

Supplementary Material for:

Hydroxyl Group as the ‘Bridge’ to Enhance the Single-Molecule Conductance by Hyperconjugation

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1. NMR Spectra

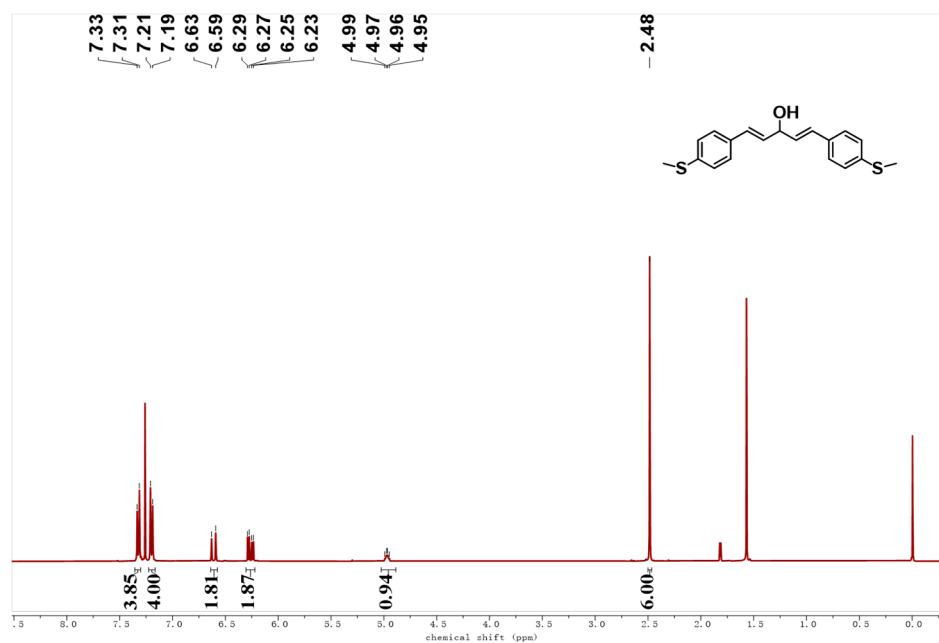


Figure S1 ¹H NMR Spectra of 1.

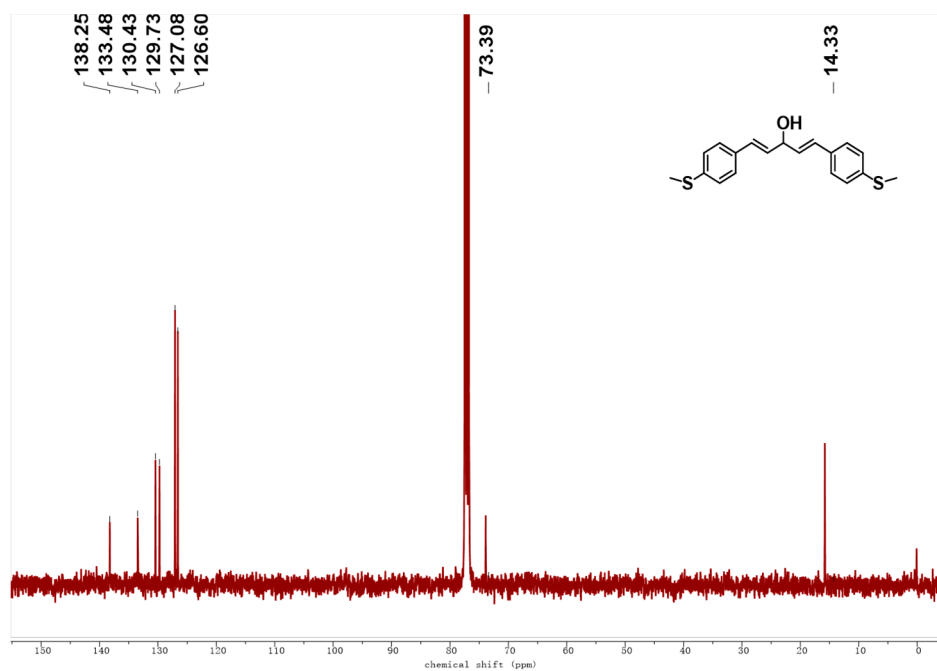


Figure S2 ¹³C NMR Spectra of 1.

