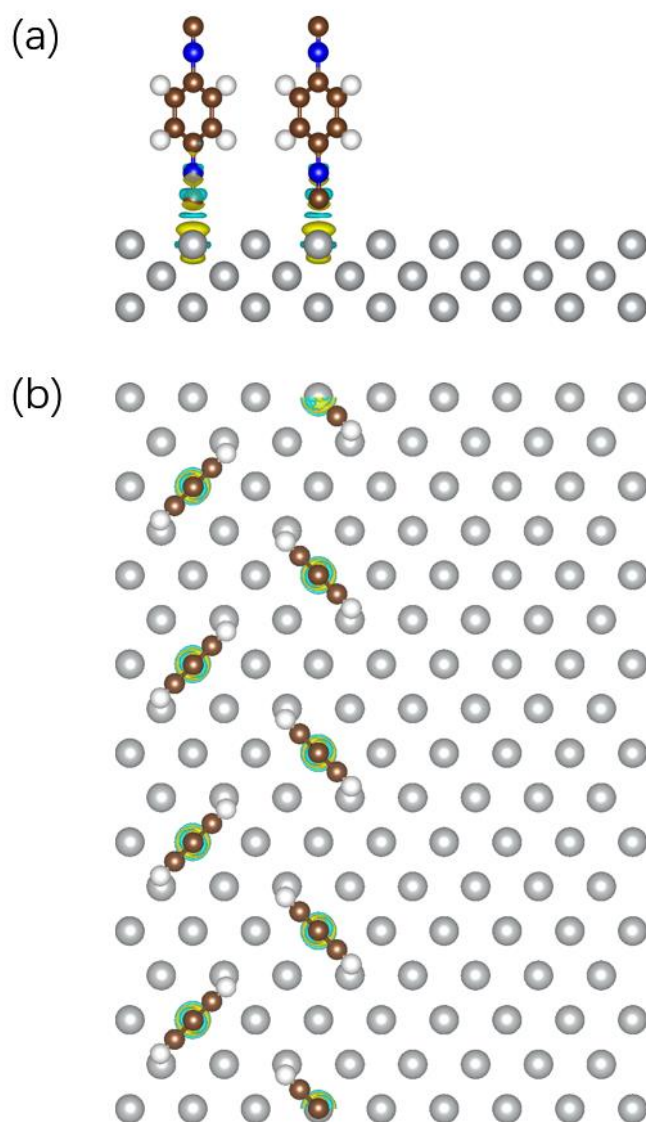


FIG S3. The differential charge density plot of a PDI double chain adsorbed on Ag(110) substrate. (a, b) display the side and top views, respectively. The blue and yellow contours represent regions with more negative charges and less negative charges, respectively. The contour value of charge density is $0.0025 \text{ e}/\text{\AA}^3$.

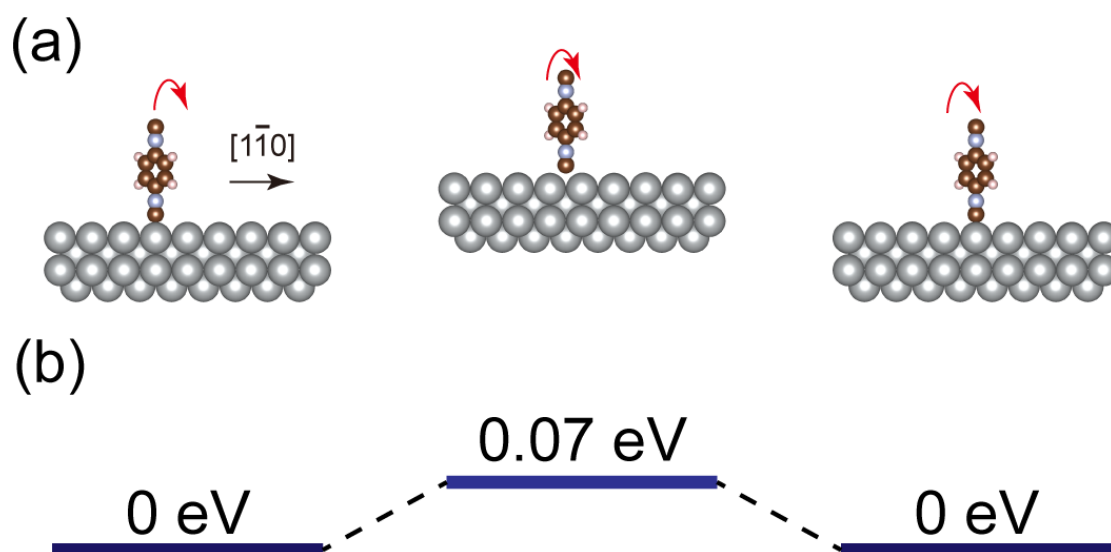


FIG S4. (a) DFT simulation showing a PDI molecule migrating along $[1\bar{1}0]$ orientation on Ag(110) substrate, with an energy barrier of 0.07 eV indicated in (b).