Electronic and optical properties of carbon nanotubes : A first-principles study

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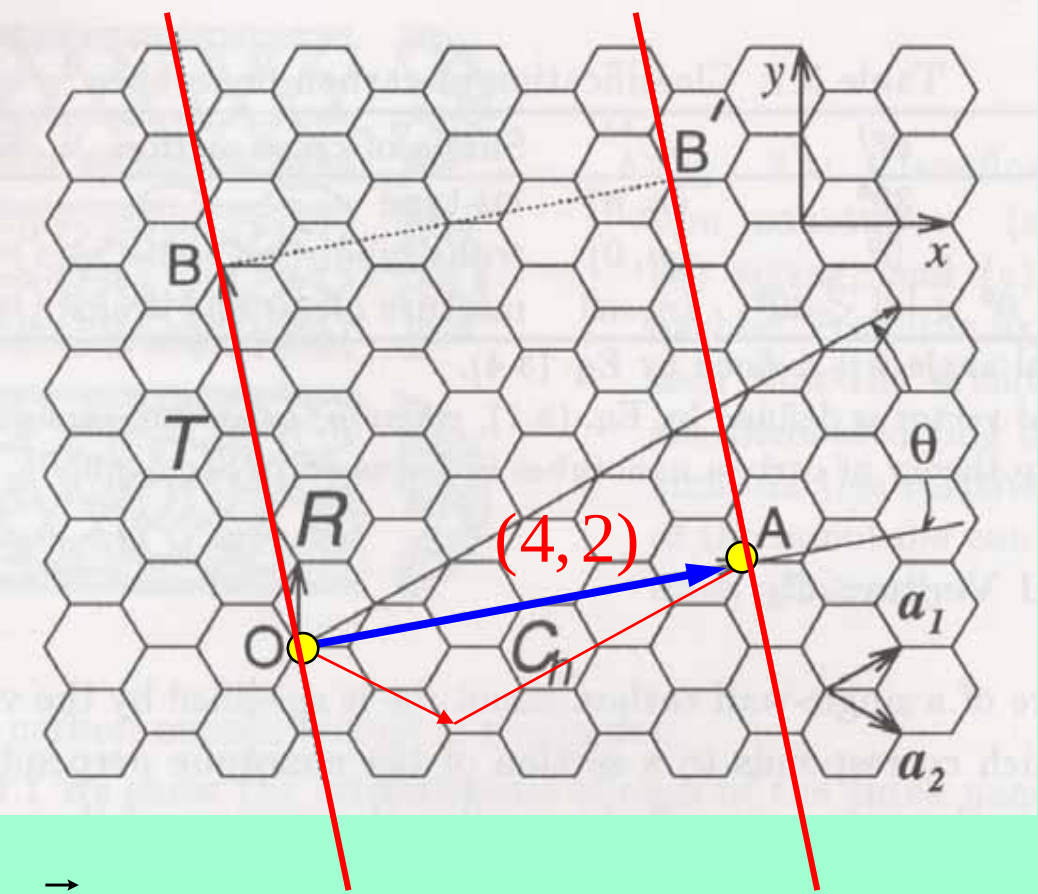
OUTLINE

The Electronic and Optical Properties of SWCNTs

The Effect of the External Electric Field on the Electronic and Optical Properties of SWCNTs

The Effect of the Intertube Interaction on the Electronic and Optical Properties of SWCNT Bundles

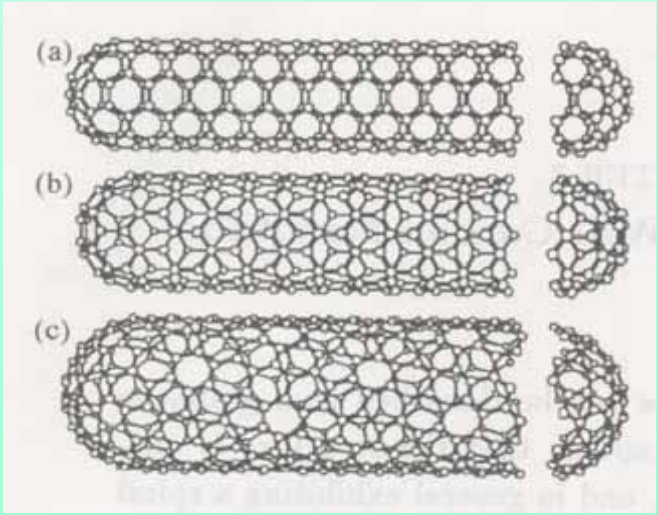
Single Walled Carbon Nanotubes (SWCNTs)