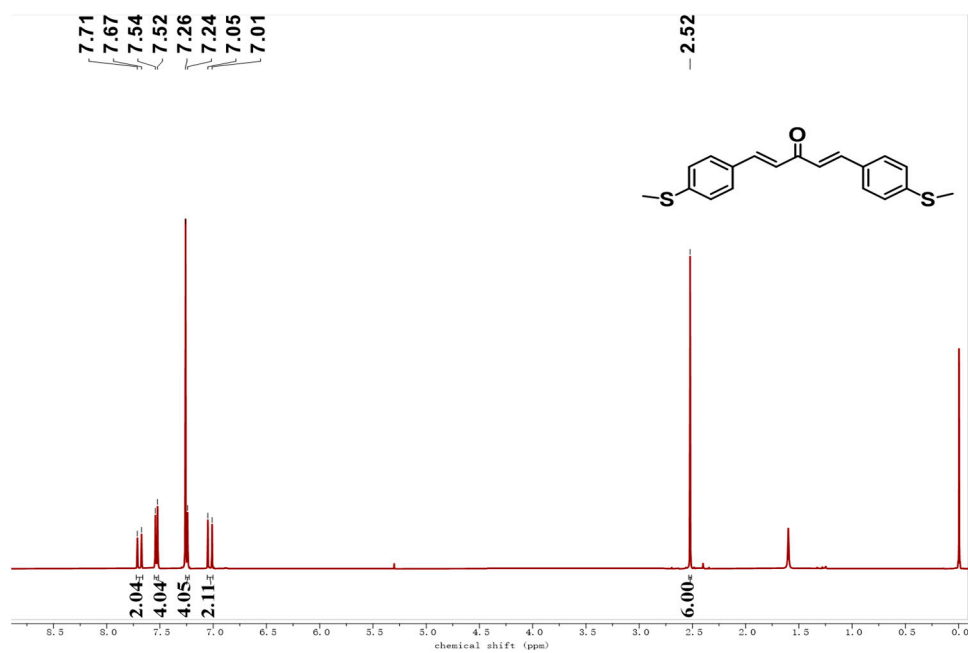


Figure S3 ¹H NMR Spectra of **2**.

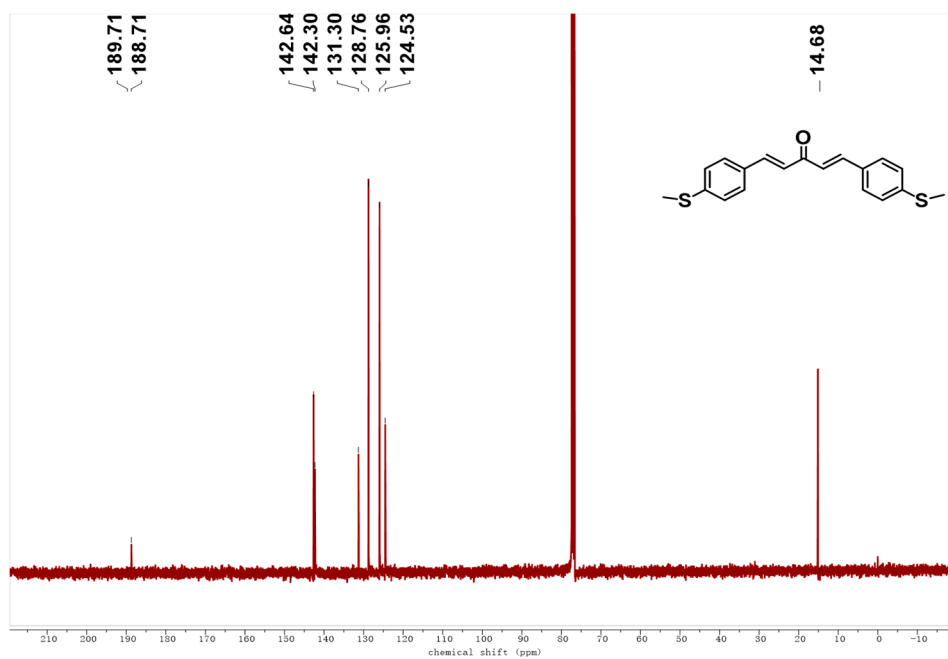


Figure S4 ¹³C NMR Spectra of **2**.

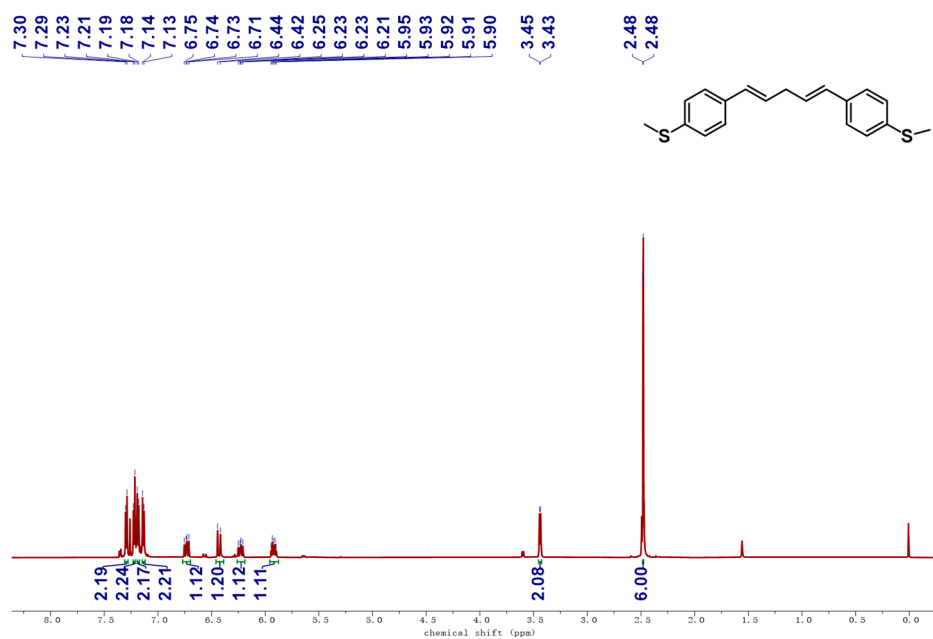


Figure S5 ¹H NMR Spectra of 3.

