SUPPLEMENTARY

UNRAVELING THE ELECTRONIC PROPERTIES OF SILICON CARBIDE MONOLAYERS WITH Si/C VACANCIES

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Table S1. Bond lengths in SiCML obtained using various DFT exchange-correlation functionals

	l_{C-C}, Å			
	HSE06	PBE0	M06	PBE*
h-SiC	–	–	–	–
m-SiC	1.433	1.432	1.432	1.465
op-SiC	1.473, 1.447	1.476, 1.448	1.473, 1.447	1.478
	l_{Si-C}, Å			
	HSE06	PBE0	M06	PBE*
h-SiC	1.787	1.787	1.782	1.780
m-SiC	1.795	1.793	1.790	1.829
op-SiC	1.807	1.809	1.807	1.833
	l_{Si-Si}, Å			
	HSE06	PBE0	M06	
h-SiC	–	–	–	–
m-SiC	–	–	–	–
op-SiC	2.129	2.131	2.129	–

*[doi: 10.1038/s41598-020-68765-x]

Table S2. Direct band gap values (eV) in SiCML obtained using various DFT exchange-correlation functionals

Method	h-SiC	m-SiC	op-SiC
HSE06, this work	3.19 (K → K)	1.00 (Γ → Γ)	0.03
PBE0, this work	3.79 (K → K)	1.37 (Γ → Γ)	0.04
M06, this work	3.96 (K → K)	1.63 (Γ → Γ)	0.06
HSE06	3.43 (K → K), 3.38 (K →M) [62]	1.09 [66]	
PBE	2.56 [63] 2.14 (K → K) [4] 2.52 (K → K) [67] [68]	0.56 [63] 0.60 [66]	0.04 [63]
PW91+ GW_0	3.90 [1]		

