Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information-*Ab Initio* Study Revealing Remarkable Oscillatory Effects and Negative Differential Resistance in the Molecular Device of Silicon Carbide Chains

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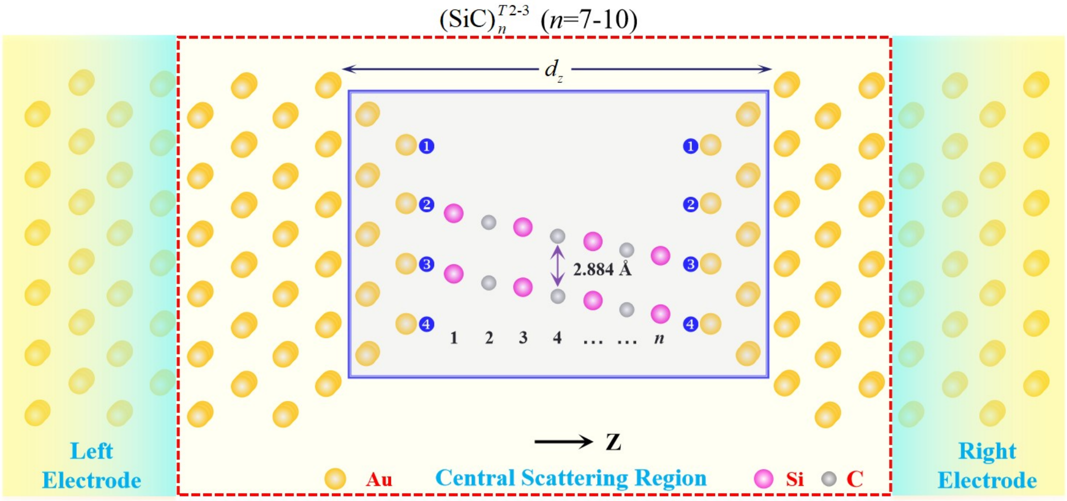

Fig. S1. The structure of $(\mathrm{SiC})_n^{T2-3}$ (n=7-10) with double inclined SiC chains, which take sites 2 and 3 on the left electrode, 3 and 4 on the right electrode, to bridge the two semi-infinite Au (100)-3×6 electrodes.

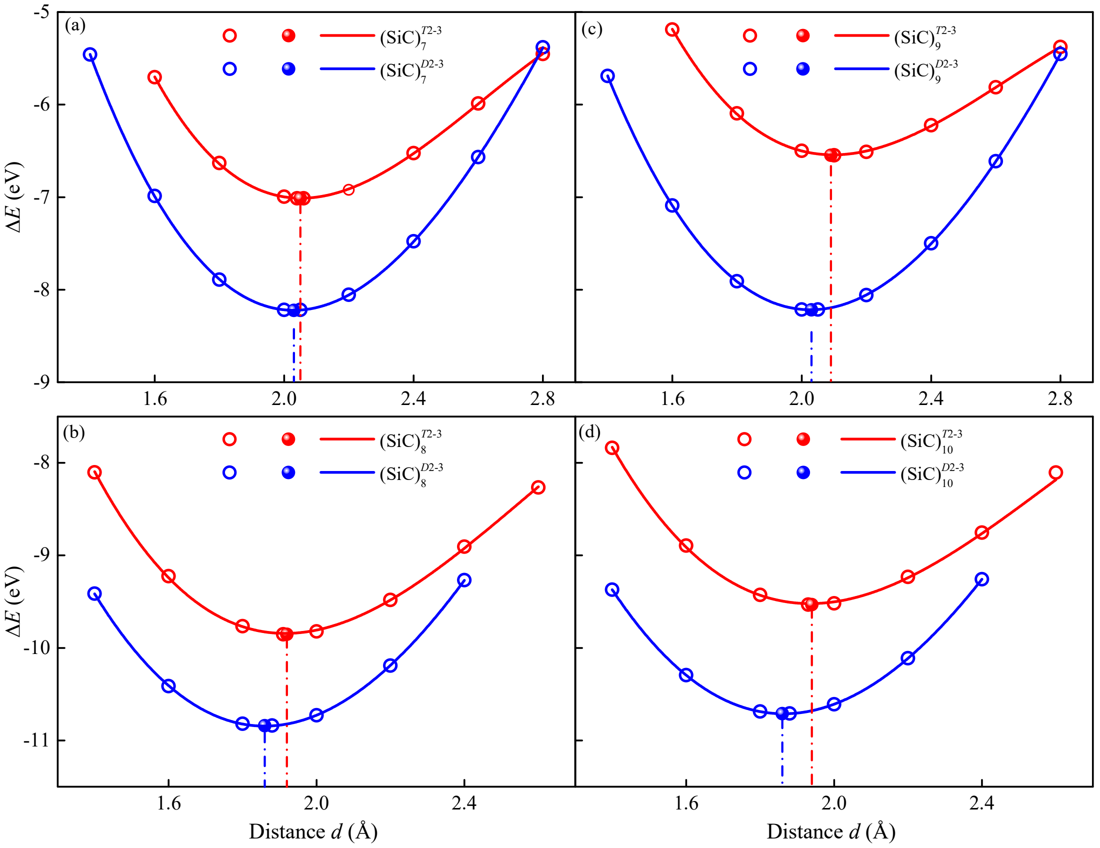

Fig. S2 The cohesion energy Δ*E* as a function of distance *d* in $(\mathrm{SiC})_n^{T2-3}$ and $(\mathrm{SiC})_n^{D2-3}$ (*n*=7-10). The empty symbols represent Δ*E* at different distances. The solid ball stands for the point close to the minimum of Δ*E*, and its abscissa corresponds to the optimal distance (dotted vertical line) of the structure at equilibrium. The solid line is a numerical fitting to the data.

