Supplementary Information

Modulating the bandgap of Cr-intercalated bilayer graphene via combining the 18-electron rule and the 2D superatomic-molecule theory

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[#] These authors contributed equally to this work.

Contents:

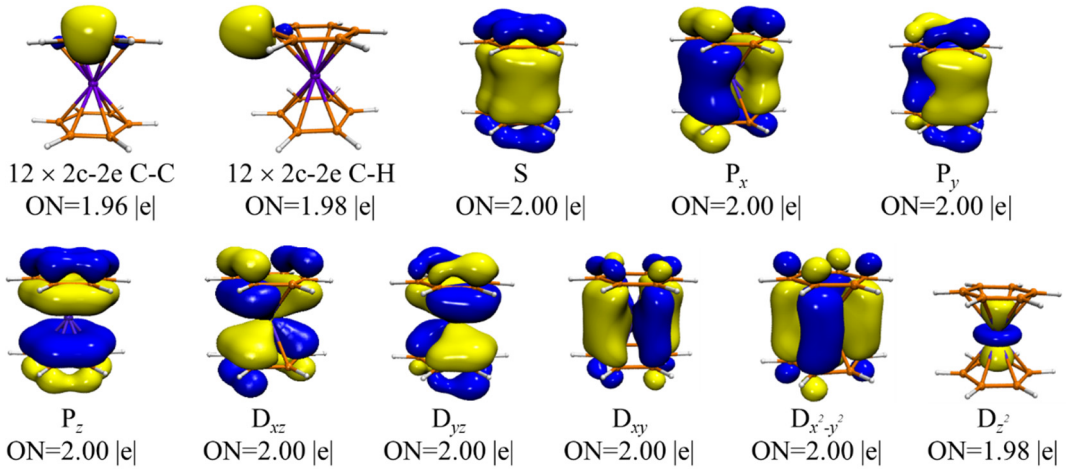

Fig. S1 Chemical bonding picture of $Cr(C_6H_6)_2$

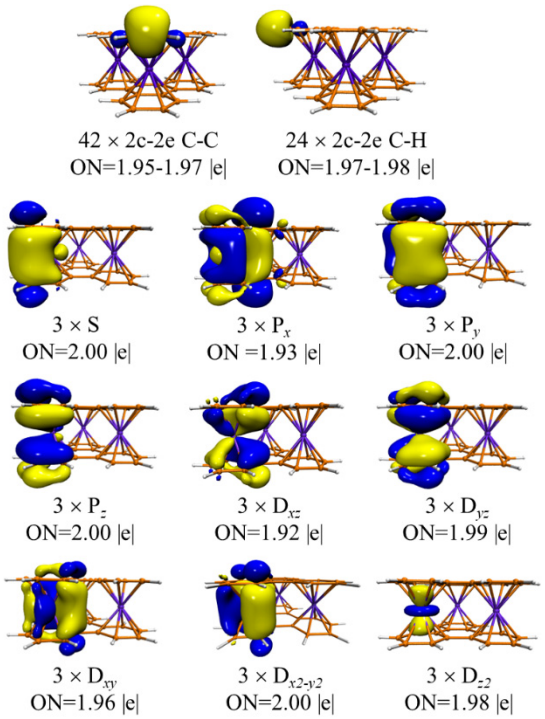

Fig. S2 Chemical bonding picture of $Cr_3C_{36}H_{24}$.

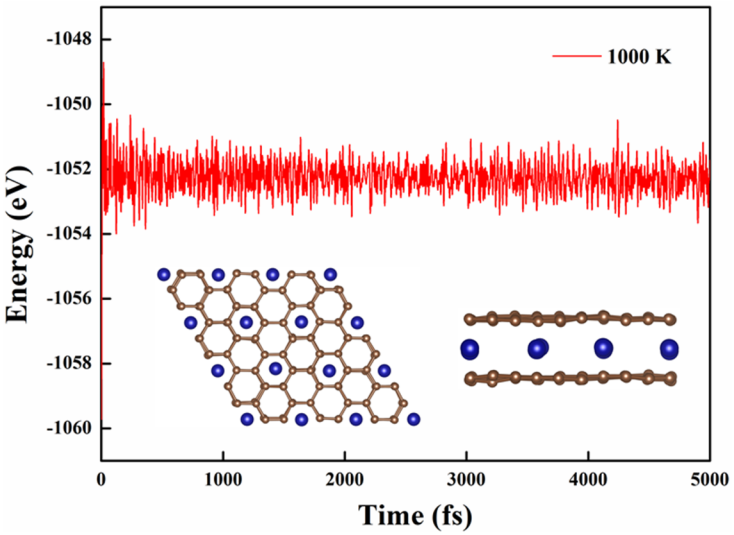

Fig. S3 Energy fluctuation depending on simulated time in molecular dynamics simulation of $C_{12}Cr$ monolayer at 1000 K after 5 ps simulation.