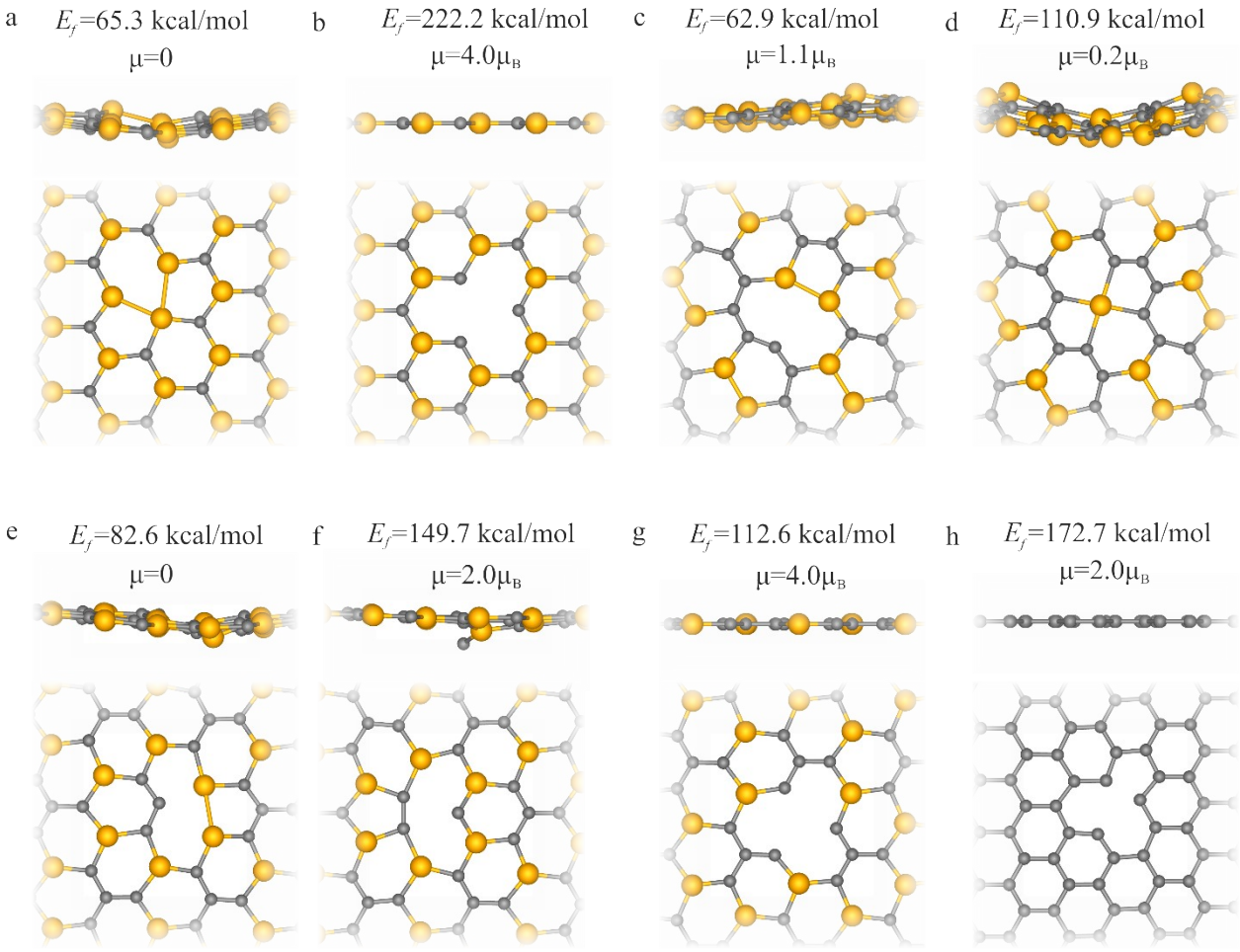


Figure S1. Atomic structure of SiCML with various vacancy defect: a) h-SiC-vac-C, b) h-SiC-vac-Si, c) op-SiC-vac-C, d) op-SiC-vac-Si, e) m-SiC-vac-C^1, f) m-SiC-vac-C^2, g) m-SiC-vac-Si, h) graphene-vac.

Table S3. Total atomic charges (Mulliken) and total atomic spins for the atoms of a selvedge in SiCML models.

Atom number of a *selvedge*	Atom number in a cell	Atomic charge, |e|	Total atomic spin, μ_B
h-SiC-vac-Si			
C1	22	-0.356	1.234
Si2	82	0.674	0.002
C3	27	-0.758	0.115
Si4	91	0.676	0.001
C5	60	-0.353	1.248
Si6	97	0.675	-0.022
C7	36	-0.769	0.094
Si8	92	0.669	-0.021
C9	59	-0.435	0.857
Si10	83	0.670	-0.021
C11	18	-0.769	0.094
Si12	78	0.674	-0.021
m-SiC-vac-Si			
C1	34	-0.2090	1.1774
C2	35	0.0604	-0.1274
C3	80	-0.5542	0.1760
Si4	98	0.6924	0.0034

C5	23	-0.2262	1.0893
C6	15	0.0557	-0.1356
C7	8	-0.5496	0.2165
Si8	91	0.7057	-0.0352
C9	14	-0.2375	0.9333
C10	13	0.0645	-0.1092
C11	22	-0.5626	0.1550
Si12	95	0.6932	-0.0498
h-SiC-vac-C			
Si1	119	0.3754	-0.00002
C2	22	-0.6866	0.00001
Si3	81	0.7651	-0.00001
C4	27	-0.6951	0.00000
Si5	90	0.3948	-0.00004
C6	35	-0.7241	-0.00001
Si7	100	0.7476	0.00000
C8	40	-0.7121	0.00000
Si9	98	0.3547	0.00006
C10	36	-0.6912	-0.00001
Si11	91	0.7626	0.00000
C12	59	-0.6938	-0.00001
m-SiC-vac-C[1]			
Si1	102	0.485	-0.0003
C2	42	-0.482	0.0001
Si3	43	0.485	-0.0003
C4	44	-0.482	0.0001
Si5	98	0.359	0.0005
C6	23	-0.511	-0.0003
Si7	95	0.763	0.0003
C8	34	-0.526	-0.0005
C9	35	-0.066	0.0006
C10	41	-0.530	-0.0004
Si11	107	0.796	0.0001
C12	47	-0.518	0.0000
m-SiC-vac-C[2]			
Si1	41	-0.4115	0.2202
C2	107	0.6951	0.0450
Si3	47	-0.5596	0.0581
C4	102	0.6692	-0.0208
Si5	35	-0.4405	0.8935
C6	98	0.6692	-0.0208
Si7	23	-0.5598	0.0582
C8	95	0.6951	0.0449
Si9	34	-0.4115	0.2203
C10	94	0.7274	0.1037
Si11	33	-0.5496	-0.0166
C12	101	0.7274	0.1037
op-SiC-vac-Si			
Si1	89	0.5726	0.0289
C2	19	-0.1698	0.0123
C3	20	-0.1846	-0.0029
C4	6	-0.0952	-0.0034

C5	5	-0.1950	0.0131
C6	71	-0.2256	-0.0075
Si7	119	0.4674	0.0228
C8	70	-0.2309	-0.0072
C9	80	-0.1696	0.0124
C10	79	-0.1848	-0.0030
C11	4	-0.0950	-0.0033
C12	10	-0.1952	0.0129
op-SiC-vac-C			
Si1	84	0.3446	0.0180
Si2	89	0.3088	0.1118
C3	9	-0.2201	-0.0249
C4	4	-0.1702	0.0550
C5	78	-0.2341	-0.0907
C6	79	0.0186	0.8658
C7	69	-0.2699	-0.0745
Si8	119	0.4705	0.0477
C9	70	-0.4502	-0.0058

Table S4. Bond lengths (Å) and positions of the extrema of the total static and electrostatic potentials and electron density, defined as segments along the interatomic bond line, as well as the values of potentials and electron density at extrema along the bond line (a.u.)