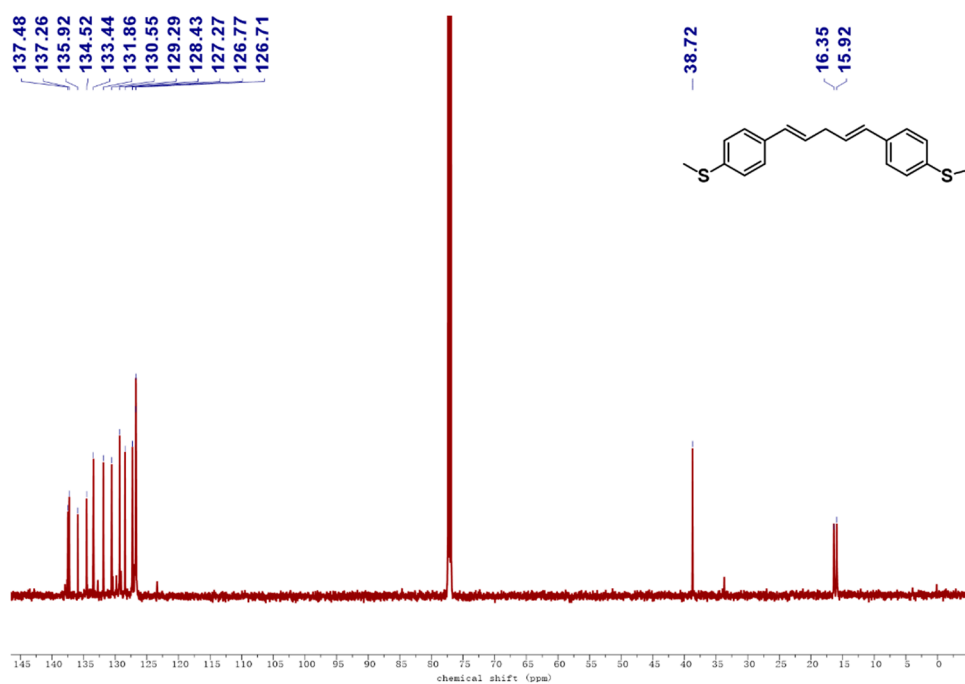


Figure S6 ¹³C NMR Spectra of 3.

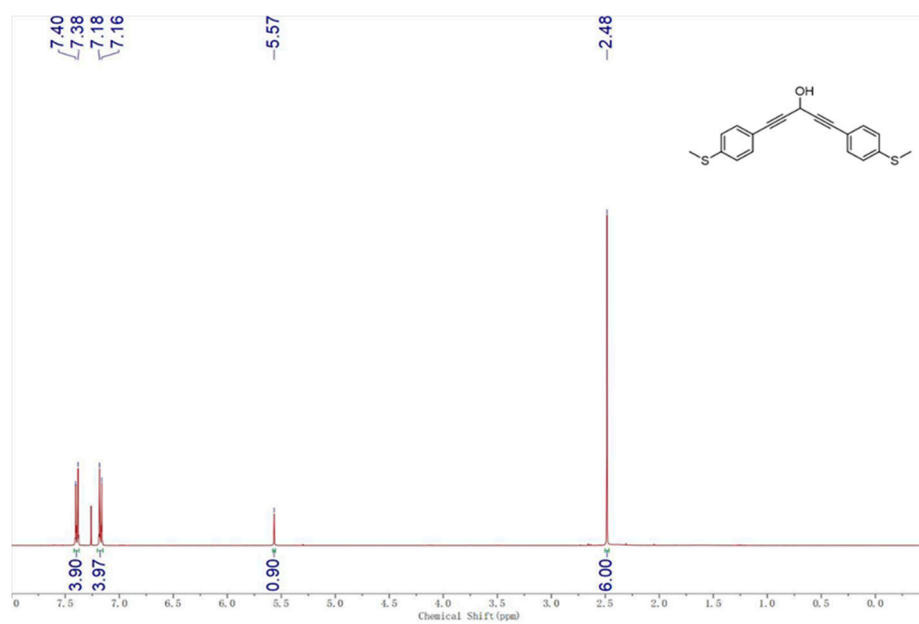


Figure S7 ¹H NMR Spectra of **4**.

