Electronic Supplementary Information for

Periodicity of Band Gaps of Chiral α-Graphyne Nanotubes

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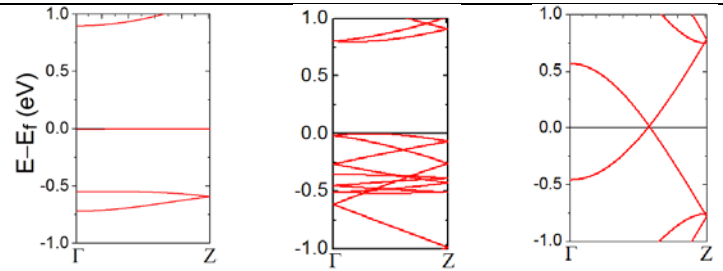
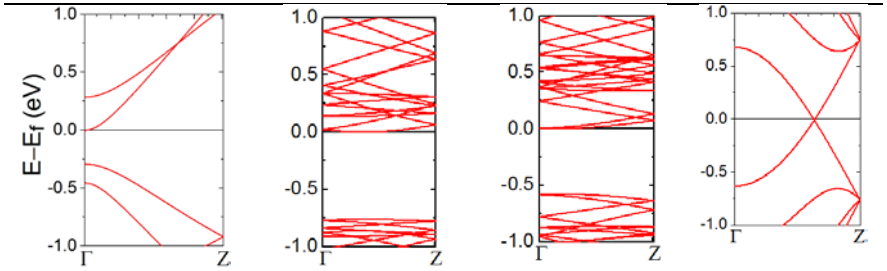
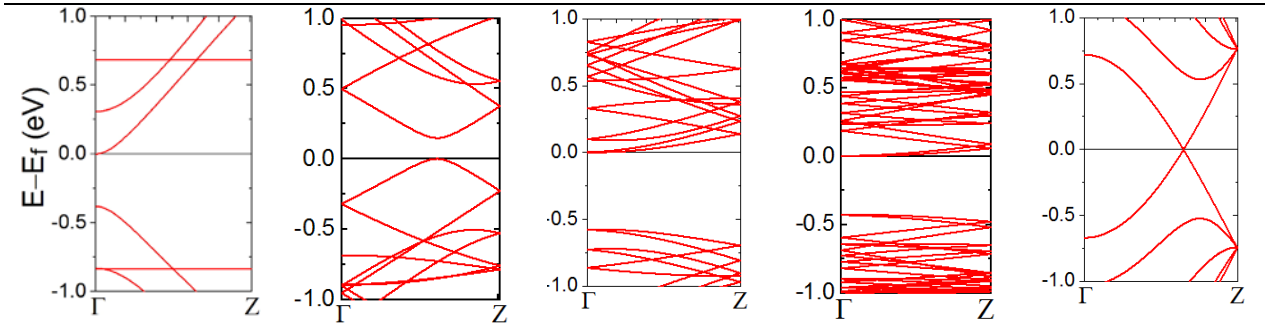
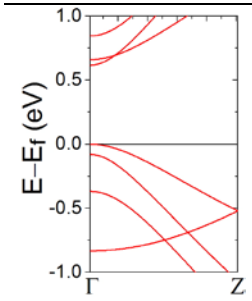
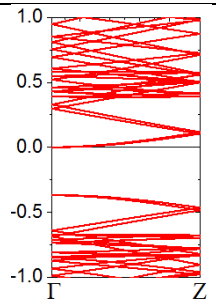
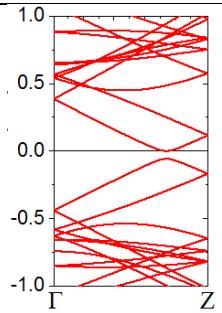
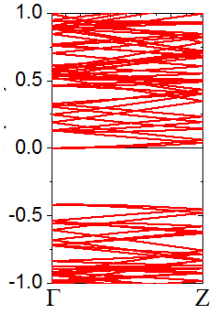
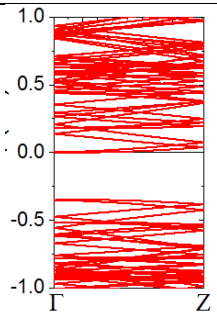
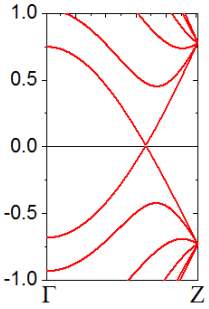
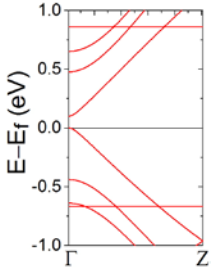
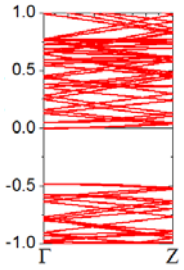
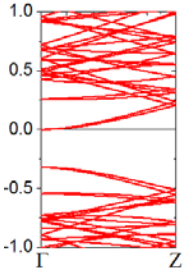
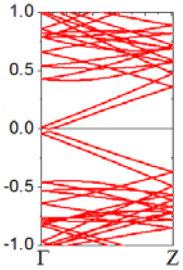
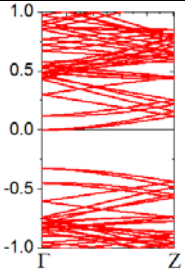
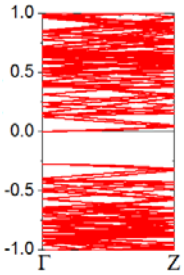
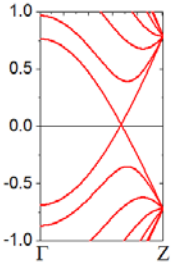
1. Band structures of αGNTs calculated by DFTB method.