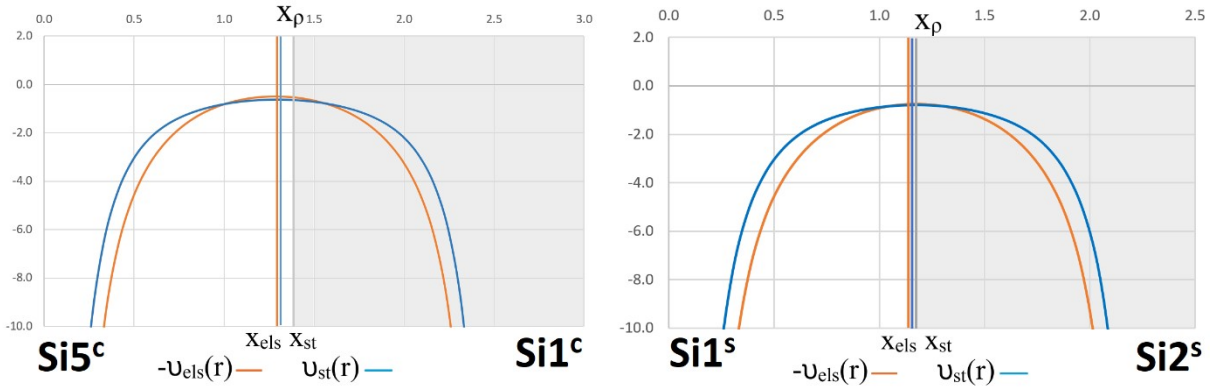
Bond	Distance	x_{els}	$\upsilon_{els}(r)$	x_{st}	$\upsilon_{st}(r)$	x_{ρ}	$\rho(r)$	$\Delta_{st\text{-}els}$	$\Delta_{\rho\text{-}st}$	$\lvert\Delta_{st\text{-}els}\text{-}\Delta_{\rho\text{-}st}\rvert$
h-SiC-vac-Si										
C60-C59 $C5^{s}…C9^{s}$	3.122	1.538	-0.123	1.567	-0.148	1.582	0.011	0.0291	0.0157	0.013
C22-C60 $C1^{s}…C5^{s}$	2.993	1.489	-0.137	1.493	-0.168	1.504	0.016	0.0038	0.0113	0.008
C22-C59 $C1^{s}…C9^{s}$	3.118	1.535	-0.124	1.567	-0.148	1.582	0.011	0.0313	0.0157	0.016
m-SiC-vac-Si										
C23-C14 $C5^{s}…C9^{s}$	3.154	1.490	-0.105	1.553	-0.166	1.585	0.013	0.0634	0.0317	0.032
C34-C23 $C1^{s}…C5^{s}$	3.084	1.534	-0.119	1.534	-0.172	1.534	0.015	0.0000	0.0000	0.000
C34-C14 $C1^{s}…C9^{s}$	3.190	1.587	-0.084	1.619	-0.167	1.635	0.011	0.0320	0.0161	0.016
h-SiC-vac-C										
C22-Si119 $C2^{s}$–$Si1^{c}$	1.863	0.824	1.004	0.852	-1.028	1.123	0.110	0.0281	0.2715	0.243
C59-Si119 $C12^{s}$–$Si1^{c}$	1.855	0.820	1.021	0.839	-1.039	1.119	0.112	0.0186	0.2797	0.261
Si90-Si119 $Si5^{s}$–$Si1^{c}$	2.595	1.291	0.486	1.291	-0.624	1.369	0.058	0.0000	0.0782	0.078
Si98-Si119 $Si9^{s}$–$Si1^{c}$	2.691	1.325	0.411	1.339	-0.566	1.434	0.049	0.0136	0.0946	0.081
m-SiC-vac-C[1]										
C35-Si102 $C9^{s}$–$Si1^{c}$	2.309	1.009	0.411	1.044	-0.619	1.311	0.065	0.0348	0.2668	0.232
C42-Si102 $C2^{s}$–$Si1^{c}$	1.908	0.844	0.904	0.863	-0.970	1.160	0.106	0.0192	0.2971	0.278
C44-Si102 $C4^{s}$–$Si1^{c}$	1.915	0.847	0.909	0.866	-0.970	1.164	0.104	0.0192	0.2984	0.279
Si98-Si102 $Si5^{s}$–$Si1^{c}$	2.514	1.251	0.566	1.251	-0.671	1.327	0.060	0.0000	0.0758	0.076