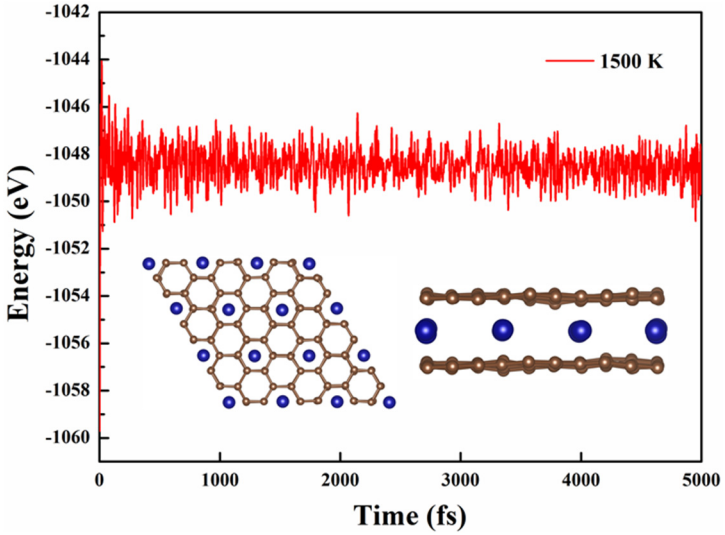

Fig. S4 Energy fluctuation depending on simulated time in molecular dynamics simulation of $C_{12}Cr$ monolayer at 1500 K after 5 ps simulation.

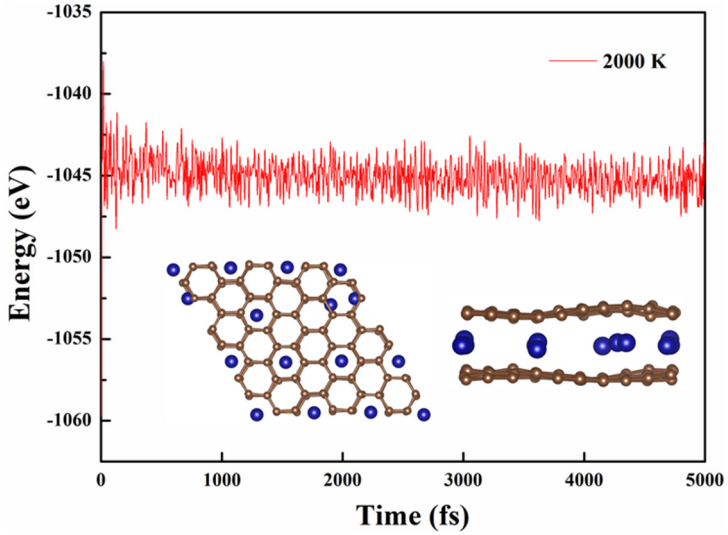

Fig. S5 Energy fluctuation depending on simulated time in molecular dynamics simulation of $C_{12}Cr$ monolayer at 2000 K after 5 ps simulation.

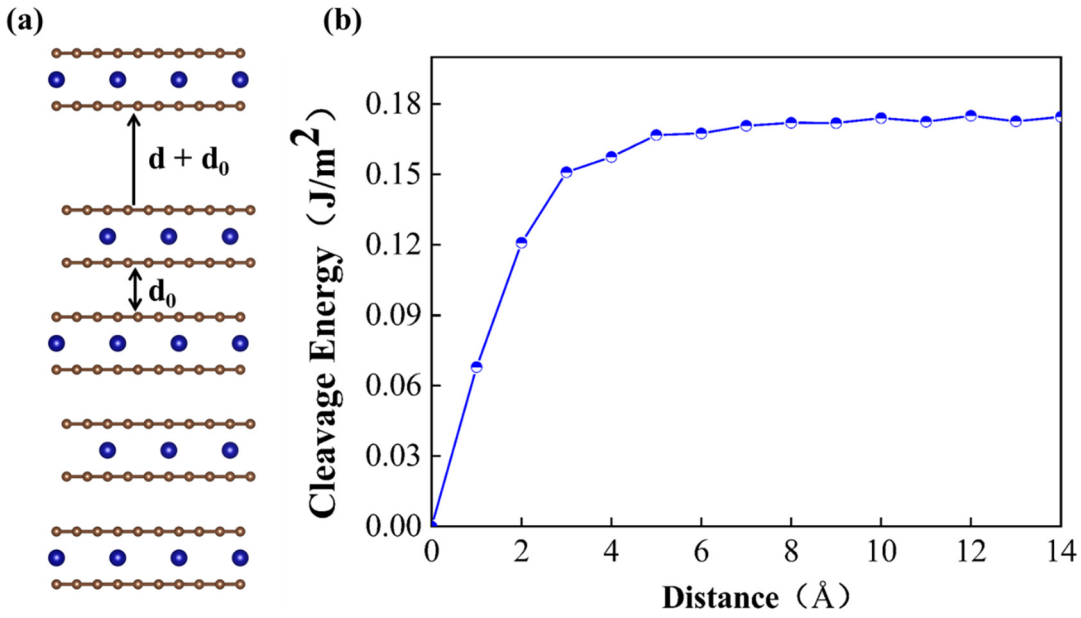

Fig. S6 (a) Separating one monolayer from neighboring four layers. (b) Cleavage energy as a function of the separation distance for a fracture in $C_{12}Cr$ bulk.