n,*m*	2,0	2,1	2,2
			
E_g (eV)	0.898	0.798	0.014

n,*m*	3,0	3,1	3,2	3,3
				
E_g (eV)	0.293	0.763	0.578	0.003

n,*m*	4,0	4,1	4,2	4,3	4,4
					
E_g (eV)	0.380	0.149	0.576	0.429	0.005

n,*m*	5,0	5,1	5,2	5,3	5,4	5,5
						
E_g (eV)	0.618	0.369	0.058	0.419	0.349	0.019

n,*m*	6,0	6,1	6,2	6,3	6,4	6,5	6,6
							
E_g (eV)	0.101	0.481	0.321	0.029	0.327	0.279	0.012

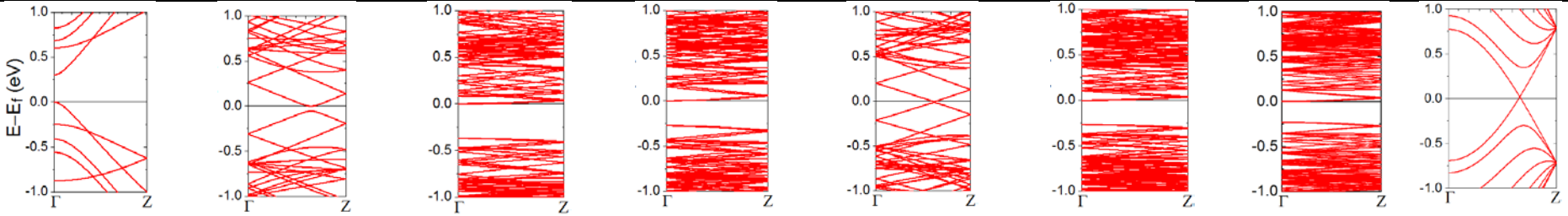
n,*m*	7,0	7,1	7,2	7,3	7,4	7,5	7,6	7,7
								
E_g (eV)	0.302	0.056	0.371	0.276	0.017	0.269	0.232	0.018

Figure S1. Band structures of Z-, A-, and C-αGNTs calculated at the DFTB/mio-1-1 level.