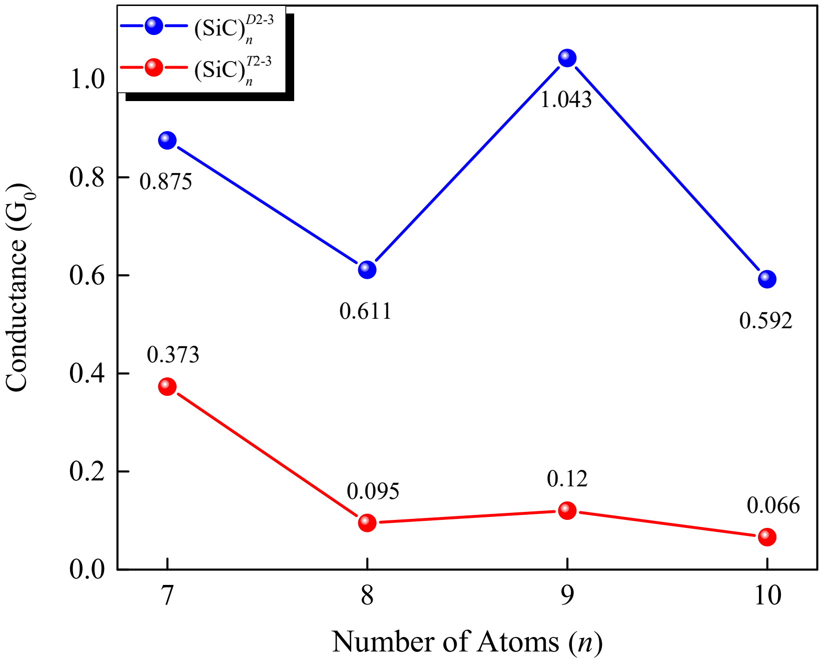

Fig. S3 The conductance of $(\mathrm{SiC})_n^{T2-3}$ and $(\mathrm{SiC})_n^{D2-3}$ (n=7-10) as a function of the number of atoms n in the chains.

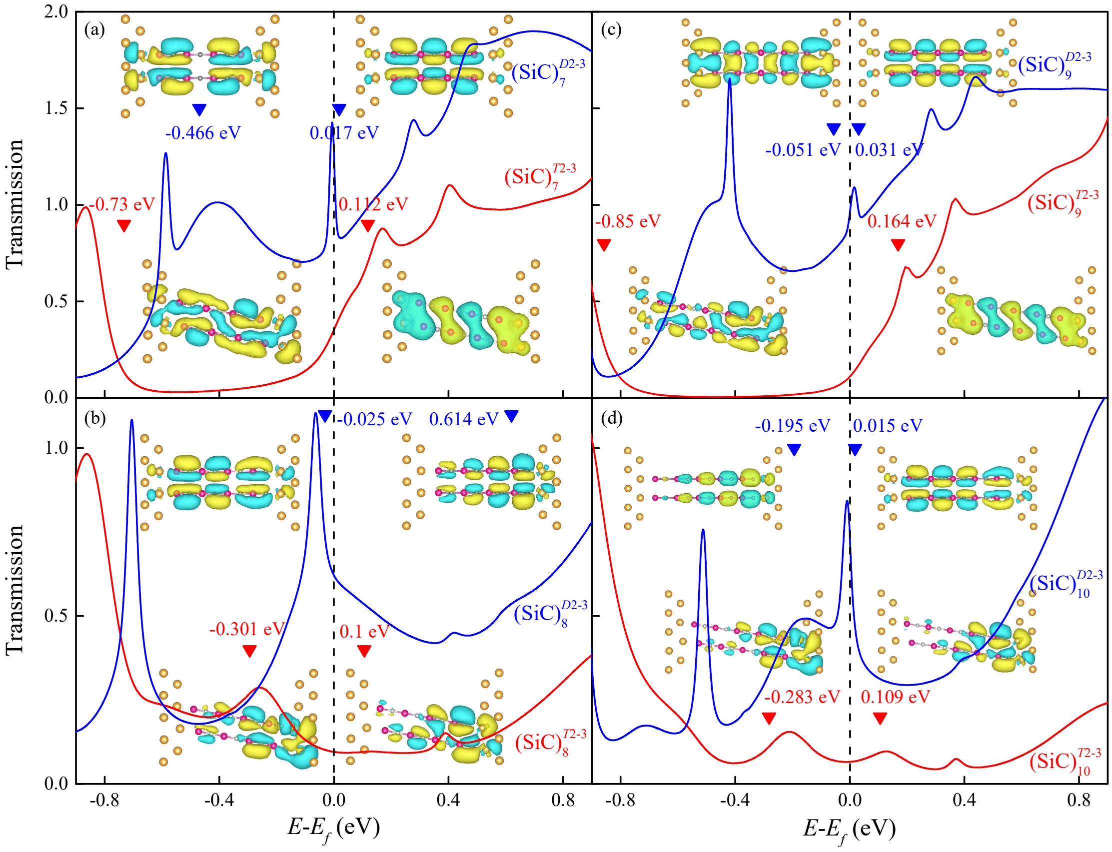

Fig. S4 The transmission spectra $T(E,V)$ of $(\mathrm{SiC})_n^{T2-3}$ and $(\mathrm{SiC})_n^{D2-3}$ (n=7-10) as a function of the electron energy at zero bias. The triangles denote eigenvalues of the MPSH states of HOMO and LUMO. Their spatial distributions are displayed nearby. The isovalue is set to be 0.03.