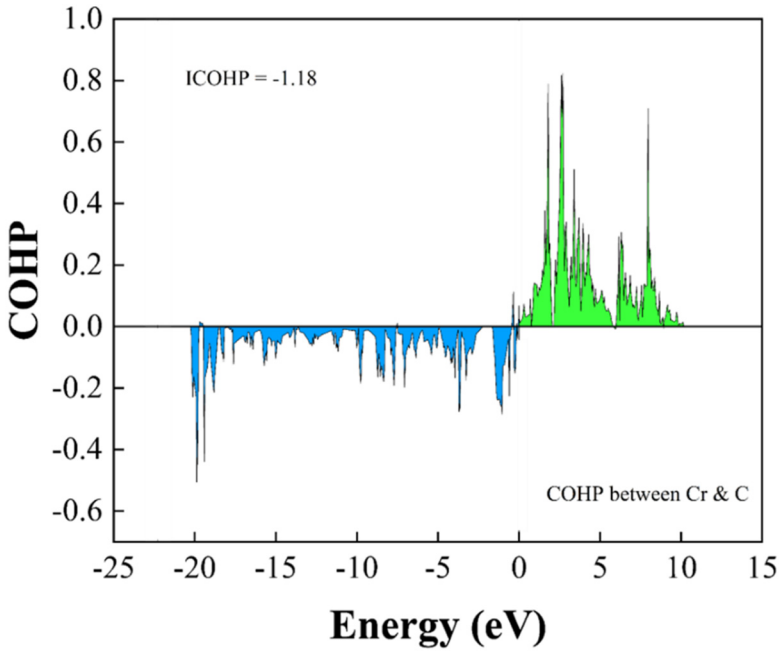

Fig. S7 Crystal orbital Hamilton population (COHP) between C atoms and Cr atoms of $C_{12}Cr$ monolayer. The Fermi level in COHP figures is set to 0 eV.

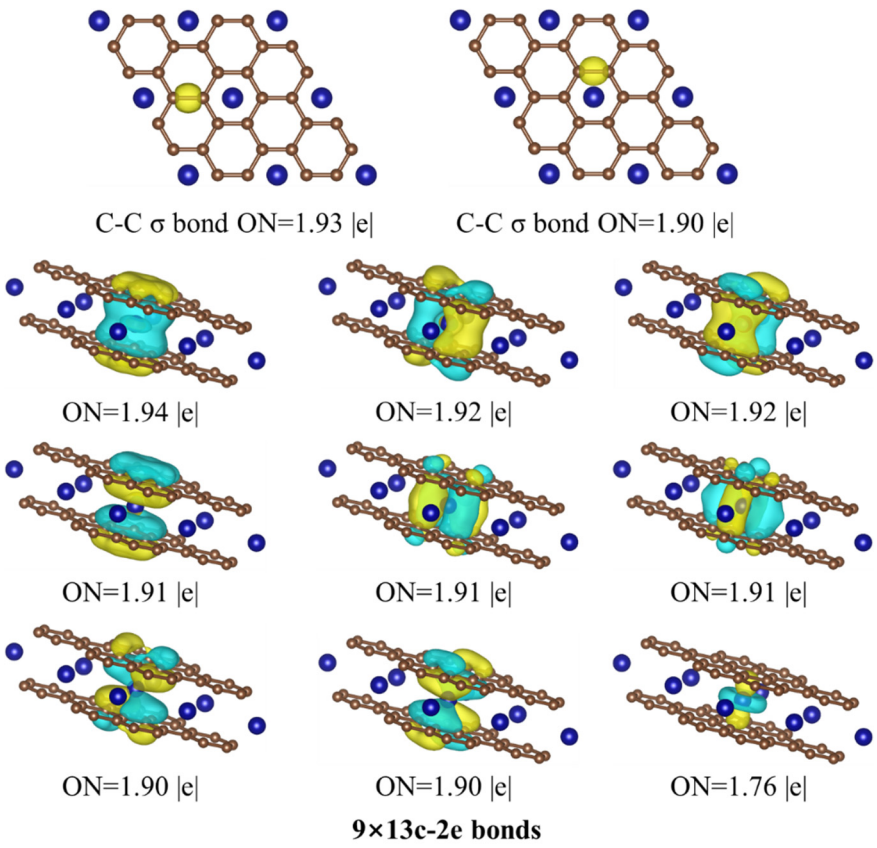

Fig. S8 Chemical bonding picture of $C_{12}Cr$ monolayer.