nonchiral nanotube

(n,n) armchair ; (n,0) zigzag

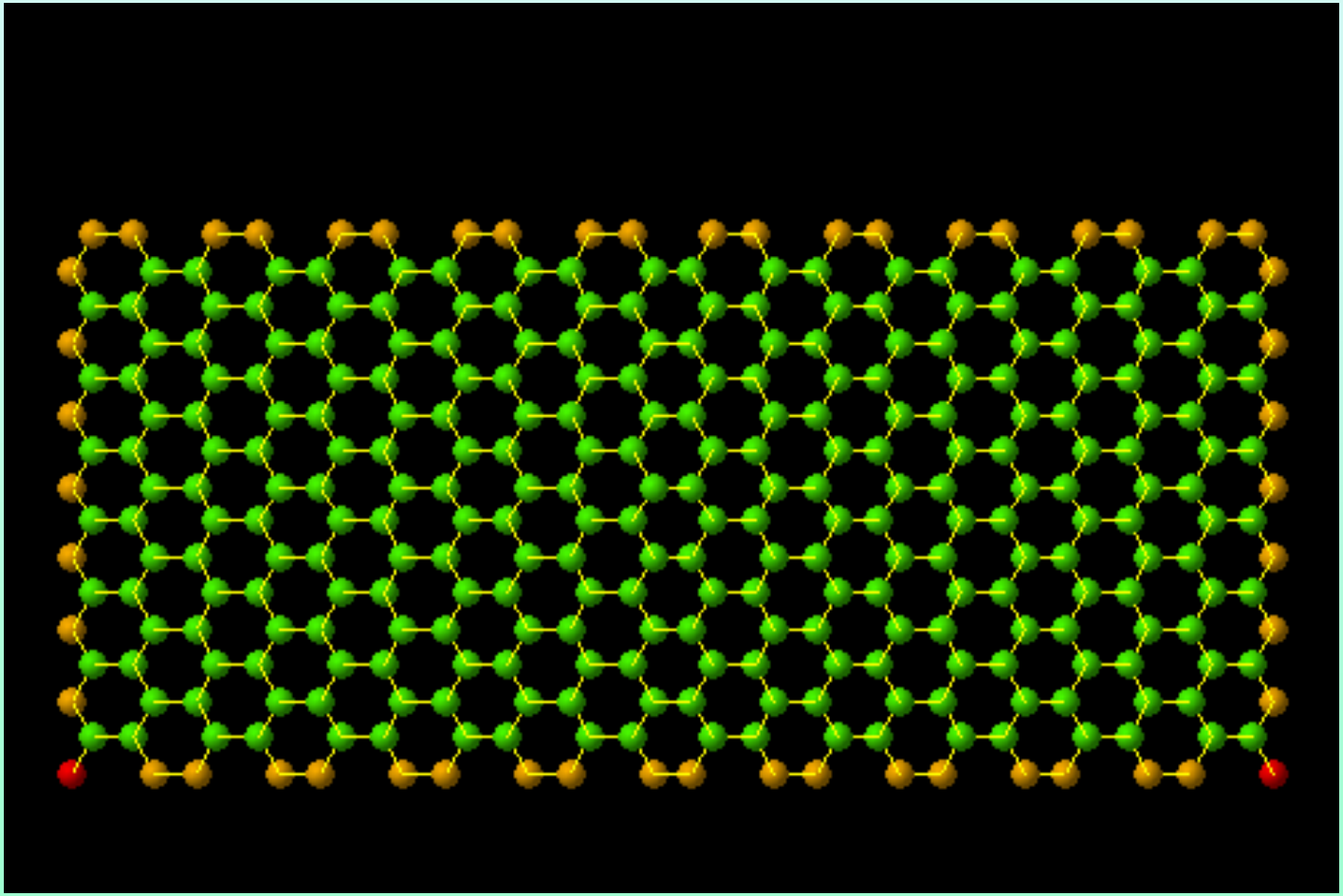
chiral nanotube otherwise



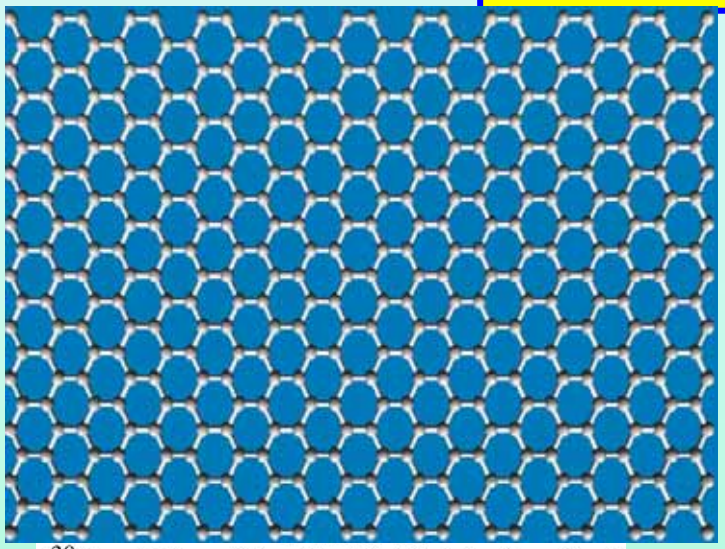