op-SiC-vac-Si										
C10-Si89 C12^{s}–Si1^{c}	2.115	0.935	0.613	0.956	-0.770	1.275	0.084	0.0212	0.3188	0.298
C19-Si89 C2^{s}–Si1^{c}	2.201	0.962	0.516	0.995	-0.698	1.305	0.074	0.0331	0.3097	0.277
C5-Si89 C5^{s}–Si1^{c}	2.116	0.936	0.611	0.956	-0.770	1.276	0.084	0.0212	0.3188	0.298
C80-Si89 C9^{s}–Si1^{c}	2.202	0.963	0.515	0.996	-0.697	1.306	0.074	0.0332	0.3098	0.277
op-SiC-vac-C										
Si84-Si89 Si1^{s}–Si2^{s}	2.347	1.168	0.750	1.168	-0.796	1.179	0.075	0.0000	0.0117	0.012
C79...Si84 C6^{s}–Si1^{s}	3.345	1.412	0.105	1.513	-0.273	1.630	0.011	0.1008	0.1177	0.017
C70-Si84 C9^{s}–Si1^{s}	1.861	0.833	1.041	0.851	-1.051	1.122	0.111	0.0187	0.2712	0.253
C5-Si84 C–Si1^{s}	1.792	0.801	1.133	0.819	-1.121	1.071	0.124	0.0180	0.2521	0.234
C9-Si89 C3^{s}–Si2^{s}	1.929	0.863	0.909	0.882	-0.969	1.173	0.105	0.0194	0.2908	0.271
m-SiC-vac-C^{2}										
C35-Si102 C4^{s}–Si5^{c}	1.749	0.782	1.245	0.800	-1.188	1.046	0.132	0.0176	0.2460	0.228
C34-Si95 C8^{s}–Si9^{c}	1.961	0.867	0.896	0.887	-0.947	1.133	0.098	0.0197	0.2463	0.227

Table S5. Bond lengths (Å) and electron density, Laplacian of electron density, kinetic and potential energy densities (a.u.) and their ratio at the bond critical points

Bond	Distance	Lap	g(r)	v(r)	v(r)/g(r)	$\rho(r_{bcp})$
h-SiC-vac-Si						
C60-C59 C5^{s}…C9^{s}	3.122	0.0225	0.0054	-0.0052	1.0	0.0114
C22-C60 C1^{s}…C5^{s}	2.993	0.0239	0.0070	-0.0081	1.2	0.0164
C22-C59 C1^{s}…C9^{s}	3.118	0.0232	0.0056	-0.0053	0.9	0.0115
C13-Si9	1.781	0.5885	0.1893	-0.2314	1.2	0.1262
m-SiC-vac-Si						
C23-C14 C5^{s}…C9^{s}	3.154	0.0228	0.0058	-0.0059	1.0	0.0127
C34-C23 C1^{s}…C5^{s}	3.084	0.0211	0.0062	-0.0072	1.2	0.0152
C34-C14 C1^{s}…C9^{s}	3.190	0.0212	0.0051	-0.0049	1.0	0.0110
C9-C17	1.432	-0.7505	0.2285	-0.6447	2.8	0.2846
C9-Si87	1.792	0.6924	0.2078	-0.2425	1.2	0.1272
h-SiC-vac-C						
C22-Si119 C2^{s}–Si1^{c}	1.863	0.2766	0.1187	-0.1682	1.4	0.1100
C59-Si119 C12^{s}–Si1^{c}	1.855	-0.2167	0.0384	-0.1311	3.4	0.1118
Si90-Si119 Si5^{s}–Si1^{c}	2.595	-0.0523	0.0171	-0.0472	2.8	0.0590
Si96-Si119 Si9^{s}–Si1^{c}	2.691	-0.0347	0.0142	-0.0371	2.6	0.0508
C22-Si77 C2^{s}–Si	1.807	-0.0157	0.0824	-0.1688	2.0	0.1211
C7-Si61	1.784	0.5572	0.1832	-0.2273	1.2	0.1255
m-SiC-vac-C^{1}						
C35-Si102 C9^{s}–Si1^{c}	2.309	-0.0674	0.0194	-0.0557	2.9	0.0656
C42-Si102 C2^{s}–Si1^{c}	1.908	0.3424	0.1255	-0.1653	1.3	0.1062
C47-Si102 C4^{s}–Si1^{c}	1.915	0.3027	0.1172	-0.1588	1.4	0.1046