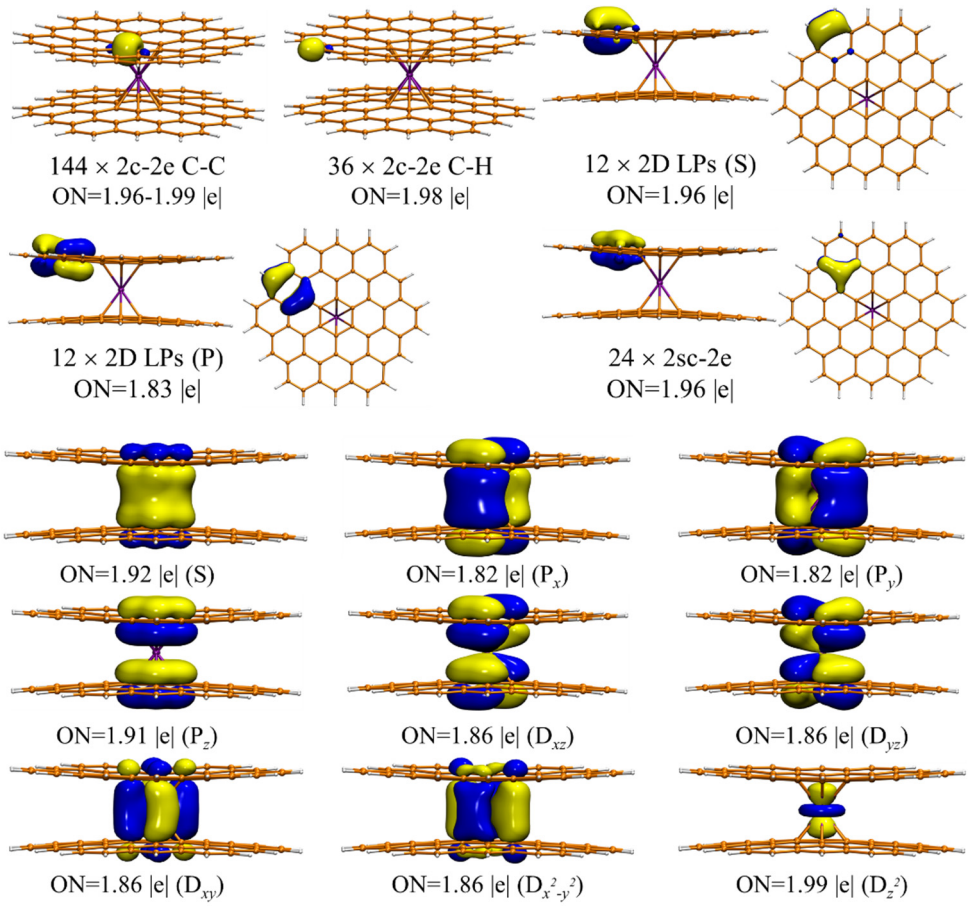

Fig. S9 Chemical bonding picture of $Cr(C_{54}H_{18})_2$.

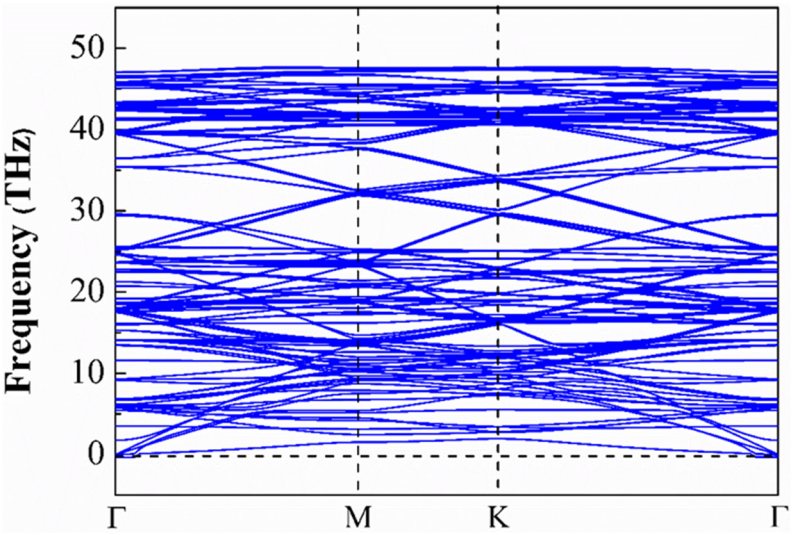

Fig. S10 Phonon dispersion of $C_{48}Cr$ monolayer.