$$\vec{C}_h = n\vec{a}_1 + m\vec{a}_2 \equiv (n,m) \qquad 0 \le m \le n$$

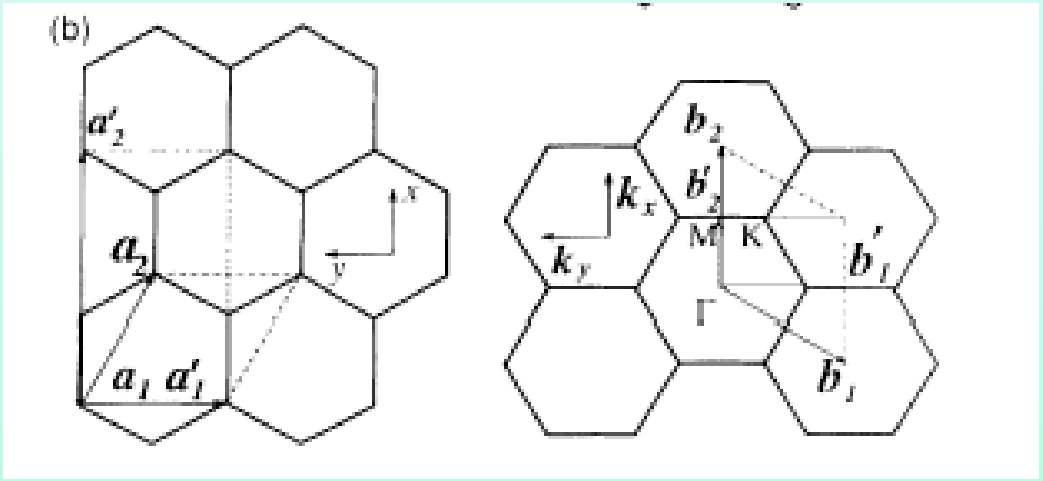
(chiral vector)