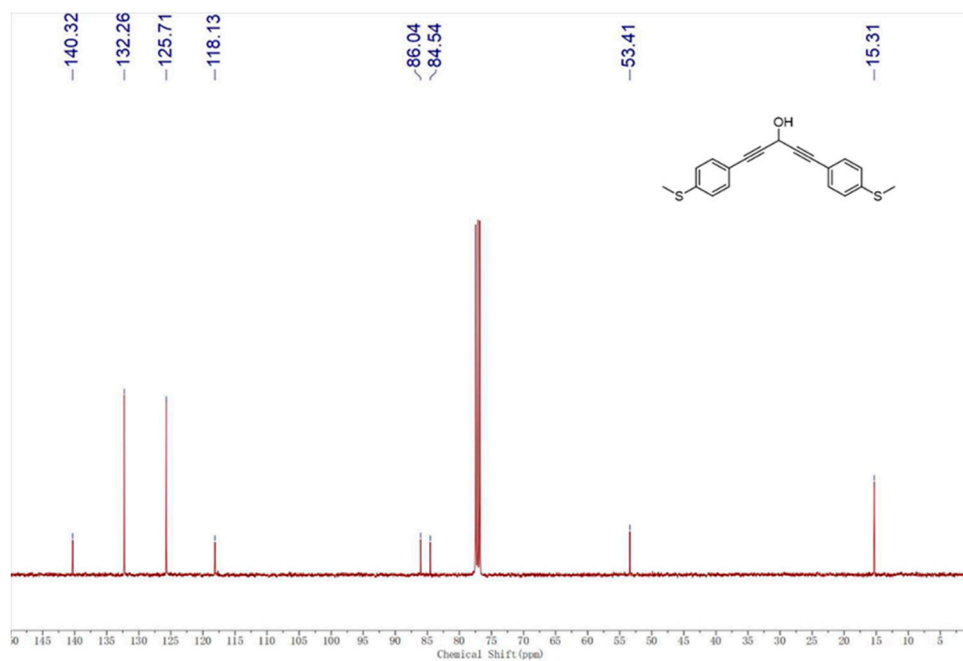


Figure S8 ¹³C NMR Spectra of **4**.

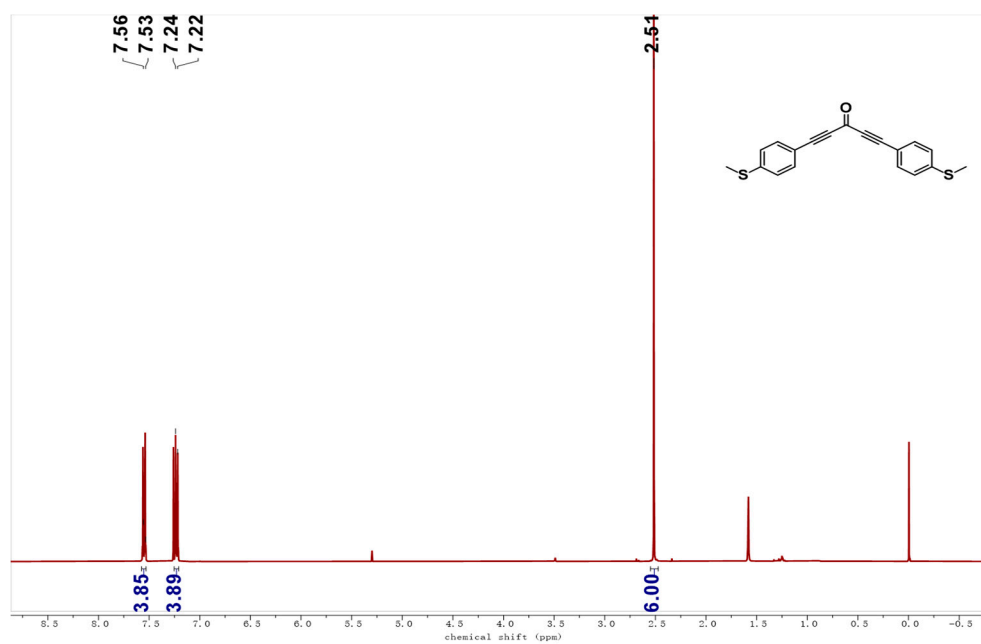


Figure S9 ¹H NMR Spectra of 5.