2. Comparison of band structures of αGNTs calculated by DFTB and DFT methods.

2.1. Comparison of band structures of Z- and A-αGNTs calculated by DFTB, VASP-PBE, VASP-HSE06, and PBE-D of Ref. 19.

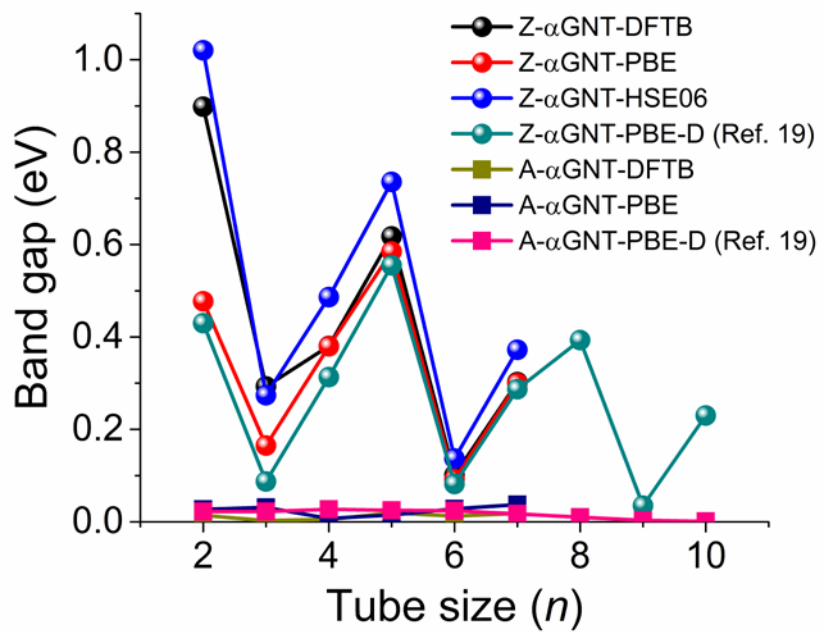
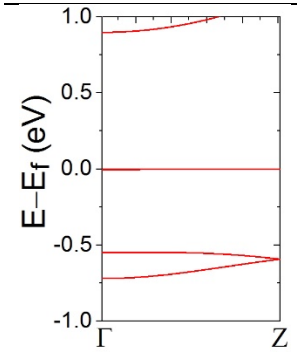
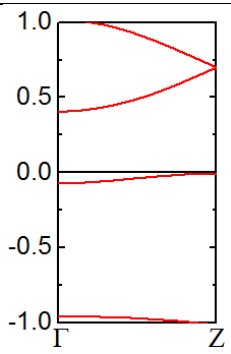
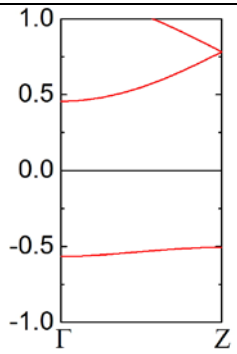
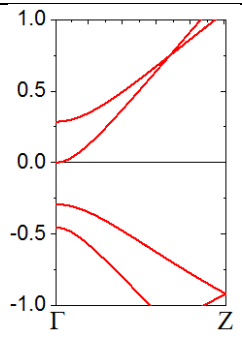
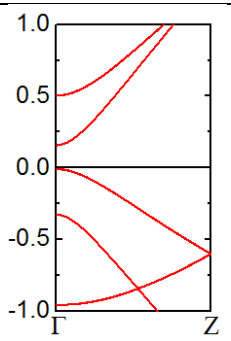
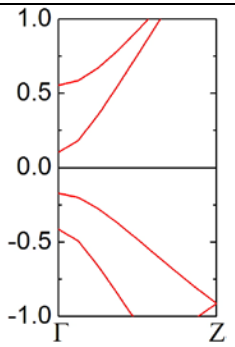
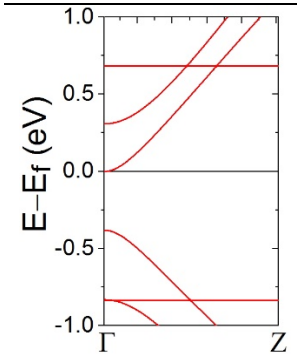
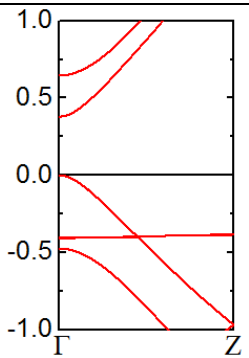
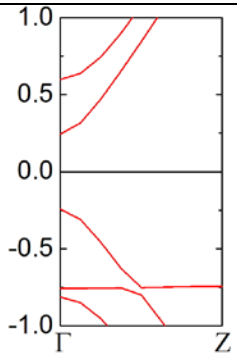
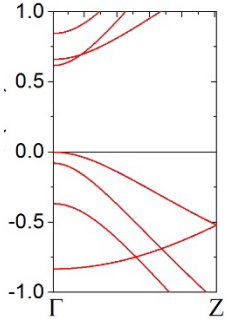
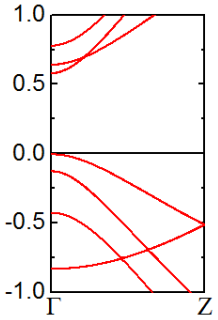
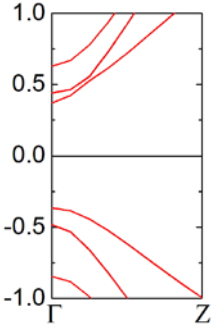


Figure S2. The variation of αGNTs' band gaps as a function of tube size calculated by various level of theories.

Band gap oscillation of Z-αGNTs were demonstrated with all the calculation methods, DFTB, VASP-PBE, PBE-D (ref. 19), and VASP-HSE06. Moreover, band gaps of small αGNTs with n = 2 – 5 calculated by DFTB were closer to VASP-HSE06 results than other DFT results. Nearly zero-gap of A-αGNTs with N_a = 2 – 7 were also well reproduced with DFTB, VASP-PBE, and PBE-D (ref. 19) calculations.