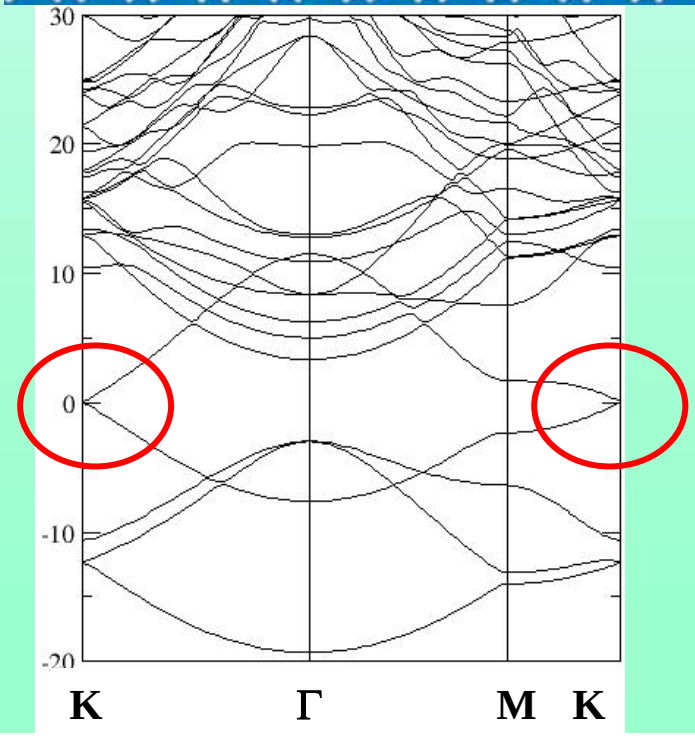
Armchair tube (10,10)