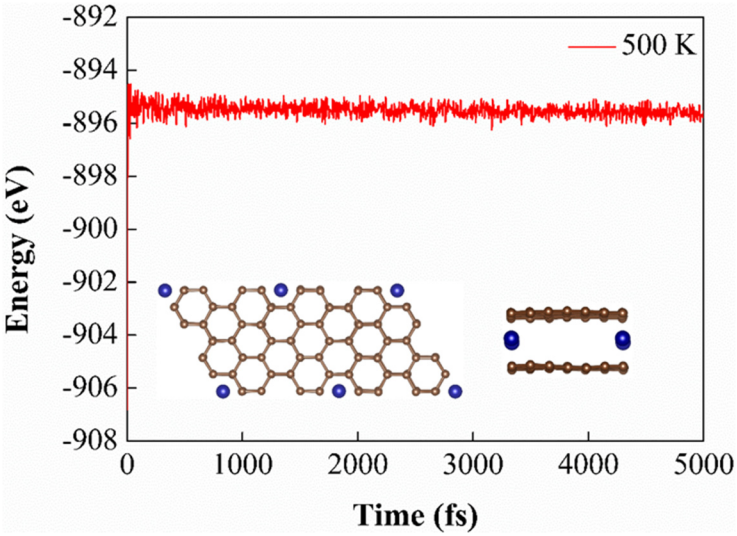

Fig. S11 Energy fluctuation depending on simulated time in molecular dynamics simulation of $C_{48}Cr$ monolayer at 500 K after 5 ps simulation.

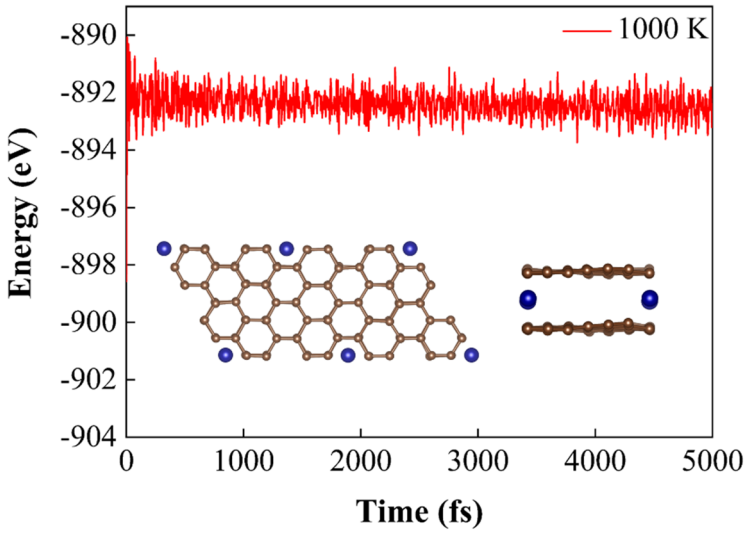

Fig. S12 Energy fluctuation depending on simulated time in molecular dynamics simulation of $C_{48}Cr$ monolayer at 1000 K after 5 ps simulation.

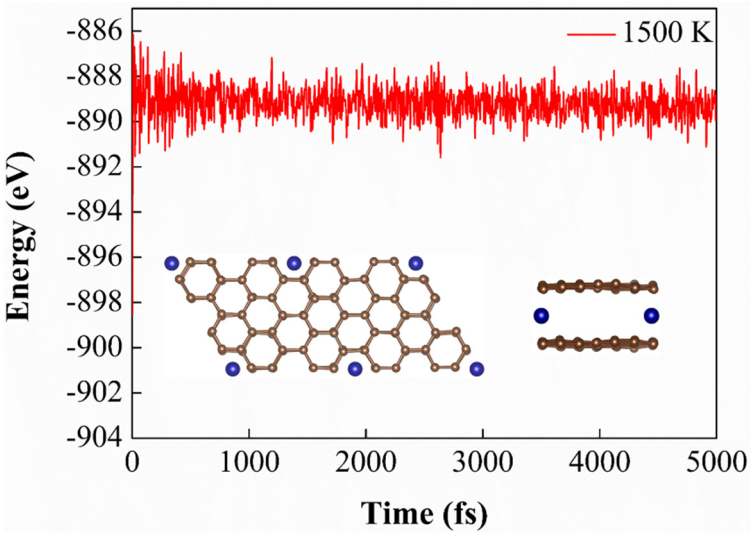

Fig. S13 Energy fluctuation depending on simulated time in molecular dynamics simulation of $C_{48}Cr$ monolayer at 1500 K after 5 ps simulation.

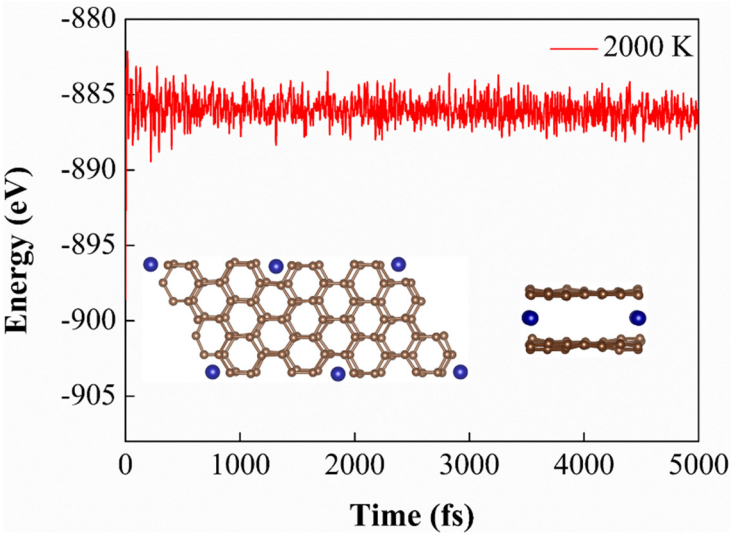

Fig. S14 Energy fluctuation depending on simulated time in molecular dynamics simulation of $C_{48}Cr$ monolayer at 2000 K after 5 ps simulation.

