

Electronic Supplementary Information

Adsorbing the PVDF Polymer via Noncovalent Interaction to Effectively Tune the Electronic and Magnetic Properties of Zigzag SiC Nanoribbons

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(I) The computational test on the sampled wider 10-zSiCNR systems modified by the PVDF polymer

Our computational results reveal that surface-adsorbing the PVDF polymer via noncovalent interaction can break the magnetic degeneracy between the FM and AFM states for the pristine 8-zSiCNR. Further, we also perform the computational test on the wider 10-zSiCNR, where three joint structures are sampled involving FH-10-zSiCNR-center-*l* and FH-10-zSiCNR-eSi-*l* systems with the fluorine-hydrogen (FH) side of PVDF interacting with the zigzag SiC chain at the ribbon center or Si-edge, as well as HH-10-zSiCNR-center-*r* with the full-hydrogen (HH) side of PVDF interacting with the six-membered ring chain at the center, as shown in Figures S1d-S1f. It is found that the magnetic degeneracy can be still broken when depositing the PVDF polymer on the surface of the wider 10-zSiCNR, where FH-10-zSiCNR-center-*l* is the FM metallicity, while FH-10-zSiCNR-eSi-*l* and HH-10-zSiCNR-center-*r* exhibit the AFM half-metallic behavior (Table S1 and Figure S1), consistent with ones of the parallel 8-zSiCNR systems modified by the PVDF polymer. Obviously, surface-adsorbing PVDF polymer can effectively break the magnetic degeneracy of zSiCNRs. In this study, in view of the computation accuracy and efficiency, the medium-sized 8-zSiCNR is selected as a prototype system to detailedly investigate the effect of surface-adsorbing PVDF polymer on the electronic and magnetic properties of zSiCNRs, as presented in the main text, where 8-zSiCNR is directly denoted as zSiCNR in the related discussions for the convenience.

Table S1 The relative energies ΔE (meV) of different magnetic couplings to the ground state, the total magnetic moment M_{tot} , the adsorption energy E_{ad} , the distance $d_{\text{SiCNR-PVDF}}$ between PVDF and zSiCNR, the electronic property, the band gap in minority channel for the half-metallicity for the modified 10-zSiCNR systems. The NM, FM and AFM represent the nonmagnetic, ferromagnetic and antiferromagnetic spin couplings, respectively.

Systems	ΔE (meV)			M_{tot} (μB)	$d_{\text{SiCNR-PVDF}}$ (\AA)	E_{ad} (eV)	Electronic property	The gap in minority channel (eV)
	NM	FM	AFM					
10-zSiCNR	180.2	0.0	0.0	2.2/0.0	-	-	-	-
FH-10-zSiCNR-center- <i>l</i>	166.3	0.0	2.4	2.1	2.90	-1.205	Metallicity	-
FH-10-zSiCNR-eSi- <i>l</i>	191.2	1.3	0.0	0.0	2.86	-0.961	Half-metallicity	0.582
HH-10-zSiCNR-center- <i>r</i>	178.7	2.9	0.0	0.0	2.90	-0.870	Half-metallicity	0.560