Si98-Si102 $Si5^s$–$Si1^c$	2.514	-0.0538	0.0187	-0.0514	2.7	0.0620
op-SiC-vac-Si						
C10-Si89 $C12^s$–$Si1^c$	2.115	0.0176	0.0497	-0.0950	1.9	0.0846
C19-Si89 $C2^s$–$Si1^c$	2.201	-0.0487	0.0302	-0.0727	2.4	0.0751
C5-Si89 $C5^s$–$Si1^c$	2.116	-0.0737	0.0343	-0.0872	2.5	0.0844
C80-Si89 $C9^s$–$Si1^c$	2.202	-0.0547	0.0292	-0.0720	2.5	0.0750
C18-C94	1.797	0.2355	0.1332	-0.2076	1.6	0.1285
C4-C79	1.440	-0.7592	0.2258	-0.6414	2.8	0.2840
C1-C73	1.457	-0.6736	0.2106	-0.5895	2.8	0.2695
op-SiC-vac-C						
Si84-Si89 $Si1^s$–$Si2^s$	2.347	-0.0905	0.0249	-0.0725	2.9	0.0770
C79-Si84 $C6^s$–$Si1^s$	3.345	0.0187	0.0050	-0.0053	1.1	0.0123
C70-Si84 $C9^s$–$Si1^s$	1.861	0.3480	0.1319	-0.1767	1.3	0.1112
C9-Si89 $C3^s$–$Si2^s$	1.929	0.2732	0.1130	-0.1576	1.4	0.1053
C70-Si119 $C9^s$–$Si8^s$	1.770	0.5041	0.1810	-0.2360	1.3	0.1309
C9-C4 $C3^s$–$C4^s$	1.426	-0.7330	0.2350	-0.6526	2.8	0.2862
C4-C78 $C4^s$–$C5^s$	1.452	-0.6955	0.2125	-0.5989	2.8	0.2723
C78-C79 $C5^s$–$C6^s$	1.364	-0.8147	0.2794	-0.7624	2.7	0.3134
C79-C69 $C6^s$–$C7^s$	1.385	-0.8034	0.2598	-0.7205	2.8	0.3036
m-SiC-vac-C^2						
C35-Si102 $C4^s$–$Si5^c$	1.749	0.2961	0.1483	-0.2225	1.5	0.1325
C34-Si95 $C8^s$–$Si9^c$	1.961	0.0880	0.0738	-0.1255	1.7	0.0973
C34-C41 $C8^s$–$C9^c$	1.496	-0.4846	0.1801	-0.4814	2.7	0.2371



a b

Figure S2. Superposition of potentials along the bond lines in a) h-SiC-vac-C monolayer, $Si5^s$…$Si1^s$ (2.595 Å); b) op-SiC-vac-C monolayer, $Si1^s$…$Si2^s$ (2.347 Å).

The expression for the phase space defined Fisher information density, PS-FID

Moreover, due to indistinguishability of electrons, the latter can be reduced within the framework of DFT to the non-negative distribution function $f(\mathbf{r},\mathbf{p})$ normalized to the number of electrons N and depended on a position and a momentum of only one electron. The non-interacting electronic position-space properties are extracted from $f(\mathbf{r},\mathbf{p})$ by integration over the dynamical variable p.

Neither electron density $\rho(r) = \int dpf(r,p),\ nor$ $f(\mathbf{r},\mathbf{p})$ itself do not exhibit electron pairs explicitly. However, minimizing the Fisher information for distribution function $f(\mathbf{r},\mathbf{p})$, we arrive at the phase-space-defined Fisher information density (PS-FID) [doi:10.1016/j.chemphys.2014.03.006]:

$$i_f(\mathbf{r}) = \int d\mathbf{p} \frac{\nabla_{\mathrm{p}} f(\mathbf{r},\mathbf{p}) \cdot \nabla_{\mathrm{p}} f(\mathbf{r},\mathbf{p})}{f(\mathbf{r},\mathbf{p})} \qquad 1.1.$$