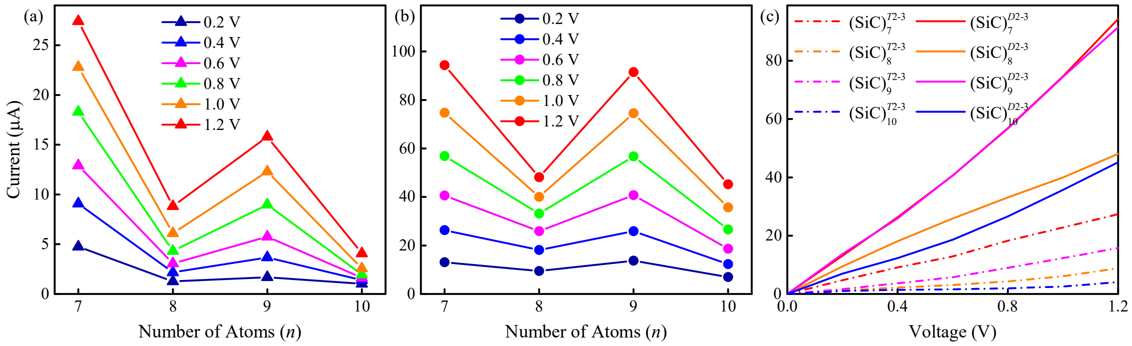

Fig. S5 The current of (a) $(\mathrm{SiC})_n^{T2-3}$ and (b) $(\mathrm{SiC})_n^{D2-3}$ (n=7-10) as a function of the number of atoms n. (c): The current as a function of bias in the range of 0 to 1.2 V.

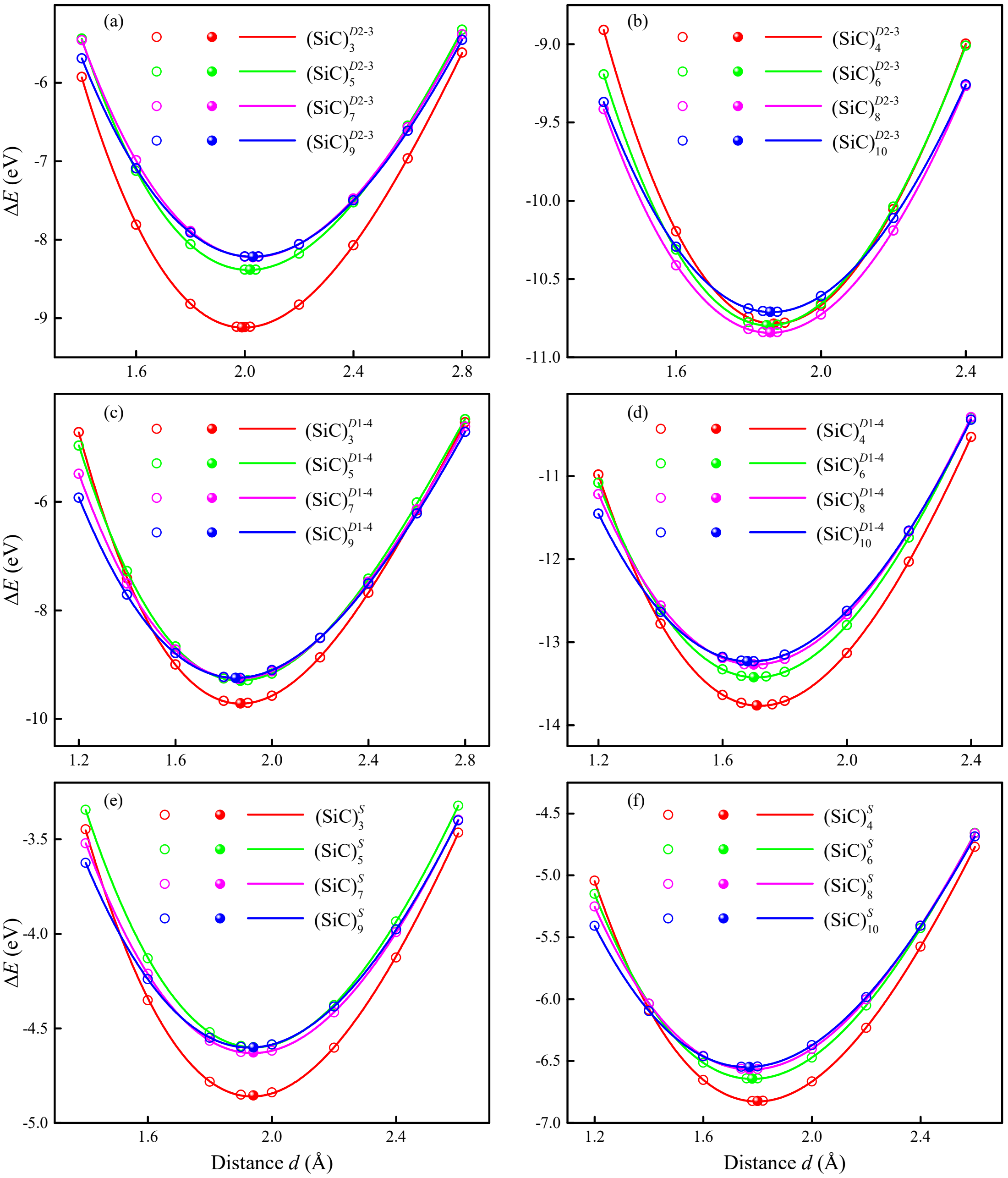

Fig. S6 The cohesion energy ΔE as a function of distance d: (a-b) $(\mathrm{SiC})_n^{D2-3}$, (c-d) $(\mathrm{SiC})_n^{D1-4}$, and (e-f) $(\mathrm{SiC})_n^{S}$ (n=3-10). The empty circles represent ΔE at different distance. The solid ball stands for the point close to the minimum of ΔE, and its abscissa corresponds to the optimal distance used in the structure at equilibrium. The solid line is a numerical fitting to the data.