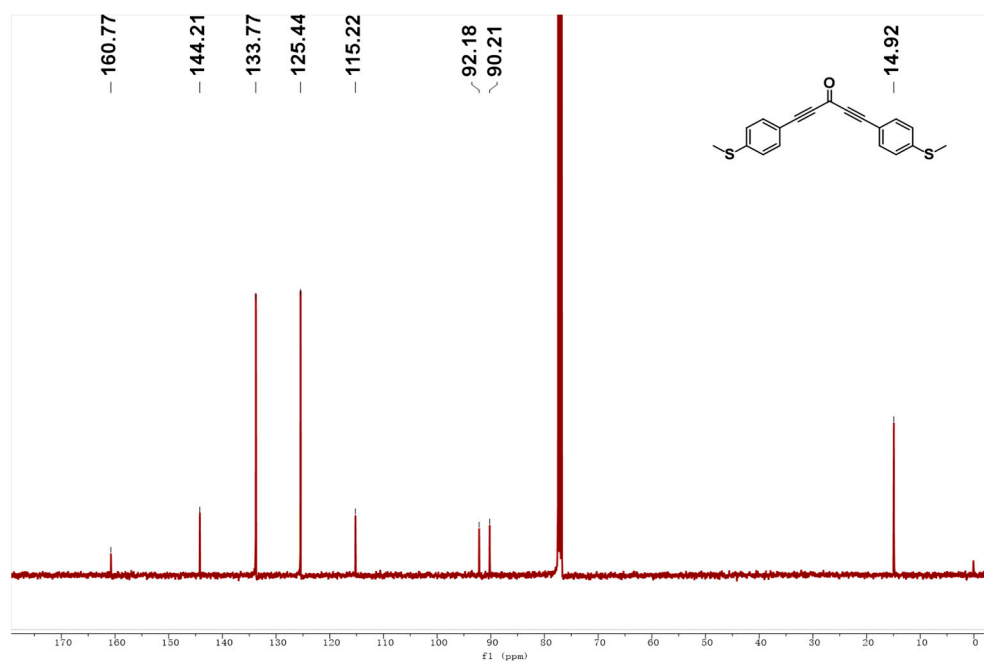


Figure S10 ¹³C NMR Spectra of 5.

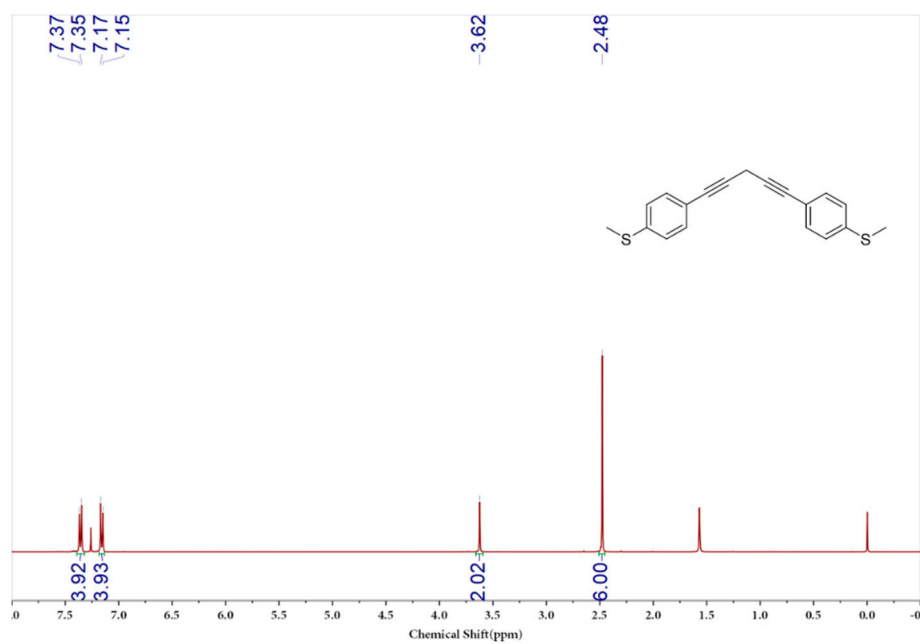


Figure S11 ¹H NMR Spectra of **6**.

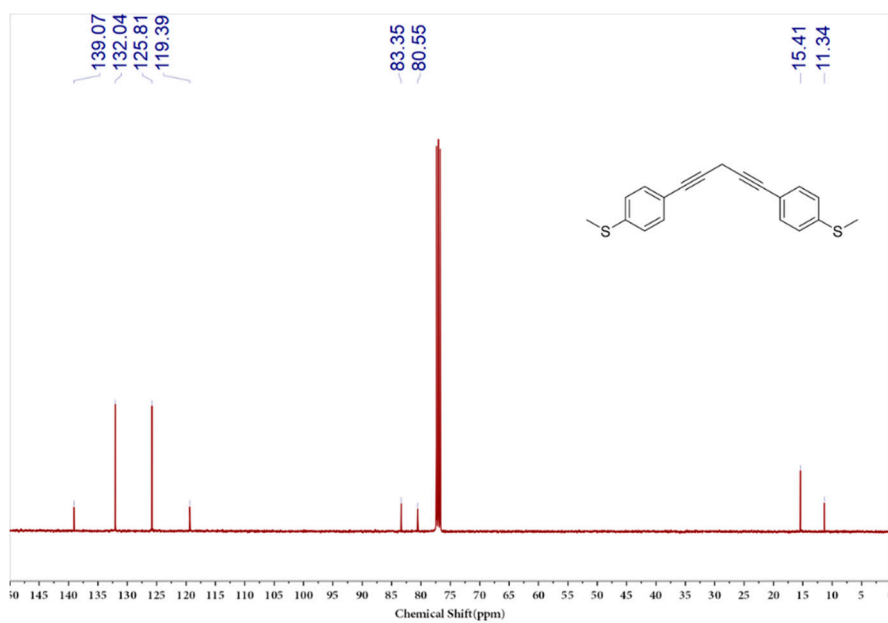


Figure S12 ¹³C NMR Spectra of **6**.