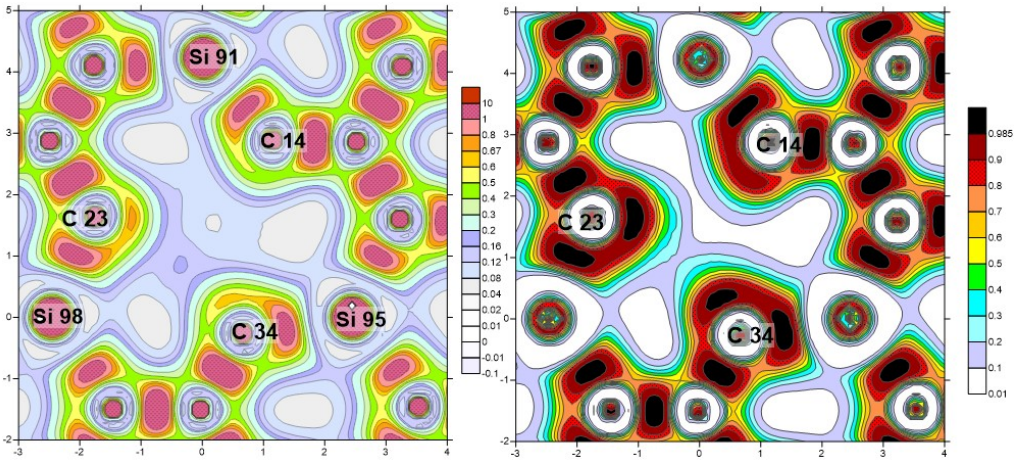
This one-electron function varied with the position r, contains information about spatial structuredness of electronic momentum distribution in the position space and leads to distribution function in the Maxwell-Boltzmann form

$$f(r,p) = \rho(r)[2\pi kT(r)]^{\frac{-3}{2}} \exp\{-\frac{1}{2kT(r)}p^2\} \qquad 1.2,$$

here k is the Boltzmann constant. $T(r)$ is the local information temperature of electron gas, which may be associated with a measure of local momentum fluctuations (momentum variance) at point r, $t_{s+}(r) = t_{s+}(r) - \frac{1}{8}\nabla^2\rho(r),$ in agreement with the restrictions of the uncertainty principle. Thus, the expression for the phase-space-defined Fisher information density, PS-FID, shows the electron pairs explicitly

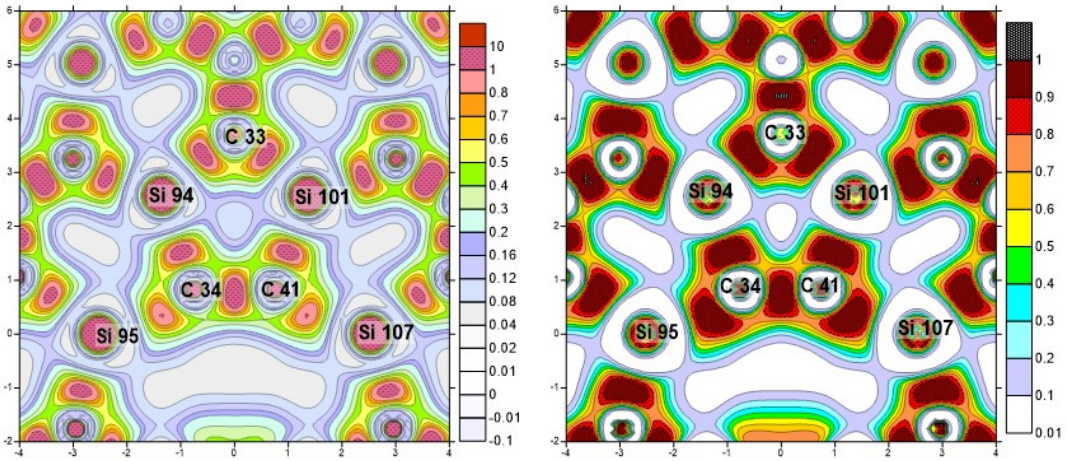
$$PS-FID(r) = \frac{9}{2}\frac{\rho(r)^2}{2t_{s+}(r) - \frac{1}{4}\nabla^2\rho(r)} \qquad 1.3.$$

Here $t_{s+}(r) = \frac{1}{2}\sum_{l}^{occ} c_l \nabla\varphi^*_l(r) \cdot \nabla\varphi_l(r)$ is the positively-defined kinetic energy density of non-interacting electrons and φ_i are the Konh-Sham orbitals.



a b

Figure S3. a) PS-FID and b) ELF distribution in the planes of m-SiC-vac-Si monolayer; kinetic energy approximation: AST kinetic energy density approximation [doi:10.1002/qua.24957] has been used



a b

Figure S4. a) PS-FID and b) ELF in the planes of m-SiC-vac-C2 monolayer; kinetic energy approximation: AST kinetic energy density approximation [doi:10.1002/qua.24957] has been used.