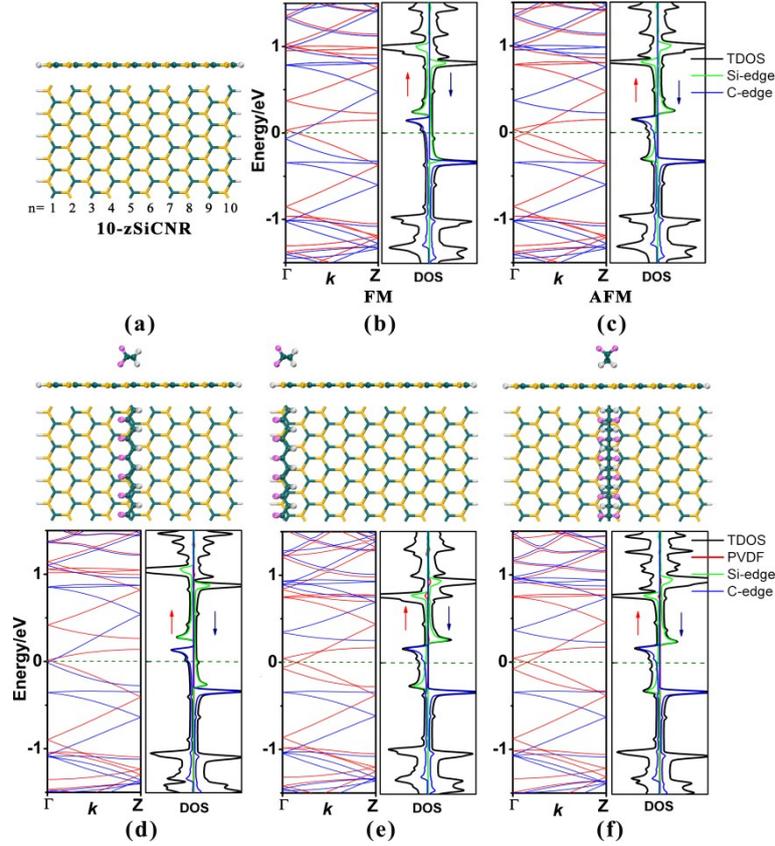


Figure S1. The geometries, band structures, and corresponding density of states of pristine 10-zSiCNR (a-c) as well as the PVDF-modified 10-zSiCNR systems: (d) FH-10-zSiCNR-center-*l*, (e) FH-10-zSiCNR-eSi-*l* and (f) HH-10-zSiCNR-center-*r*. The red and blue lines in band structures denote the spin-up (\uparrow) and spin-down (\downarrow) channels, respectively. The Fermi-level is set as zero and indicated by the green dotted line.

(II) The computations on the modified 8-zSiCNR systems with the PVDF polymer at the center

When adsorbing the PVDF polymer over the center of 8-zSiCNR, we consider all three possible adsorption sites involving one six-membered ring chain (denoted by “*r*”) at the blue area, and two component zigzag SiC chains (denoted by “*l*”) represented by the ball-and-stick model, as illustrated in Figure S2. Thus three conformations ((I)~(III)) of PVDF-modified 8-zSiCNR can be obtained in each spatial series (FH, HF, HH or FF), as shown in Figure S3. By comparison, we can find that the most stable conformations among the FH, HF, HH and FF series are a-I, b-II, c-II and d-I, respectively (Figure S3), which have been accordingly named as FH-zSiCNR-center-*l*, HF-zSiCNR-center-*r*, HH-zSiCNR-center-*r* and FF-zSiCNR-center-*l* in the main text.

(III) The changes of geometrical parameters and bader charges on the involved atoms before and after the interaction of PVDF polymer with 8-zSiCNR

The computed geometrical parameters of isolated PVDF polymer and ones in the most favorable joint configurations are presented in Table S2, Figures S4b and S4c. It is revealed that all the bond lengths and angles of PVDF in joint structures are close to the corresponding one of isolated PVDF, indicating that the change of geometrical structure of PVDF polymer after interacting with zSiCNR is very small owing to the rigid σ -framework. Similar situation can be also observed in the computed bader charges on the involved atoms in the PVDF polymer, that is, the bader charges on PVDF in joint systems have the similar distribution to one of the isolated PVDF polymer (Figure S4d), which is mainly attributed to the large electronegativity difference between the involved H and F atoms. Combining the related geometrical parameters and bader charges, we can reasonably conclude that the change of dipole moment for the PVDF polymer is small before and after its interaction with zSiCNR to form all the joint systems.

Table S2 The main geometrical parameters involving the related bond lengths and angles for the isolated PVDF polymer and ones in the most favorable joint configurations.