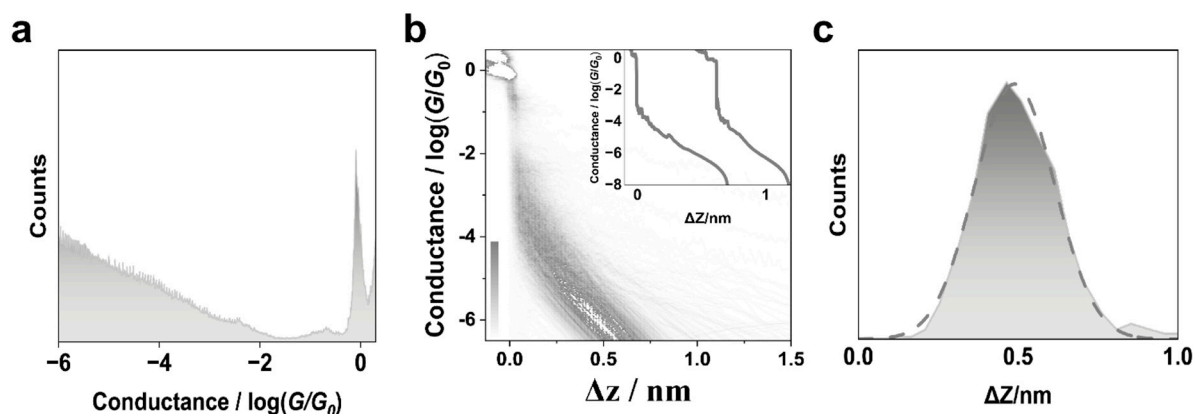


Figure S13 One-dimension conductance histogram, two-dimension conductance histogram and the relative displacement distribution determined from $10^{0.3} G_0$ to $10^{-6.0} G_0$ using pure solvent for the calibration of the stretching rate of the electrodes pair.

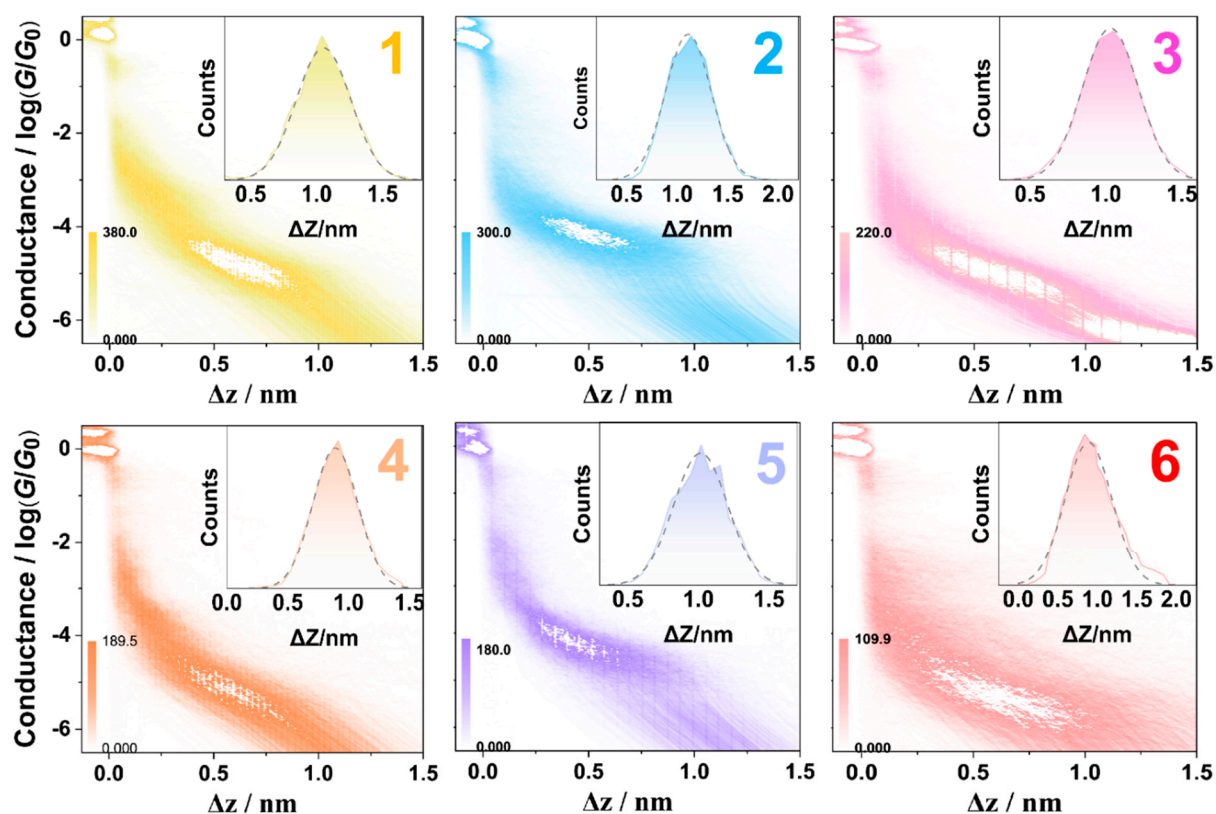


Figure S14 Two-dimensional conductance histograms and statistical junction lengths of molecules 1-6.