Table S6. The band gaps for the defected and the pristine (defect-less) structures of SiCML

Model	Band gap (eV)	
	Spin UP	Spin DOWN
h-SiC	3.82 (direct)	
h-SiC-vac-C	3.14 (indirect)	
h-SiC-vac-Si	3.87 (indirect)	0.76 (indirect)
m-SiC	1.37 (direct)	
m-SiC-vac-C1	1.38 (direct)	
m-SiC-vac-C2	1.49 (direct)	0.85 (direct)
m-SiC-vac-Si	1.62 (direct)	0.47 (indirect)
op-SiC	0	
op-SiC-vac-C	0	
op-SiC-vac-Si	0	

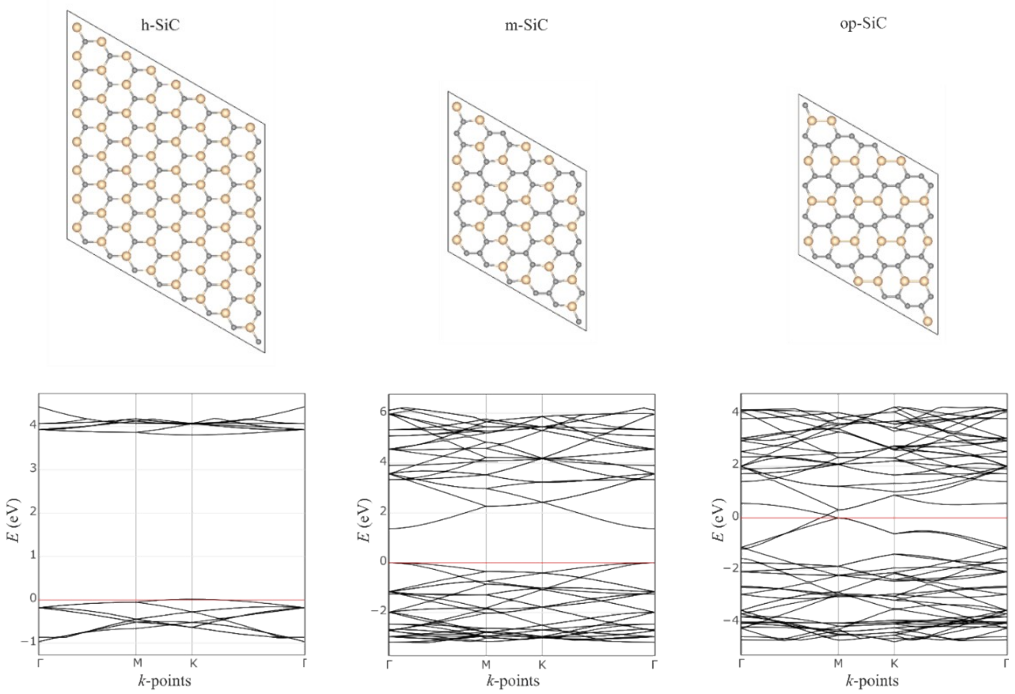


Figure S5. Band structures for defect-less SiCML models. PBE0 exchange-correlation functional in CRYSTAL program was used.