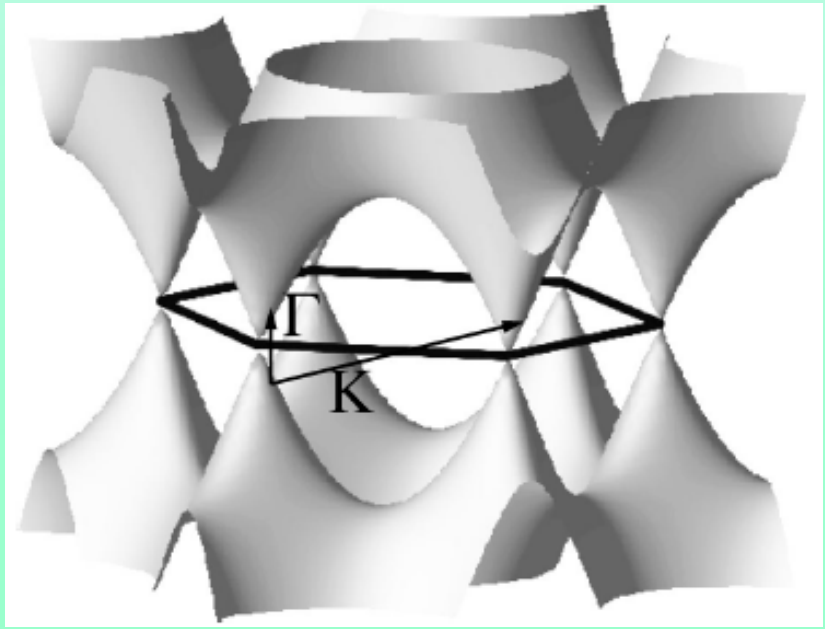


Graphite Sheet







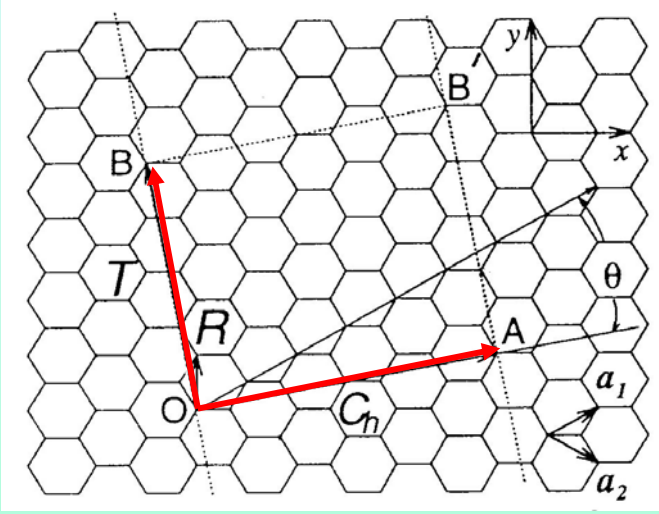


Zone Folding Method

Band Structure of Graphite Sheet ➔ the Band Structure of SWCNTs

$$\vec{C_h}\cdot\vec{K_1}=2\pi \qquad \vec{T}\cdot\vec{K_1}=0$$

$$\vec{C_h}\cdot\vec{K_2}=0 \qquad \vec{T}\cdot\vec{K_2}=2\pi$$

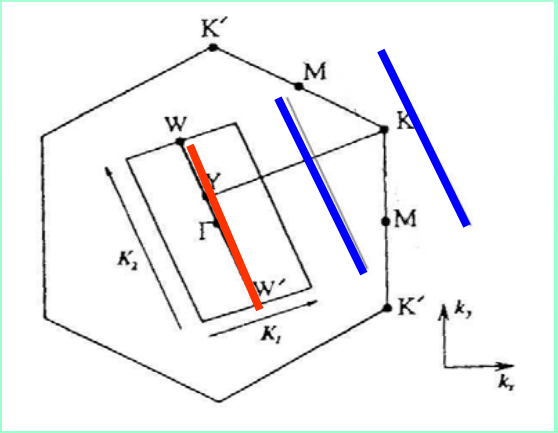


$$\psi(\vec{r}+\vec{C_h})=\psi(\vec{r}) \Rightarrow \exp(i\vec{k}\cdot\vec{C_h})=1$$

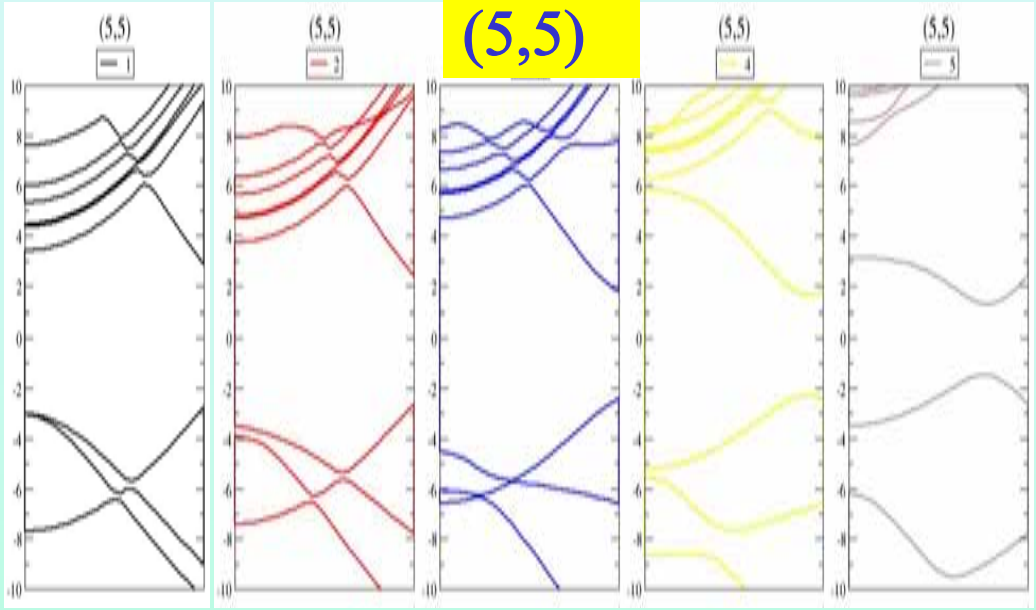
$$\vec{k}=x_1\vec{K_1}+x_2\vec{K_2} \Rightarrow \vec{k}\cdot\vec{C}_n=x_1\vec{K_1}\cdot\vec{C_h}+x_2\vec{K_2}\cdot\vec{C_h}=2\pi q$$