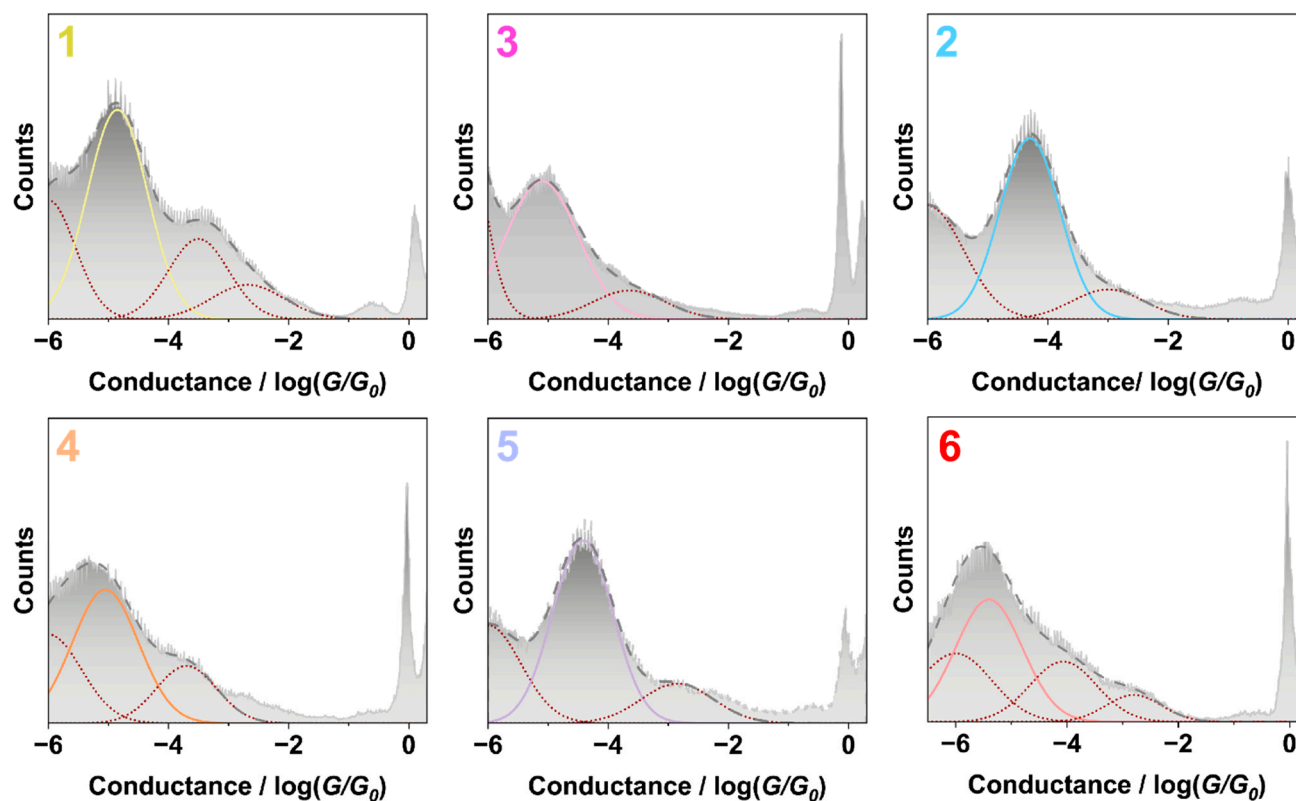


Figure S15 One-dimensional histograms of molecules **1-6** with the fitting curves of background (dashed brown lines), multimodal fitting (dashed grey lines) and the most probable conductance (colored peaks).