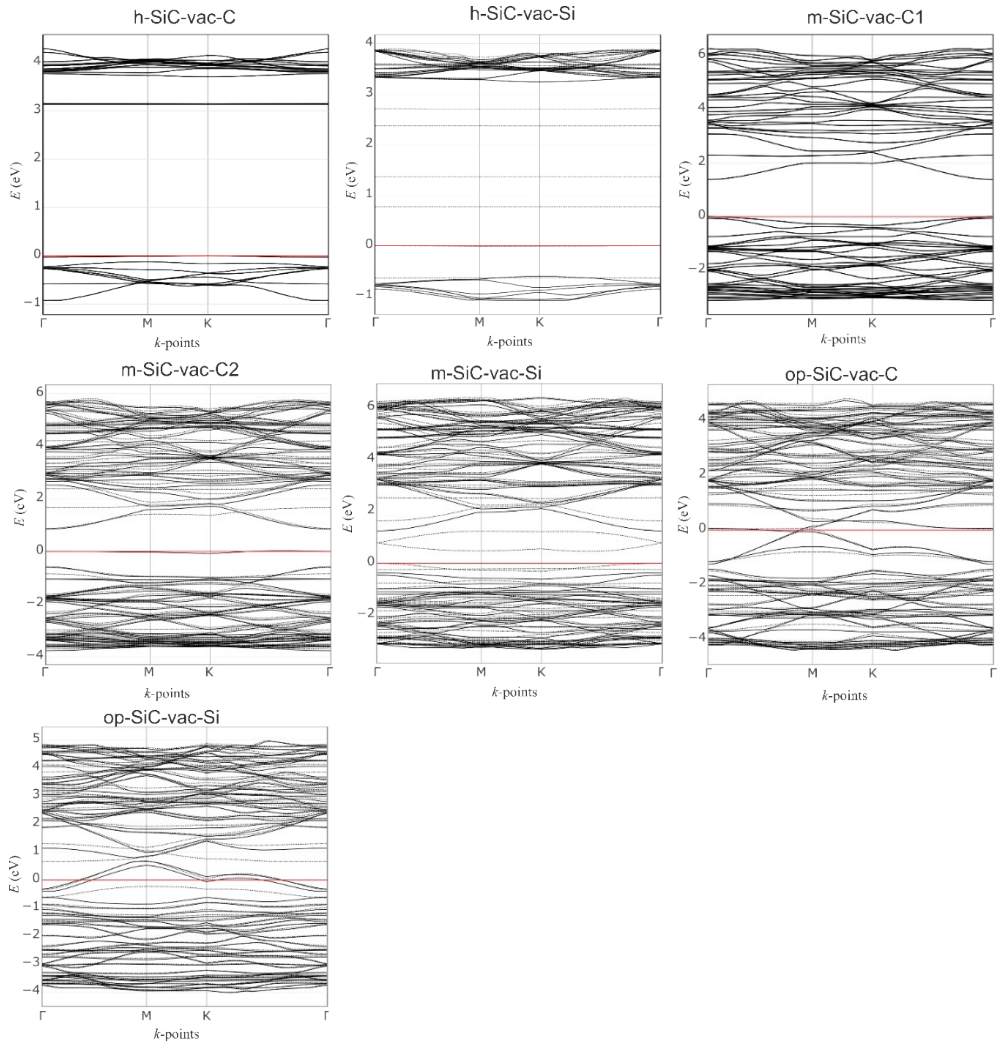


Figure S6. Band structures for the structural models of defective SiCML; spin-polarized calculations. PBE0 exchange-correlation functional in CRYSTAL program was used.

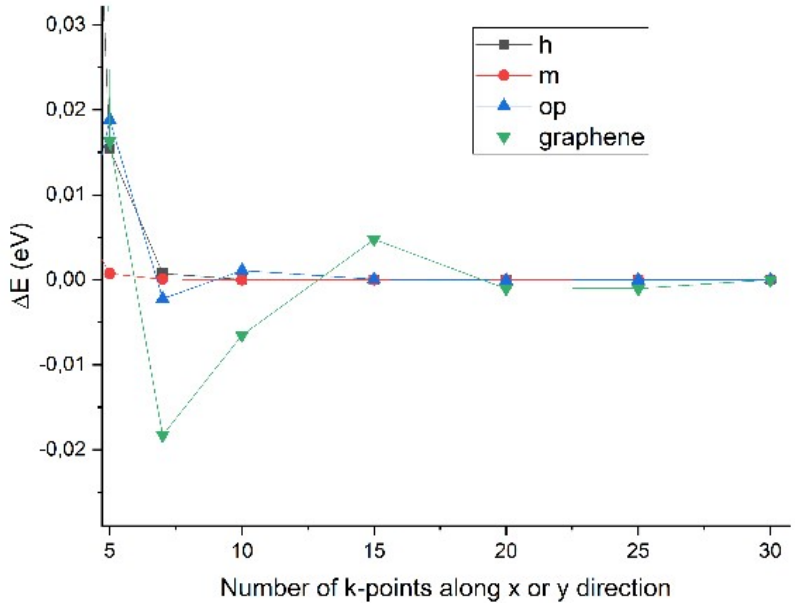


Figure S7. The dependence of ΔE on the number of k-points along Ox and Oy directions.

Electronic chemical potential estimation

$e_{Graphene} = E_{Graphene}/n_{Graphene}$ – the energy of one C atom in graphene.

$e_{Silicene} = E_{Silicene}/n_{Silicene}$ – the energy of one Si atom in silicene.

For defect-free h-SiC monolayer [57]:

$$\mu_C = E_{SiC} - \frac{E_{Silicene}}{n_{Silicene}},$$

$$\mu_{Si} = E_{SiC} - \frac{E_{Graphene}}{n_{Graphene}},$$

For defect-free m-SiC and op-SiC monolayers [57]:

$$\mu_C = \frac{E_{SiC}/6 - E_{Silicene}/n_{Silicene}}{2},$$

$$\mu_C = \frac{E_{SiC}}{6} - 2\frac{E_{Graphene}}{n_{Graphene}}.$$

Table S7. Electronic chemical potential for removed atoms estimated by various approaches, eV

Model	Atom in vacuum		Pristine MLs: graphene or silicene		Si,C MLs [57]	
	μ_C	μ_{Si}	μ_C	μ_{Si}	μ_C	μ_{Si}
h-SiC	-154.1	-123.9	-162.2	-128.0	-162.0	-127.9
m-SiC					-161.8	-127.2

op-SiC					-161.4	-126.5