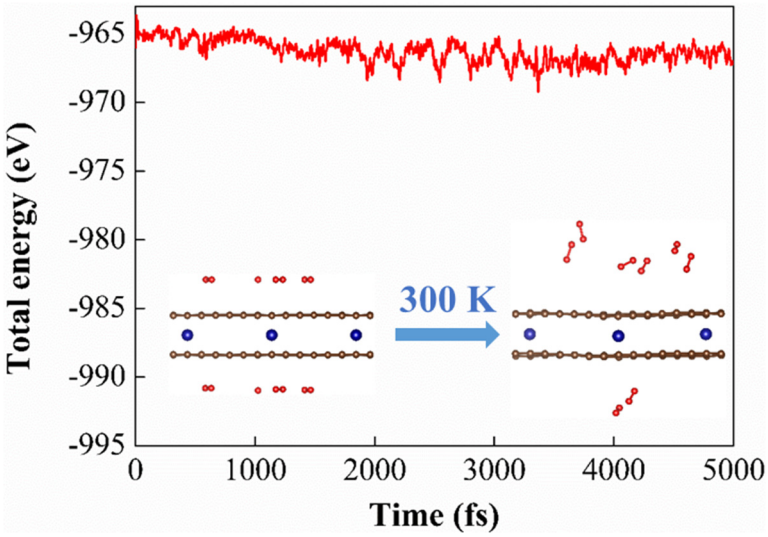

Fig. S15 Total energy and a snapshot of $C_{48}Cr$ monolayer with 8 O_2 molecules after a 5 ps AIMD simulation at 300 K.

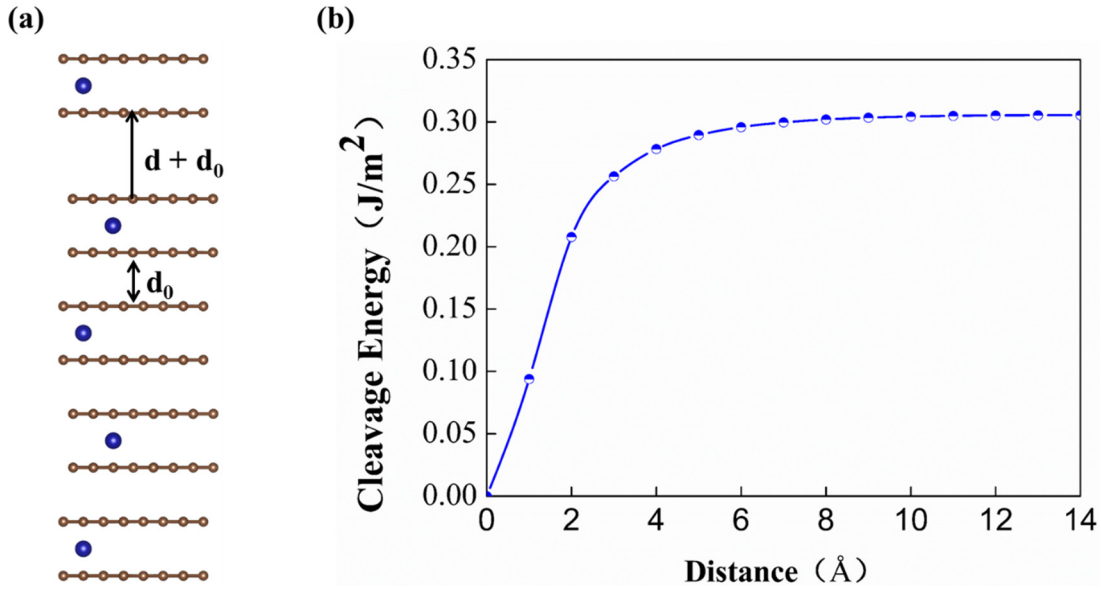

Fig. S16 (a) Separating one monolayer from neighboring four layers. (b) Cleavage energy as a function of the separation distance for a fracture in $C_{48}Cr$ bulk.