2.2. Comparison of band structures of Z-, A-, and C-αGNTs calculated by DFTB, VASP-PBE, and VASP-HSE06 done by us.

i) *n*–*m* = *n* (Z-αGNT)

n,*m* 2,0 (DFTB)	(VASP-PBE)	(VASP-HSE06)	3,0 (DFTB)	(VASP-PBE)	(VASP-HSE06)
					
n,*m* 4,0 (DFTB)	(VASP-PBE)	(VASP-HSE06)	5,0 (DFTB)	(VASP-PBE)	(VASP-HSE06)
					

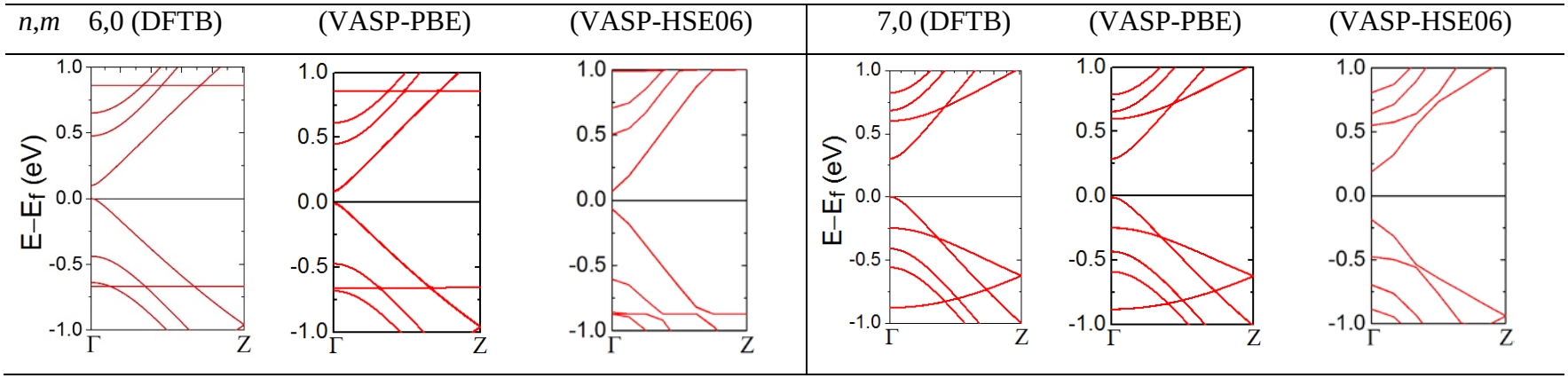


Figure S3. Band structures of Z-αGNTs with n = 2 – 7 calculated at the DFTB, VASP-PBE, and VASP-HSE06 level of theories.

ii) n–m = 0 (A-αGNT)