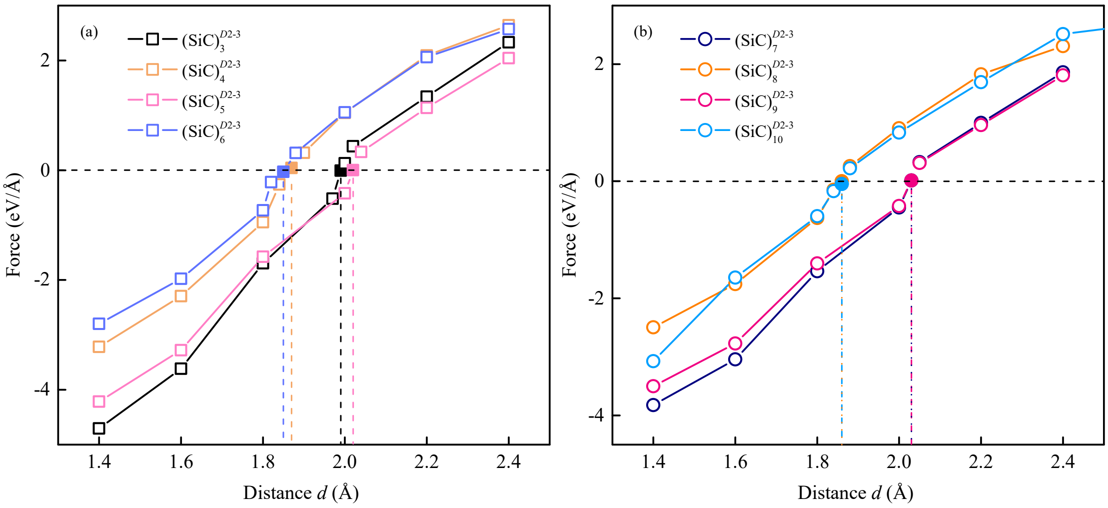

Fig S7 The tensile force F_z calculated as a function of distance d for $(\mathrm{SiC})_n^{D2-3}$ (n=3-10): (a) n=3-6, (b) n=7-10. The positive (negative) values of F_z signify that the resultant inter-atomic force is repulsive (attractive), and the molecular junction is under compressive (tensile) strain. The solid square represents the F_z close to zero, and the corresponding dotted vertical line denotes the optimal distance for $(\mathrm{SiC})_n^{D2-3}$, where the lines of n=8, 10 and n=7, 9 are coincide. The optimal distance employed in the simulation corresponds to the point close to near-zero tensile force F_z and the local minimum of cohesion energy ΔE, indicating that such structure is at equilibrium, approximating its most stable state in $(\mathrm{SiC})_n^{D2-3}$.

Table S1 Optimized bond lengths (in Å) in systems $(\mathrm{SiC})_n^{D2-3}$, $(\mathrm{SiC})_n^{D1-4}$, and $(\mathrm{SiC})_n^{S}$ (n=3-10). The column indicates the number of atoms in the chains. The row indicates the distance between the Si and C atoms in the chains. The three rows of $d_{\text{Si-C}}$, $d_{\text{Au-Si}}$ and $d_{\text{Au-C}}$ are the average bond lengths of Si-C, Au-Si, and Au-C, respectively. Since the bond lengths involved in the upper and lower chains are the same, only the bond lengths in the upper chain are listed. The last row is the optimal distance d between the end Si/C atoms in SiC chains and the pyramidal Au atoms.

Bonds in $(\mathrm{SiC})_n^{D2\text{-}3}$	n							
	3	4	5	6	7	8	9	10
Si1-C2	1.669	1.663	1.666	1.661	1.667	1.666	1.667	1.668
C2-Si3	1.670	1.683	1.666	1.662	1.666	1.662	1.663	1.662
Si3-C4		1.680	1.665	1.650	1.655	1.652	1.654	1.656
C4-Si5			1.666	1.670	1.655	1.652	1.652	1.650
Si5-C6				1.678	1.665	1.650	1.652	1.650
C6-Si7					1.667	1.669	1.654	1.652
Si7-C8						1.679	1.662	1.648
C8-Si9							1.667	1.669
Si9-C10								1.679
$d_{\text{Si-C}}$	1.670	1.675	1.666	1.664	1.663	1.661	1.659	1.659
$d_{\text{Au-Si}}$	2.278	2.289	2.286	2.281	2.289	2.286	2.287	2.282
$d_{\text{Au-C}}$		1.933		1.929		1.929		1.929
d	1.99	1.87	2.02	1.85	2.03	1.86	2.03	1.86

Bonds in $(\mathrm{SiC})_n^{D1-4}$	n							
	3	4	5	6	7	8	9	10
Si1-C2	1.670	1.643	1.659	1.645	1.658	1.648	1.658	1.648
C2-Si3	1.671	1.682	1.656	1.667	1.654	1.664	1.654	1.66
Si3-C4		1.651	1.657	1.632	1.642	1.632	1.642	1.633
C4-Si5			1.659	1.667	1.642	1.654	1.641	1.649

Si5-C6				1.653	1.655	1.632	1.640	1.631
C6-Si7					1.658	1.665	1.641	1.649
Si7-C8						1.655	1.654	1.632
C8-Si9							1.658	1.662
Si9-C10								1.657
$d_{\mathrm{Si\text{-}C}}$	1.671	1.659	1.658	1.653	1.652	1.650	1.649	1.647
$d_{\mathrm{Au\text{-}Si}}$	2.245	2.256	2.245	2.252	2.247	2.257	2.248	2.251
$d_{\mathrm{Au\text{-}C}}$		1.907		1.902		1.901		1.900
d	1.87	1.71	1.87	1.70	1.86	1.70	1.85	1.68

Bonds in	n							
$(\mathrm{SiC})_n^S$	3	4	5	6	7	8	9	10
Si1-C2	1.670	1.647	1.659	1.647	1.659	1.649	1.660	1.652
C2-Si3	1.669	1.685	1.655	1.669	1.655	1.663	1.655	1.661
Si3-C4		1.652	1.655	1.632	1.643	1.633	1.643	1.635
C4-Si5			1.659	1.670	1.643	1.654	1.641	1.650
Si5-C6				1.651	1.654	1.632	1.641	1.633
C6-Si7					1.660	1.666	1.643	1.652
Si7-C8						1.653	1.654	1.632
C8-Si9							1.660	1.666
Si9-C10								1.654
$d_{\mathrm{Si\text{-}C}}$	1.670	1.661	1.657	1.654	1.652	1.650	1.650	1.648
$d_{\mathrm{Au\text{-}Si}}$	2.248	2.263	2.251	2.259	2.252	2.258	2.256	2.258
$d_{\mathrm{Au\text{-}C}}$		1.918		1.911		1.910		1.910
d	1.94	1.8	1.94	1.78	1.94	1.77	1.94	1.77