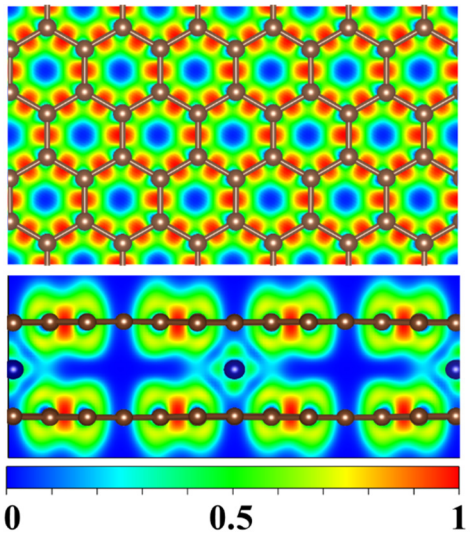

Fig. S17 ELF contour planes in the graphene layer (top) and in the Cr and C plane (below).

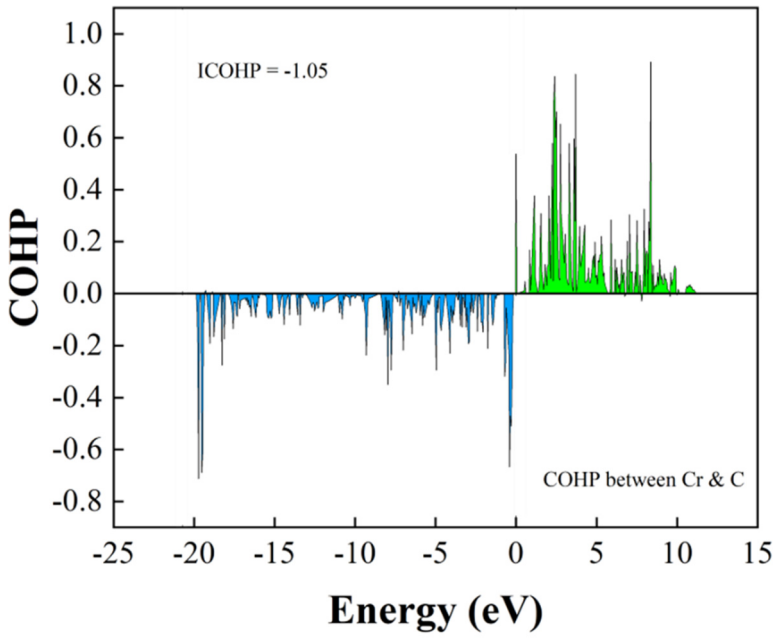

Fig. S18 Crystal orbital Hamilton population (COHP) between C atoms and Cr atoms of $C_{48}Cr$ monolayer. The Fermi level in COHP figures is set to 0 eV.

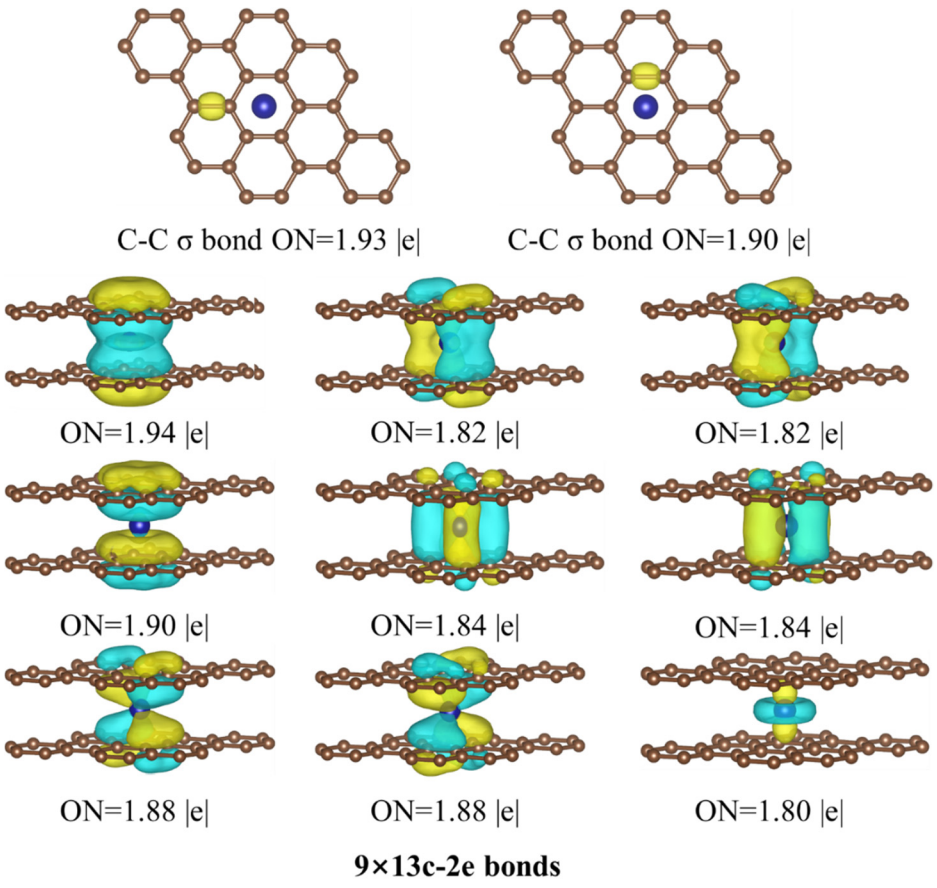

Fig. S19 SSAdNDP analysis showing localized C-C 2c-2e and 13c-2e C_6CrC_6 orbitals of $C_{48}Cr$ monolayer.