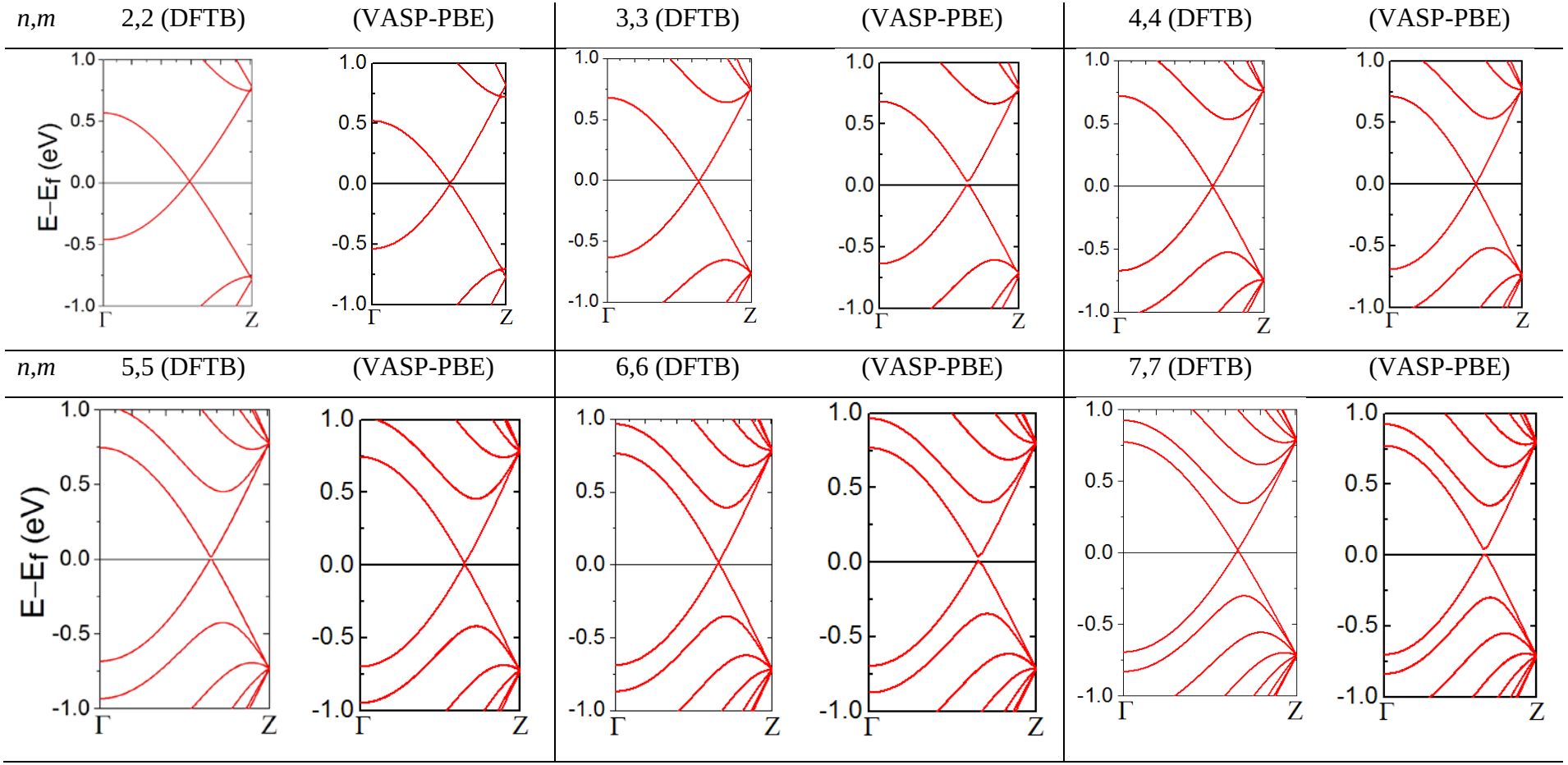


Figure S4. Band structures of A-αGNTs with n = 2 – 7 calculated at the DFTB/mio-1-1 and PBE levels.

iii) *n*–*m* = 3*a* (*n* = 4, 5, 6; C-αGNT)

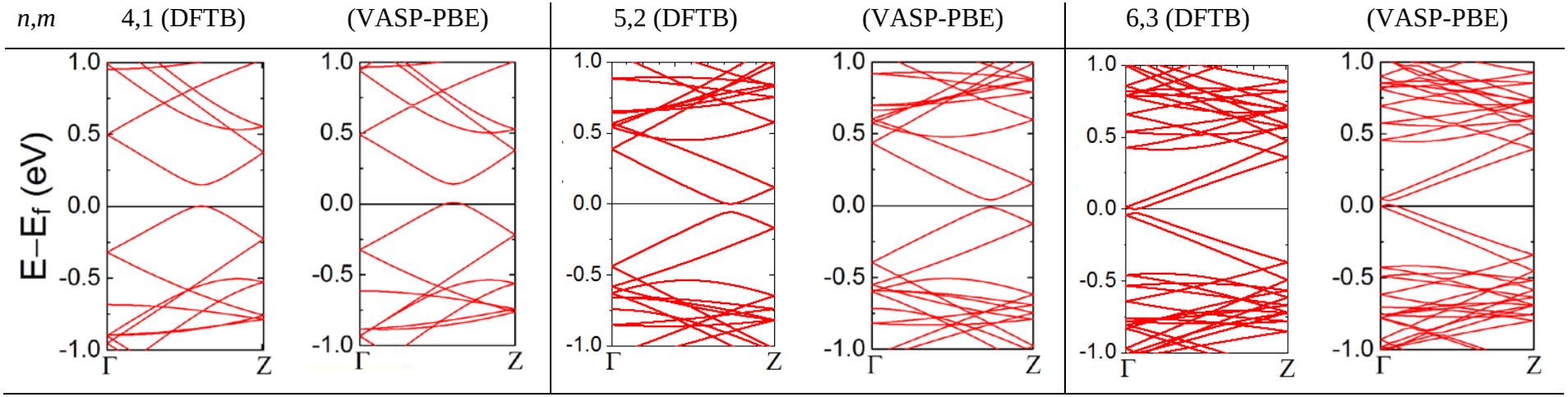


Figure S5. Band structures of selected C-αGNTs with *n*–*m* = 3*a* calculated at the DFTB and VASP-PBE level of theories.

iv) $n–m = 3a+1$ (n = 2, 3; C-αGNT)