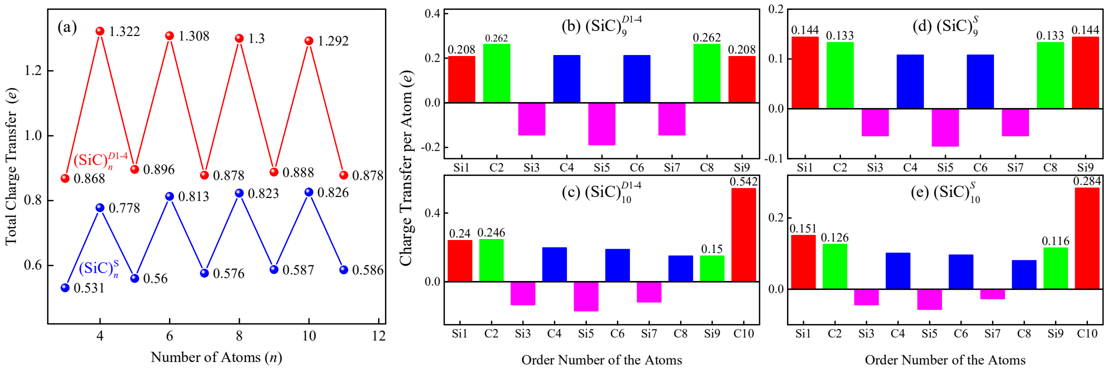

Fig. S8 (a) The total charge transferred in $(\mathrm{SiC})_n^{D1-4}$ and $(\mathrm{SiC})_n^{S}$ (n=3-11) as a function of number of atoms n in the chains. (b-e) Distribution of charge transferred along the chains in $(\mathrm{SiC})_n^{D1-4}$ and $(\mathrm{SiC})_n^{S}$ (n=9,10). Our calculation shows that the charge transferred per atom in the upper chain is the same as that in the lower one, thus twice the amount of charge per atom in one chain is illustrated.

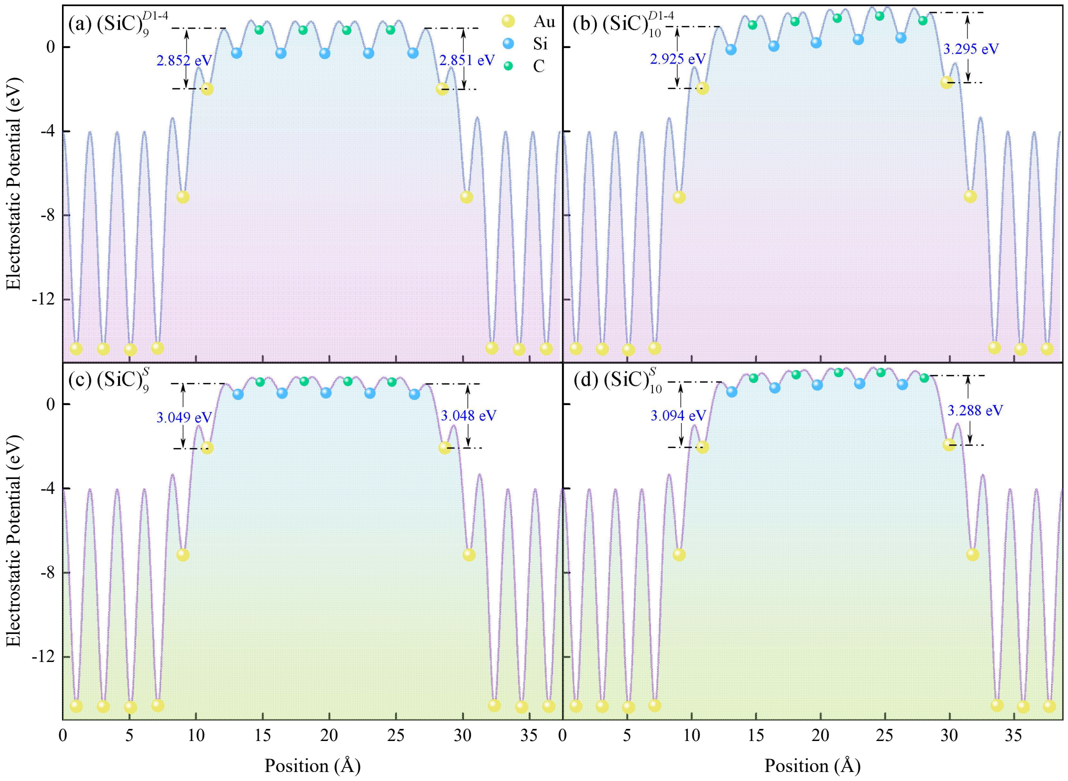

Fig. S9 The electrostatic potential as a function of position along the z direction in $(\mathrm{SiC})_n^{D1-4}$ and $(\mathrm{SiC})_n^{S}$ (n=9-10). The yellow, blue, and green balls represent the positions of tip Au, Si, and C atoms, respectively. The maximum barrier height at the molecule-electrode interface has been marked.