Table S1 The elastic constants (N/m), Young's modulus (N/m), and Poisson's ratio of the $C_{48}Cr$ monolayer.

	C_{11}	C_{12}	C_{66}	Y_{2D}	ν
$C_{48}Cr$	704.58	128.17	288.21	681.27	0.182

Structural information of $C_{12}Cr$ monolayer

Lattice parameters (Å, °)		Wyckoff position, fractional coordinates		
		x	*y*	*z*
$a = b = 4.33$	C	0.00000	0.33114	0.41863
$c = 20$	C	0.00000	0.66885	0.58136
$\alpha = \beta = 90$	C	0.66885	0.66885	0.41863
$\gamma = 120$	C	0.33114	0.33114	0.58136
	C	0.33114	0.00000	0.41863
	C	0.66885	0.00000	0.58136
	C	0.00000	0.66885	0.41863
	C	0.00000	0.33114	0.58136
	C	0.33114	0.33114	0.41863
	C	0.66885	0.66885	0.58136
	C	0.66885	0.00000	0.41863
	C	0.33114	0.00000	0.58136
	Cr	0.00000	0.00000	0.50000

CONTCAR

$C_{12}Cr$

1.00000000000000

4.3358396679504736 0.0000000000000000 0.0000000000000000

-2.1679198339752368 3.7549472992437405 0.0000000000000000

0.0000000000000000 0.0000000000000000 0.0000000000000000

C Cr

12 1

Direct

-0.0000000000000017 0.3311484600076313 0.4186327284096203

0.0000000000000017 0.6688515399923686 0.5813673015903822

0.6688515399923668 0.6688515399923686 0.4186327284096203

0.3311484600076331 0.3311484600076314 0.5813673015903822

0.3311484600076348 0.0000000000000000 0.4186327284096203

0.6688515399923650 0.0000000000000000 0.5813673015903822

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-0.0000000000000000	0.0000000000000000	0.5000000000000000

Structural information of $C_{48}Cr$ monolayer

Lattice parameters (Å, °)		Wyckoff position, fractional coordinates		
		x	*y*	*z*
$a = b = 8.56$	C	0.50220	0.66689	0.10924
$c = 20$	C	0.66713	0.66568	0.11084
$\alpha = \beta = 90$	C	0.83495	0.83350	0.11314
$\gamma = 120$	C	0.00226	0.66568	0.11082
	C	0.16841	0.66690	0.10920
	C	0.33617	0.83466	0.10921
	C	0.50219	0.83464	0.10924
	C	0.66713	0.00080	0.11085
	C	0.83495	0.00079	0.11315
	C	0.50203	0.66680	0.34403
	C	0.66696	0.66559	0.34248
	C	0.83480	0.83343	0.34034
	C	0.00209	0.66561	0.34251
	C	0.16823	0.66681	0.34405
	C	0.33600	0.83455	0.34403
	C	0.50203	0.83455	0.34403
	C	0.66697	0.00072	0.34249
	C	0.83479	0.00070	0.34035
	C	0.00227	0.83351	0.11312
	C	0.16955	0.00080	0.11310
	C	0.33740	0.00082	0.11079
	C	0.50232	0.16696	0.10920
	C	0.66835	0.16695	0.10923
	C	0.83611	0.33471	0.10925
	C	0.00225	0.16809	0.11313
	C	0.16954	0.16810	0.11312
	C	0.33739	0.33593	0.11081
	C	0.50230	0.33470	0.10920
	C	0.66835	0.50074	0.10923
	C	0.83610	0.50075	0.10925

	C	0.00226	0.33593	0.11083
	C	0.16840	0.50086	0.10921
	C	0.33616	0.50087	0.10922
	C	0.00211	0.83344	0.34036
	C	0.16940	0.00073	0.34035
	C	0.33722	0.00071	0.34249
	C	0.50214	0.16686	0.34402
	C	0.66817	0.16686	0.34403
	C	0.83593	0.33463	0.34405
	C	0.00208	0.16801	0.34034
	C	0.16938	0.16802	0.34033
	C	0.33722	0.33584	0.34249
	C	0.50214	0.33462	0.34403
	C	0.66818	0.50064	0.34404
	C	0.83593	0.50065	0.34405
	C	0.00209	0.33584	0.34250
	C	0.16823	0.50077	0.34406
	C	0.33600	0.50078	0.34404
	Cr	0.00224	0.00079	0.22673

CONTCAR

$C_{48}Cr$

1.00000000000000

8.5674573337053097 0.0000000000000000 0.0000000000000000

-4.2838079128217972 7.4197174297130859 0.0000000000000000

0.0000000000000000 0.0000000000000000 15.0000000000000000

C Cr

48 1

Direct

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