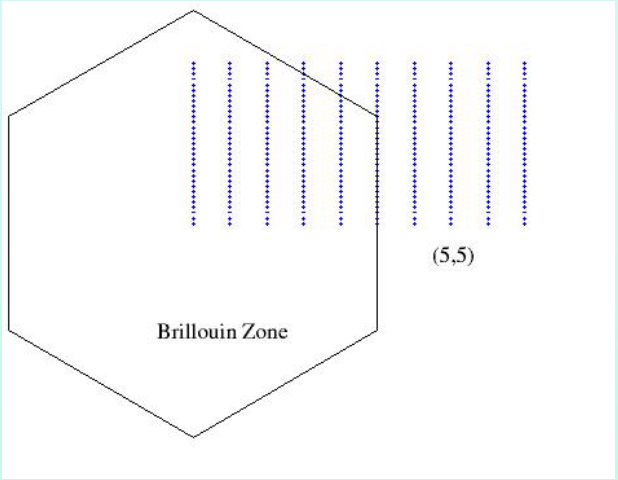
$$\vec{k}=q\vec{K_1}+x_2\frac{\vec{K_2}}{|\vec{K_2}|} \qquad K_1=2\pi/D$$

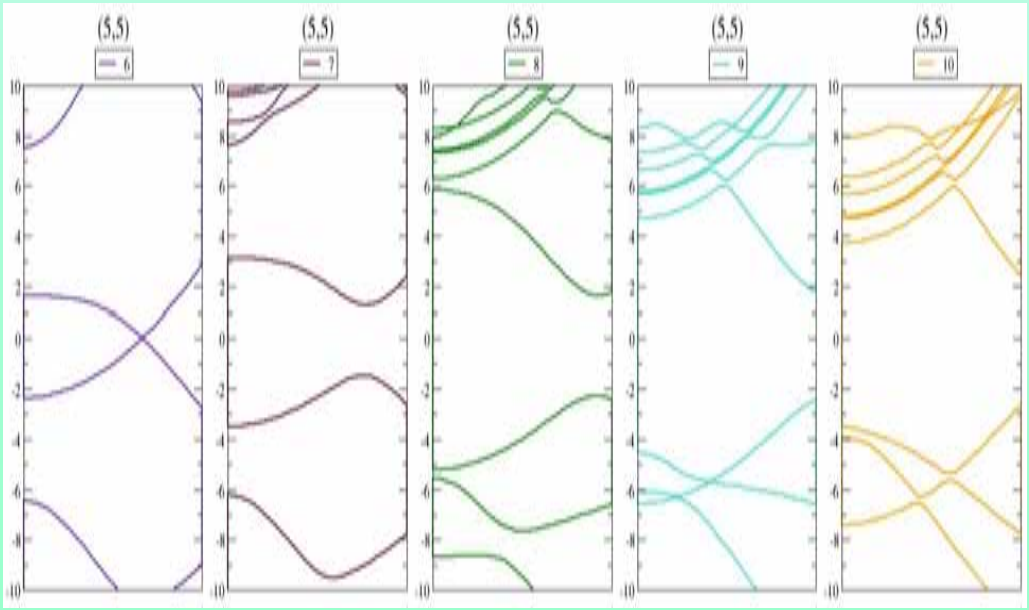
$$q=0,1,2\cdots,N-1 \qquad -\frac{\pi}{T}<x_2<\frac{\pi}{T}$$