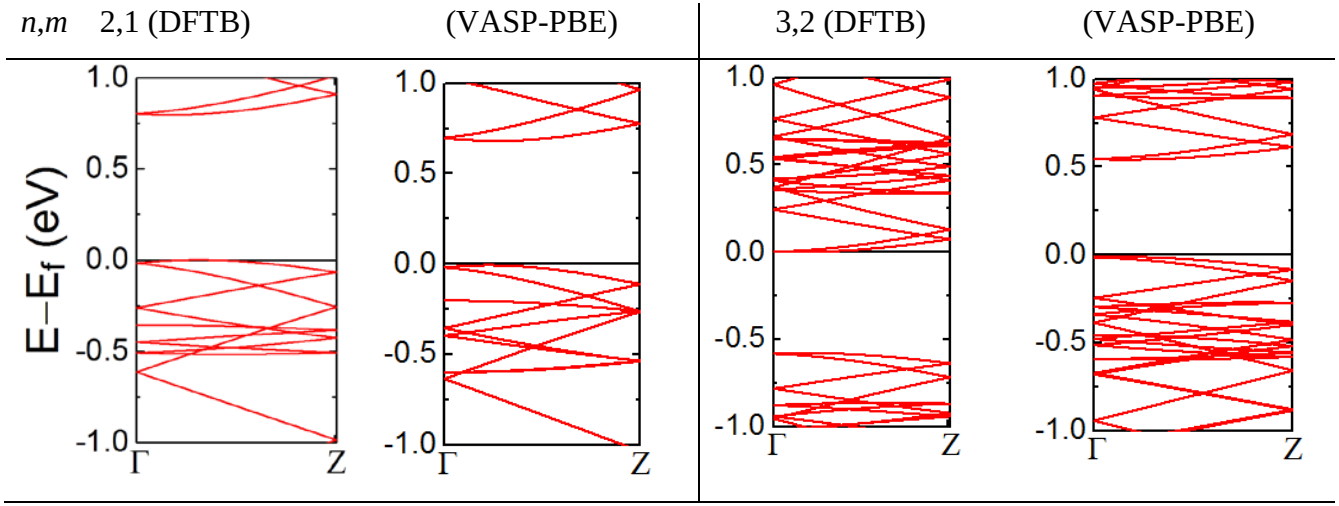


Figure S6. Band structures of selected C-αGNTs with $n–m = 3a+1$ calculated at the DFTB and VASP-PBE level of theories.

v) $n–m = 3a+2$ (C-αGNT)

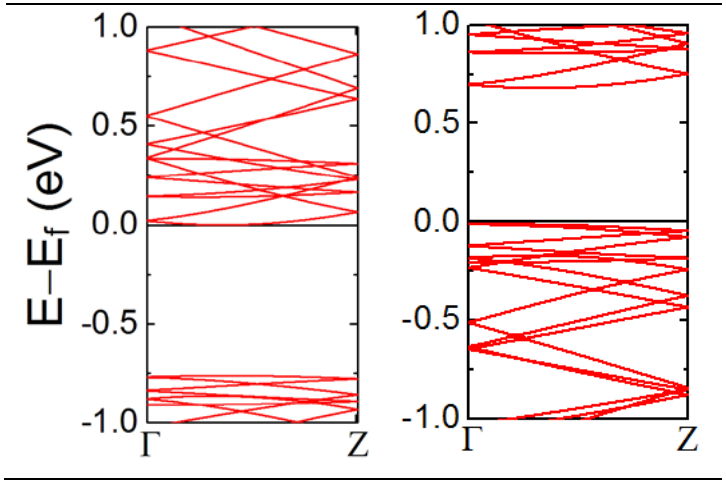


Figure S7. Band structures of selected C-αGNTs with $n–m = 3a+2$ calculated at the DFTB and VASP-PBE level of theories.