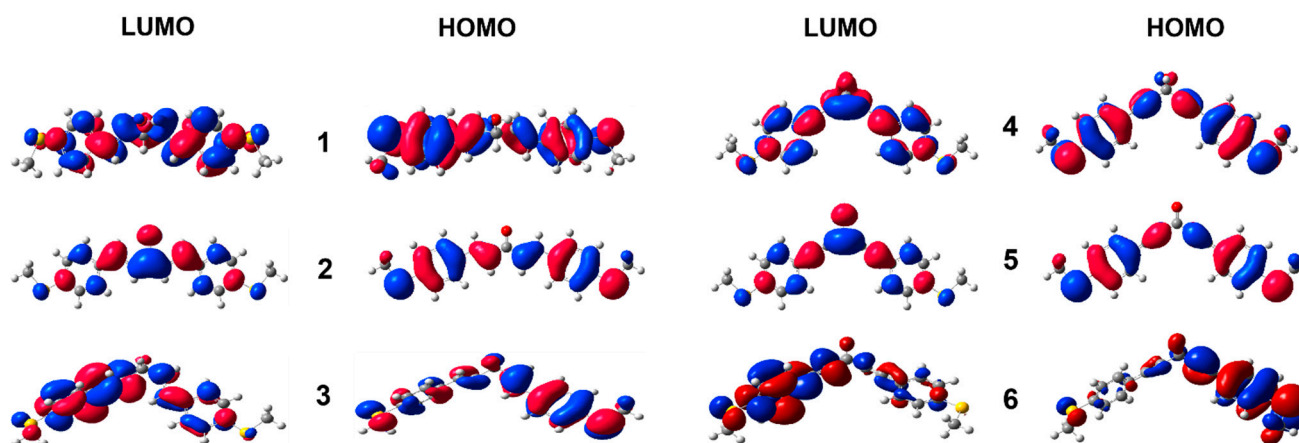


Figure S16 Distributions of frontier molecular orbitals of HOMO and LUMO of molecules **1-6** in the previous report [1]. Reproduced with permission from

ref. [1]. 2024 American Chemical Society.

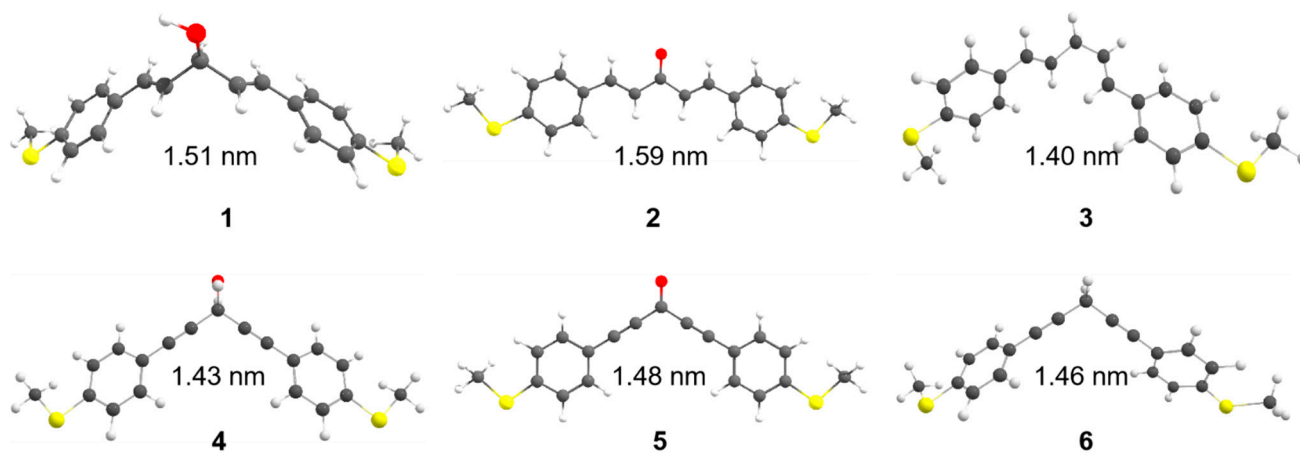


Figure S17 Theoretically optimized molecular configurations and corresponding S-S distances of molecules **1-6**.



Figure S18 Theoretical model used for transport calculations of single-molecule device.

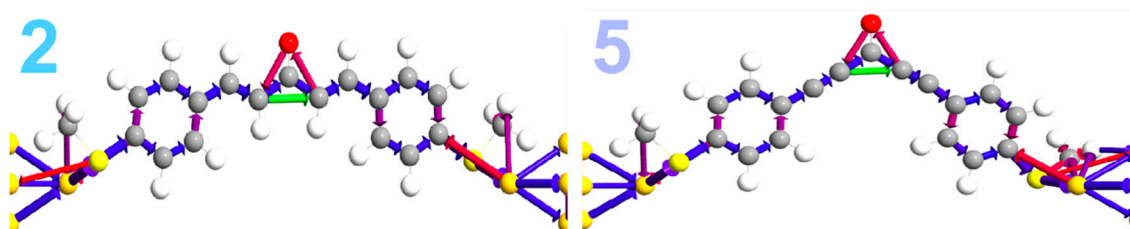


Figure S19 Transmission pathways of molecules **2** and **5**

Notes and references

1. Guo, M. M.; Jiang, Y. X.; Wang, J. Y.; Chen, Z. N.; Hou, S. M.; Zhang, Q. C., Effectively Enhancing the Conductance of Asymmetric Molecular Wires by Aligning the Energy Level and Symmetrizing the Coupling. *Langmuir* **2024**, 40, (7), 3759-3765.
<http://dx.doi.org/10.1021/acs.langmuir.3c03530>