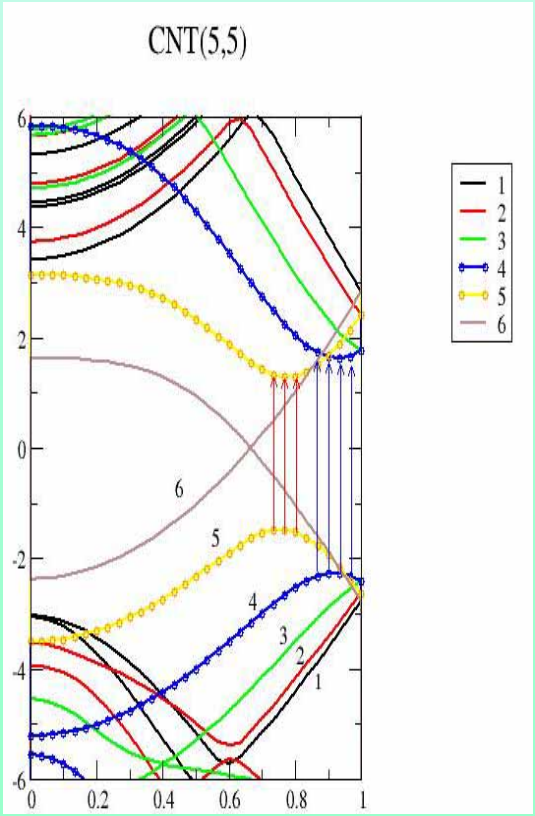


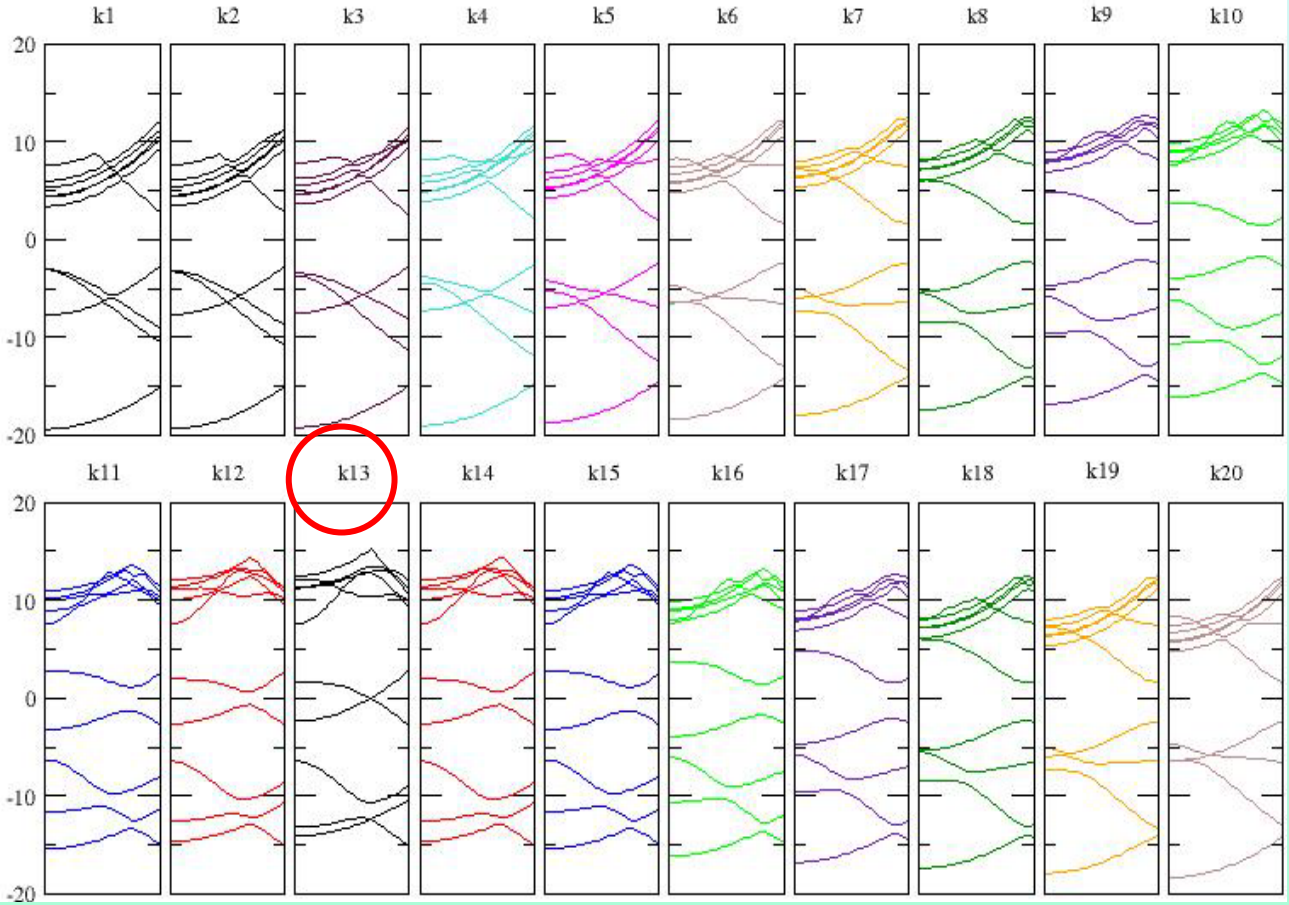
(5,5)