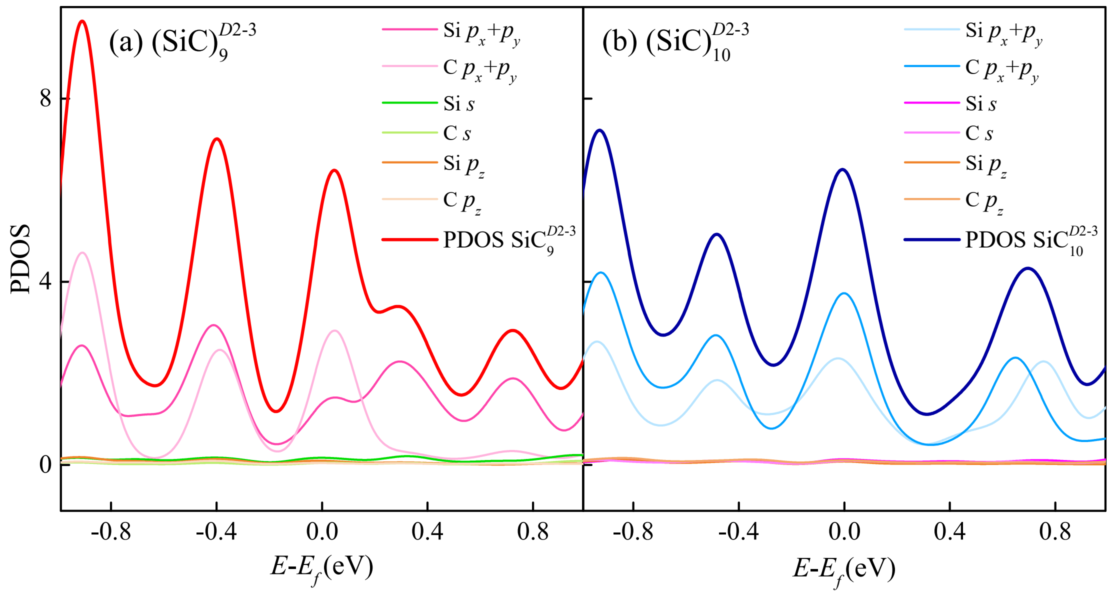

Fig. S10 The PDOS of $(\mathrm{SiC})_n^{D2-3}$ (n=9, 10) as a function of electron energy near the E$_f$.

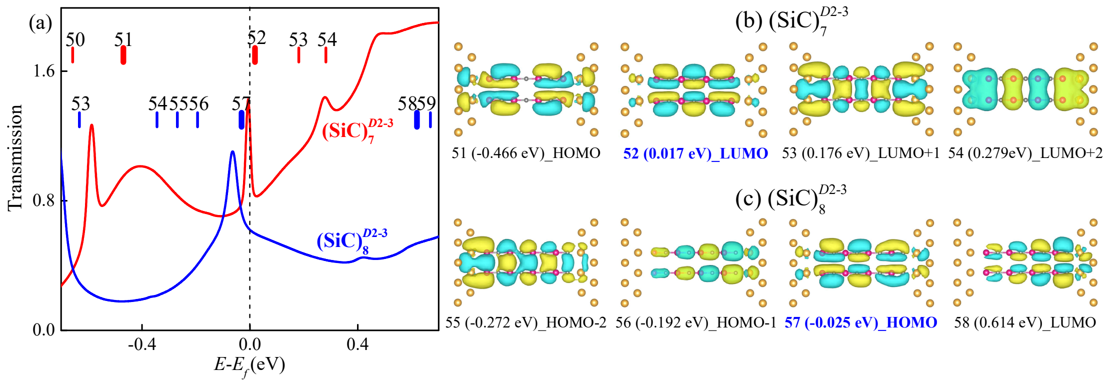

Fig. S11 (a) The transmission spectra T (E,V) of $(\mathrm{SiC})_n^{D2-3}$ (n=7, 8) as a function of electron energy at zero bias. The short lines denote eigenvalues of the MPSH states, where the thick ones stand for HOMO and LUMO, respectively. (b-c) The spatial distribution of four MPSH states of $(\mathrm{SiC})_n^{D2-3}$ (n=7, 8) close to the E_f. The molecular orbital lies closest to the E_f is highlighted in blue. The isovalue is set to be 0.03.

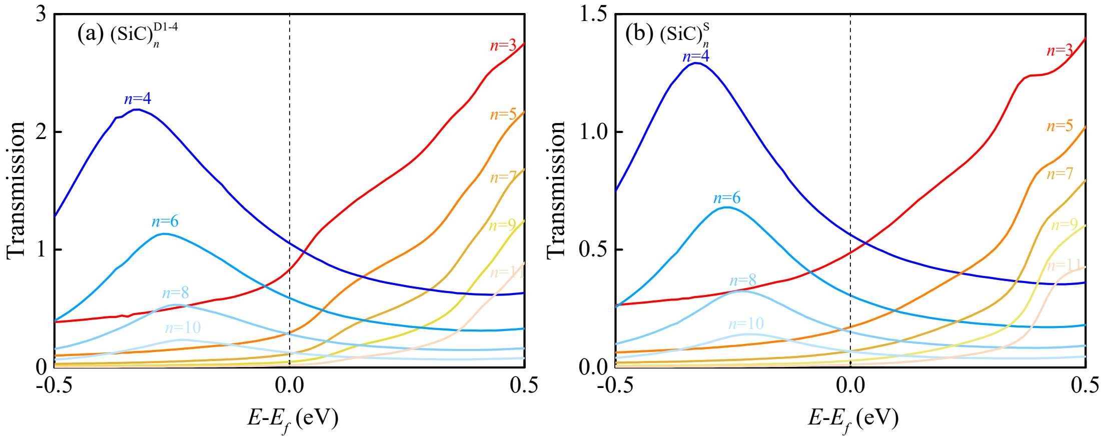

Fig. S12 (a-b) The transmission spectra *T* (*E*,*V*) of $(\mathrm{SiC})_n^{D1-4}$ and $(\mathrm{SiC})_n^{S}$ (*n*=3-11) as a function of electron energy at zero bias, respectively.