3. Density of states of αGNTs calculated by DFTB method.

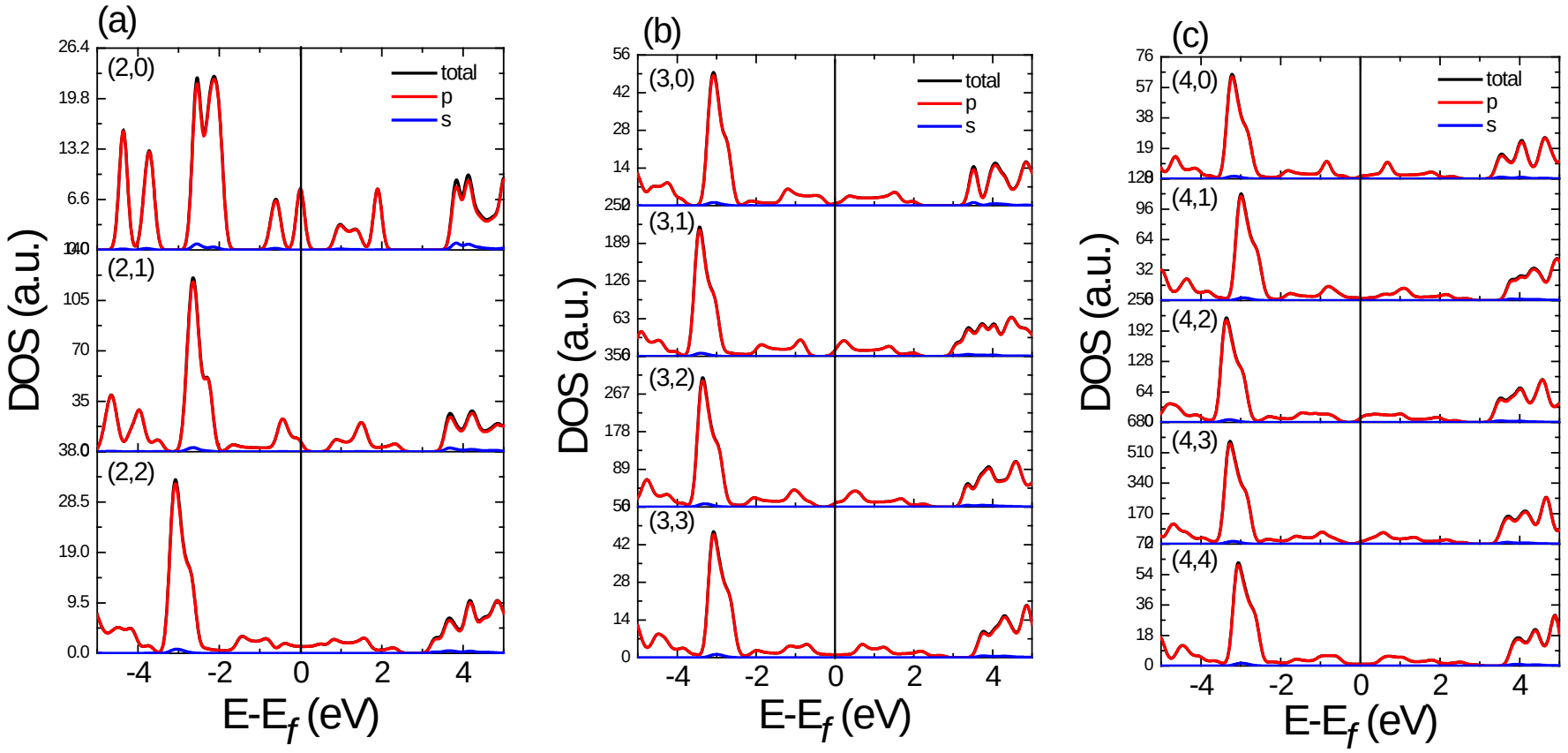


Figure S8. Density of states diagrams of αGNTs with *n* = (a) 2, (b) 3, (c) 4, (d) 5, (e) 6, and (f) 7 calculated at the DFTB method. (*n*,*m*) parameters are given.