Systems	d_{C-H} (Å)	d_{C-F} (Å)	d_{C-C} (Å)	$\angle HCH$ (°)	$\angle FCF$ (°)	$\angle CCC$ (°)	$\angle CCH$ (°)	$\angle CCF$ (°)		
Isolated PVDF	1.100	1.382	1.382	1.541	1.539	108.7	109.1	114.1	108.5	109.2
FH-zSiCNR-eSi- <i>l</i>	1.100	1.380	1.395	1.540	1.537	109.0	106.6	114.3	108.2	109.8
FH-zSiCNR-center- <i>l</i>	1.100	1.384	1.394	1.539	1.538	109.1	106.6	114.3	108.4	109.4
FH-zSiCNR-eC- <i>l</i>	1.100	1.384	1.386	1.540	1.537	109.6	106.8	114.5	108.9	109.6
HF-zSiCNR-eSi- <i>r</i>	1.100	1.380	1.390	1.540	1.538	108.7	107.1	114.2	108.7	109.6
HF-zSiCNR-center- <i>r</i>	1.100	1.380	1.388	1.539	1.538	108.9	106.8	114.2	108.6	108.9
HF-zSiCNR-eC- <i>r</i>	1.100	1.385	1.385	1.541	1.538	109.1	106.3	114.3	108.7	109.6
HH-zSiCNR-eSi- <i>r</i>	1.100	1.383	1.386	1.539	1.538	108.0	107.3	114.2	108.6	109.3
HH-zSiCNR-center- <i>r</i>	1.100	1.384	1.385	1.540	1.538	108.0	107.2	114.2	108.9	109.2
HH-zSiCNR-eC- <i>r</i>	1.100	1.388	1.389	1.538	1.536	107.7	107.1	114.3	108.9	108.8
FF-zSiCNR-eSi- <i>l</i>	1.100	1.381	1.386	1.539	1.537	108.6	106.5	114.3	108.7	109.2
FF-zSiCNR-center- <i>l</i>	1.099	1.380	1.388	1.540	1.538	108.8	106.9	114.2	108.9	108.4
FF-zSiCNR-eC- <i>l</i>	1.100	1.382	1.383	1.541	1.540	108.8	107.4	114.2	108.9	109.2

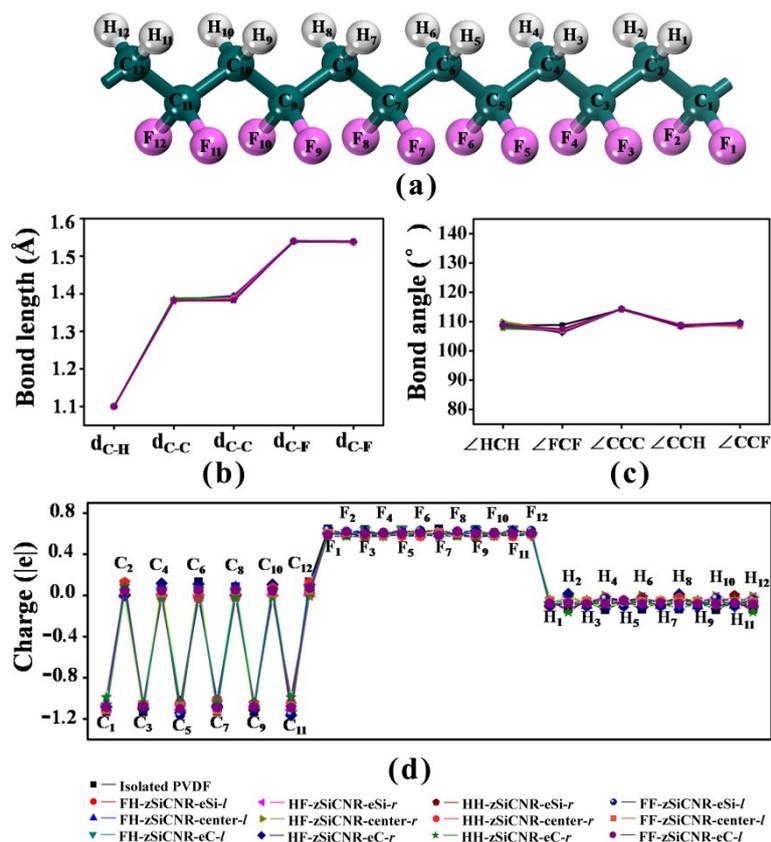


Figure S4. (a) The corresponding atomic number for the isolated PVDF polymer or ones in the most favorable jointed systems. The parallel comparison of the bond lengths (b), bond angles (c) and bader charges on the involved atoms (d) for the PVDF polymer in the isolated and jointed systems.