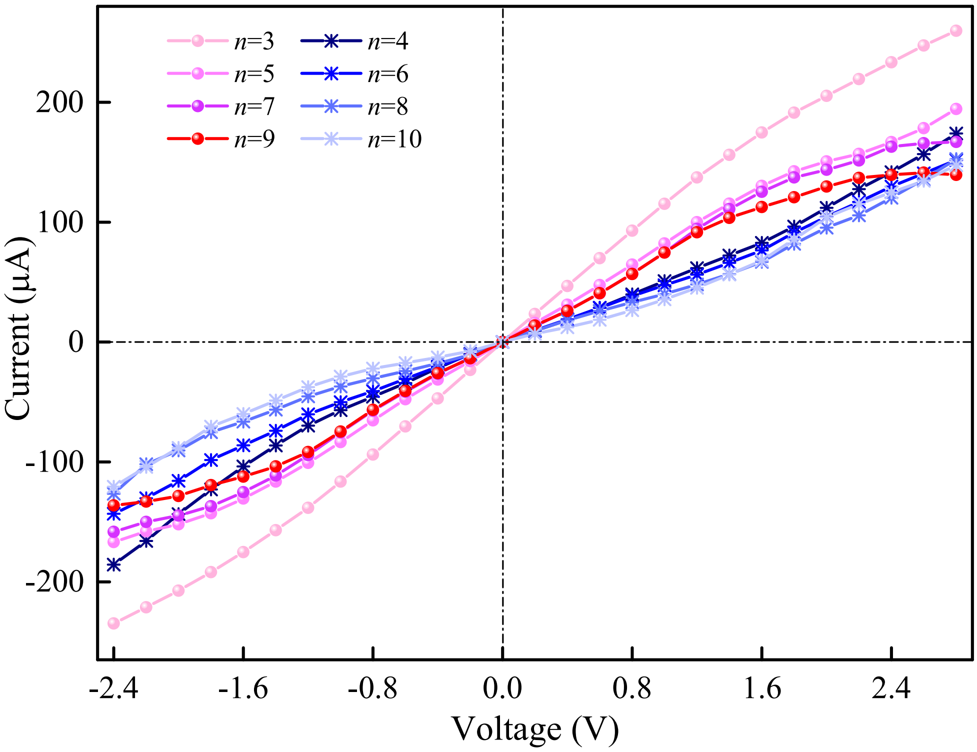

Fig. S13 The current of $(\mathrm{SiC})_n^{D2-3}$ (*n*=3-10) as a function of bias in the range of -2.4 to 2.8 V.

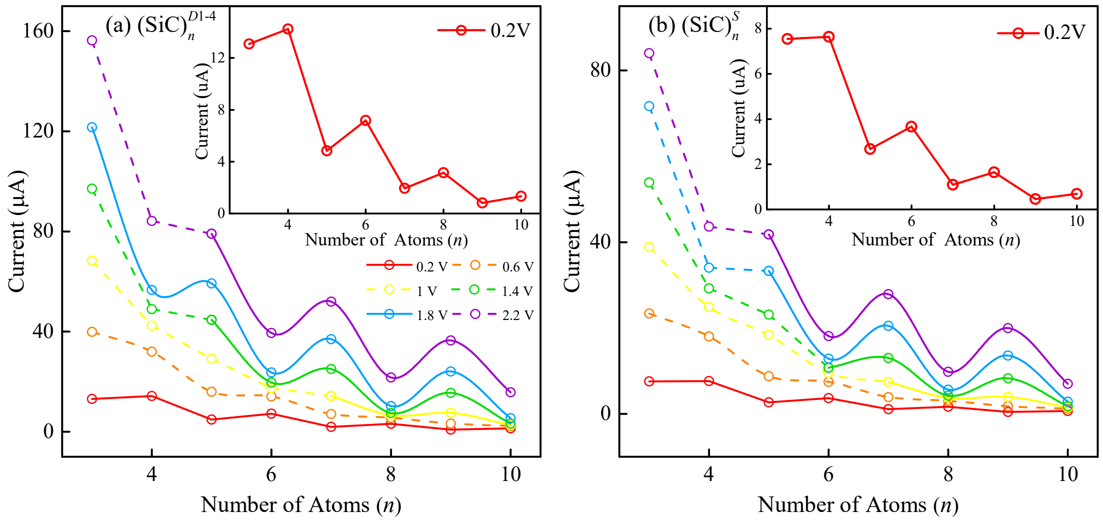

Fig. S14 The current of $(\text{SiC})_n^{D1-4}$ and $(\text{SiC})_n^{S}$ (n=3-10) as a function of number of atoms n. The partial disturbance of the current oscillation is given by dashed lines. The insets are the occasional current oscillation at 0.2 V.

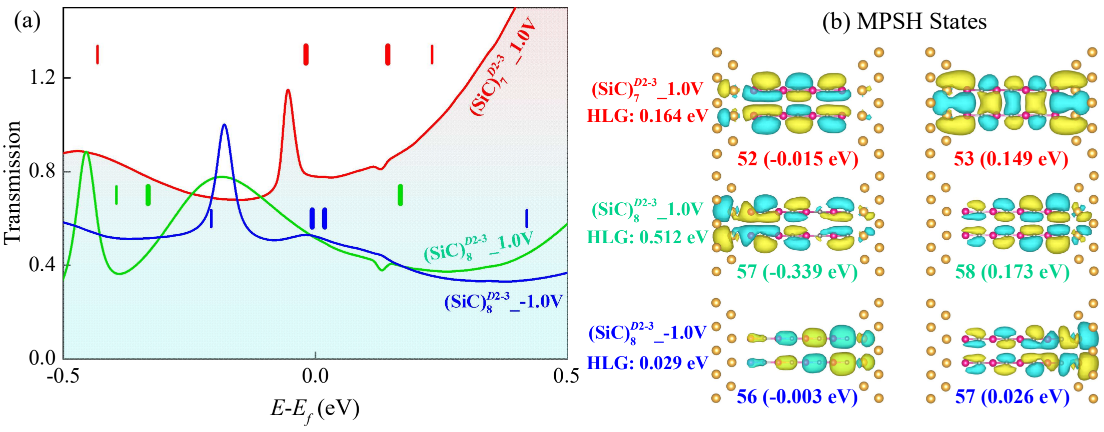

Fig. S15 (a) The transmission spectra $T(E,V)$ of $(\text{SiC})_n^{D2-3}$ (n=7, 8) under the typical biases of ±1.0 V. The shaded area stands for the transmission integral within the bias window. The short lines denote eigenvalues of the MPSH states, where the thick ones stand for HOMO and LUMO, respectively. (b) The spatial distribution of HOMO and LUMO of $(\text{SiC})_n^{D2-3}$ (n=7, 8) at ±1.0 V. The isovalue is set to be 0.03.