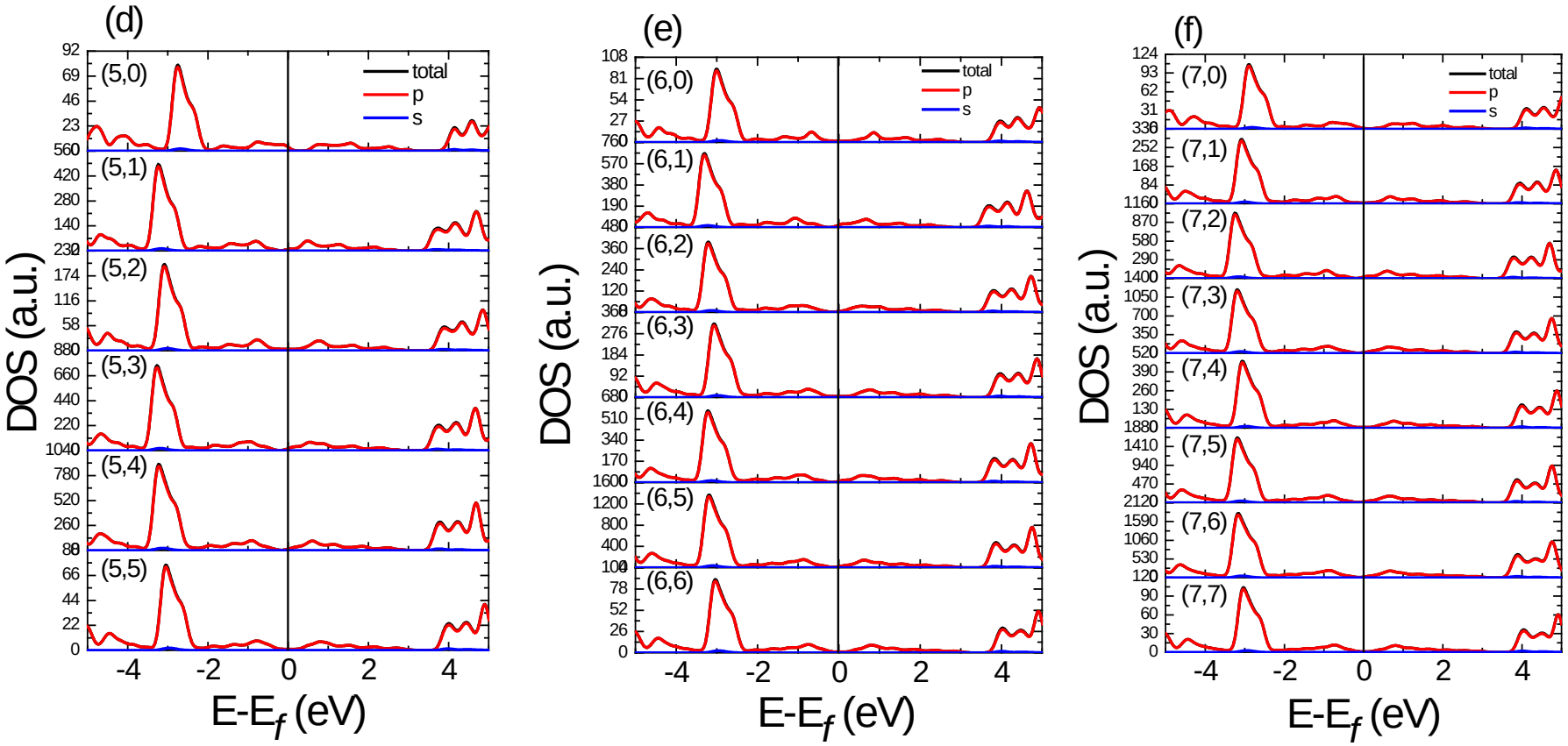


Figure S8. Density of states diagrams of αGNTs with n = (a) 2, (b) 3, (c) 4, (d) 5, (e) 6, and (f) 7 calculated at the DFTB method. (n,m) parameters are given. (Continued)

4. Empirical fitting of band gaps of αGNTs.

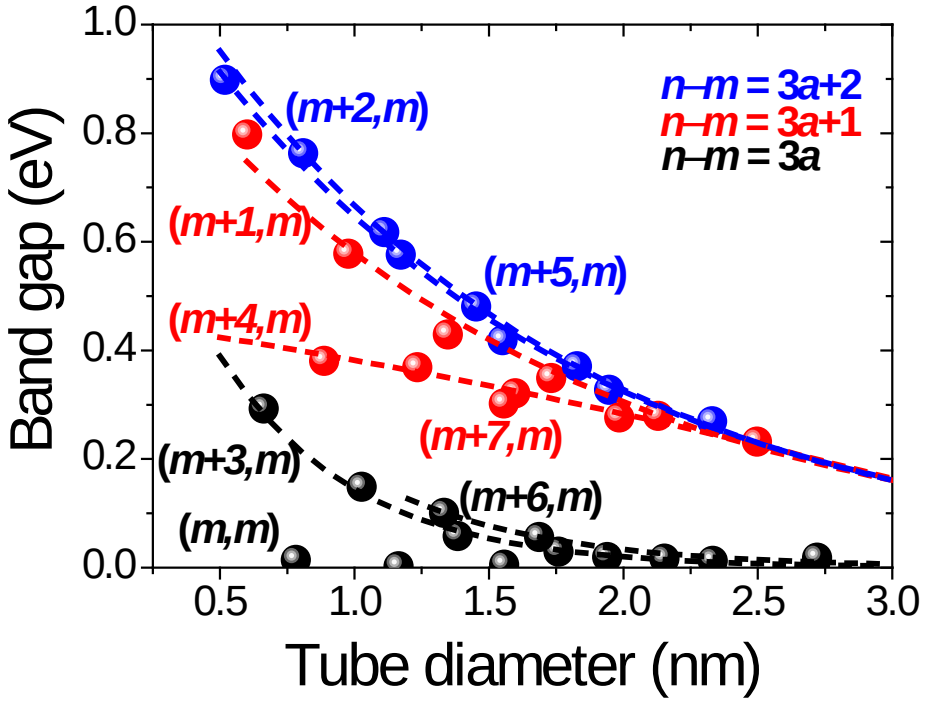


Figure S9. Empirical fitting of band gaps of αGNTs as a function of tube diameter.

Empirical equations used in Figure S9. [a]

(a) $n - m = 1$, band gap (eV) = $1.0976 * e^{-0.064x}$

(b) $n - m = 2$, band gap (eV) = $1.2889 * e^{-0.069x}$

(c) $n - m = 3$, band gap (eV) = $1.0478 * e^{-0.198x}$

(d) $n - m = 4$, band gap (eV) = $-0.0001x^2 - 0.0069x + 0.461$

(e) $n - m = 5$, band gap (eV) = $1.3567 * e^{-0.071x}$

(f) $n - m = 6$, band gap (eV) = $0.9261 * e^{-0.166x}$

[a] Tube diameter in nm was denoted as x.