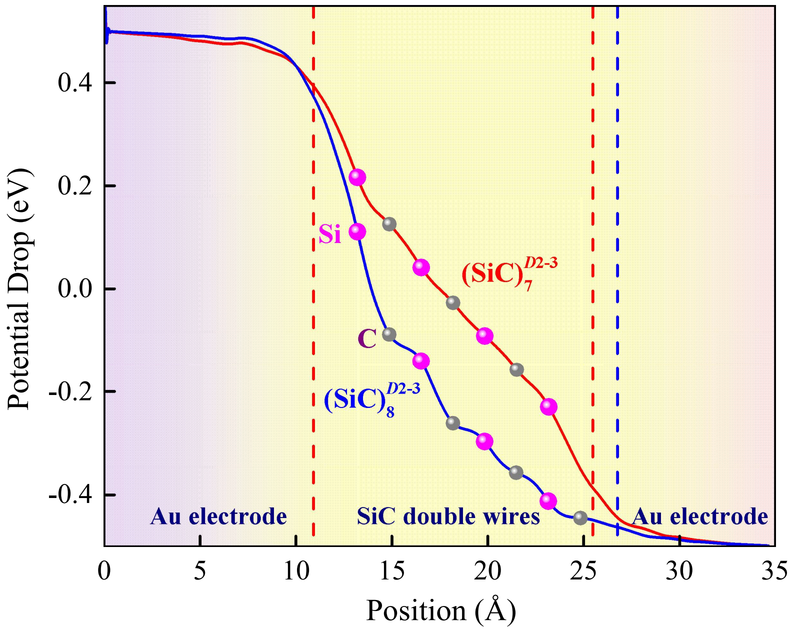

Fig. S16 The potential drop along the length of $\left(\mathrm{SiC}\right)_n^{D2-3}$ (n=7,8) at 1.0 V. The red and blue dashed lines indicate the positions of the tip Au atoms in $\left(\mathrm{SiC}\right)_n^{D2-3}$ (n=7,8), respectively. The small circles represent the positions of the atoms in the chains, and the atomic symbols are given nearby.

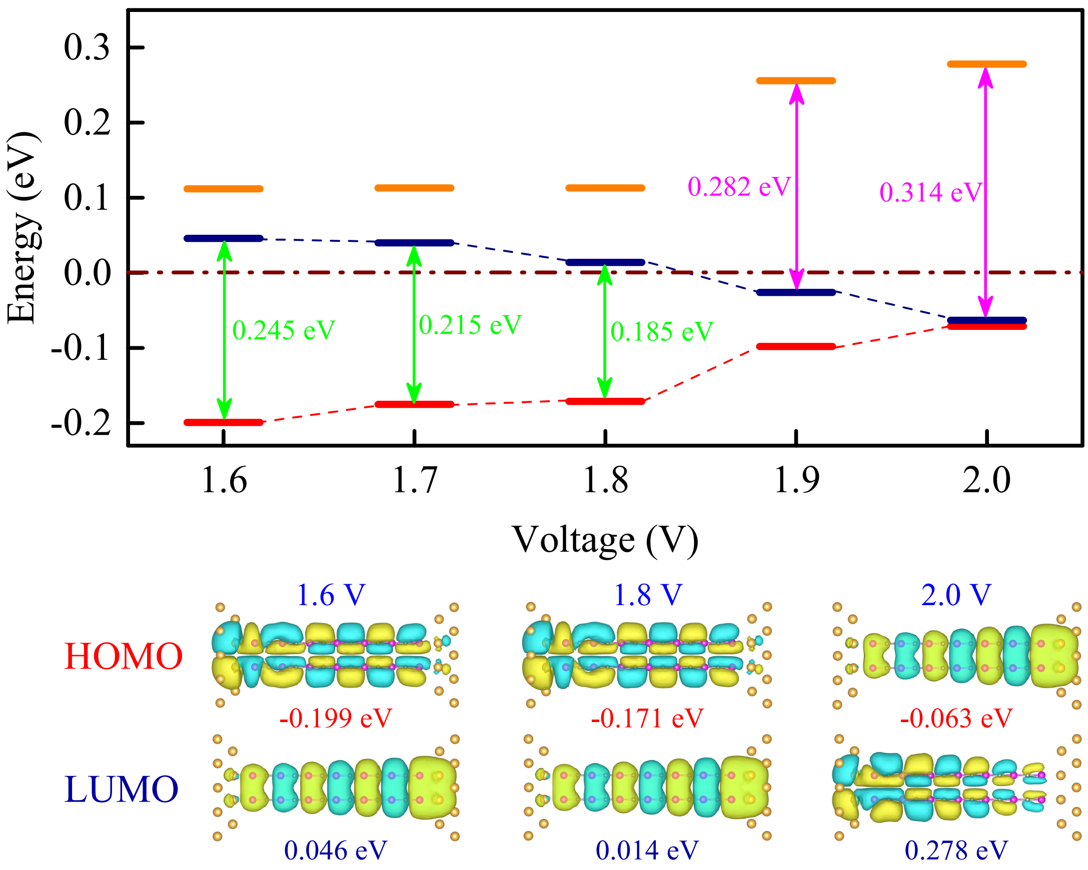

Fig. S17 The energy of molecular states close to the E_f as a function of bias near the NDR in $\left(\mathrm{SiC}\right)_{13}^{D2-3}$. The value of HLG is labeled. The spatial distributions of HOMO and LUMO are shown below.