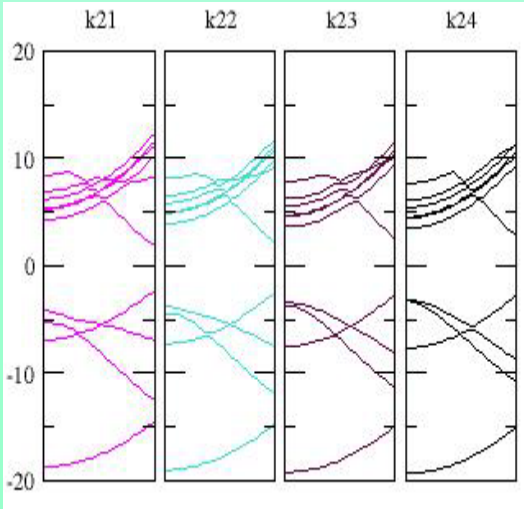

(5,5)
1
(5,5)
2
(5,5)
4
(5,5)
5

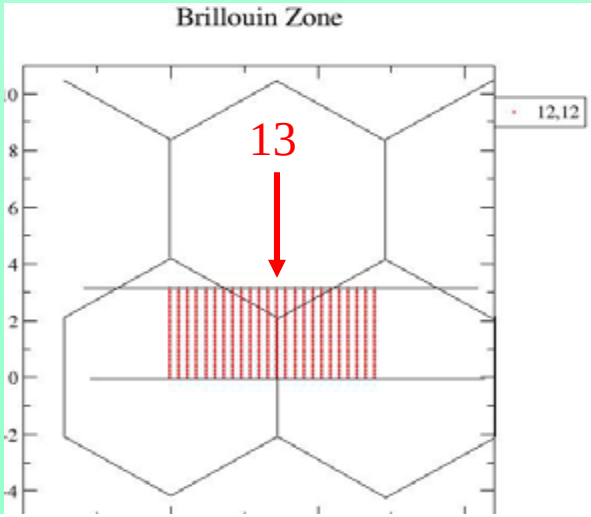

(5,5)
Brillouin Zone


(5,5)
6
(5,5)
7
(5,5)
8
(5,5)
9
(5,5)
10

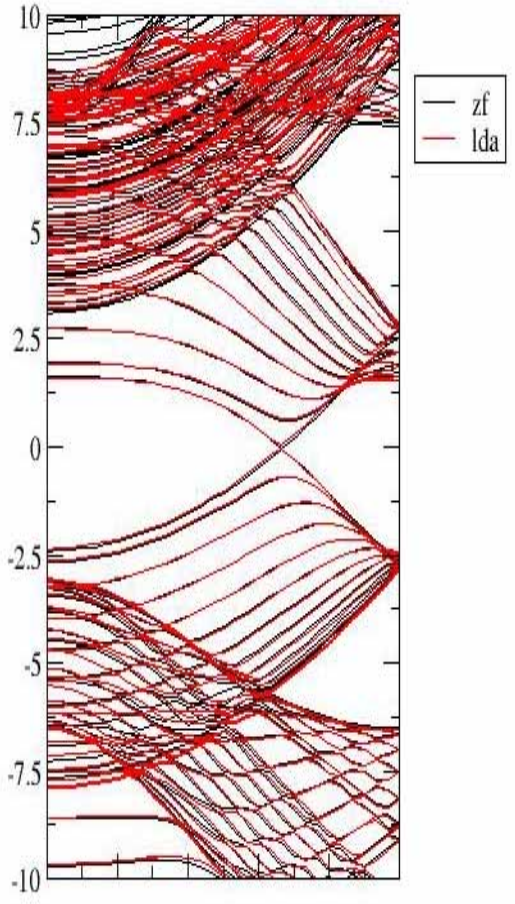

CNT(5,5)
1
2
3
4
5
6
0
0.2
0.4
0.6
0.8
1


k1
k2
k3
k4
k5
k6
k7
k8
k9
k10
k11
k12
k13
k14
k15
k16
k17
k18
k19
k20
20
10
0
-10
-20

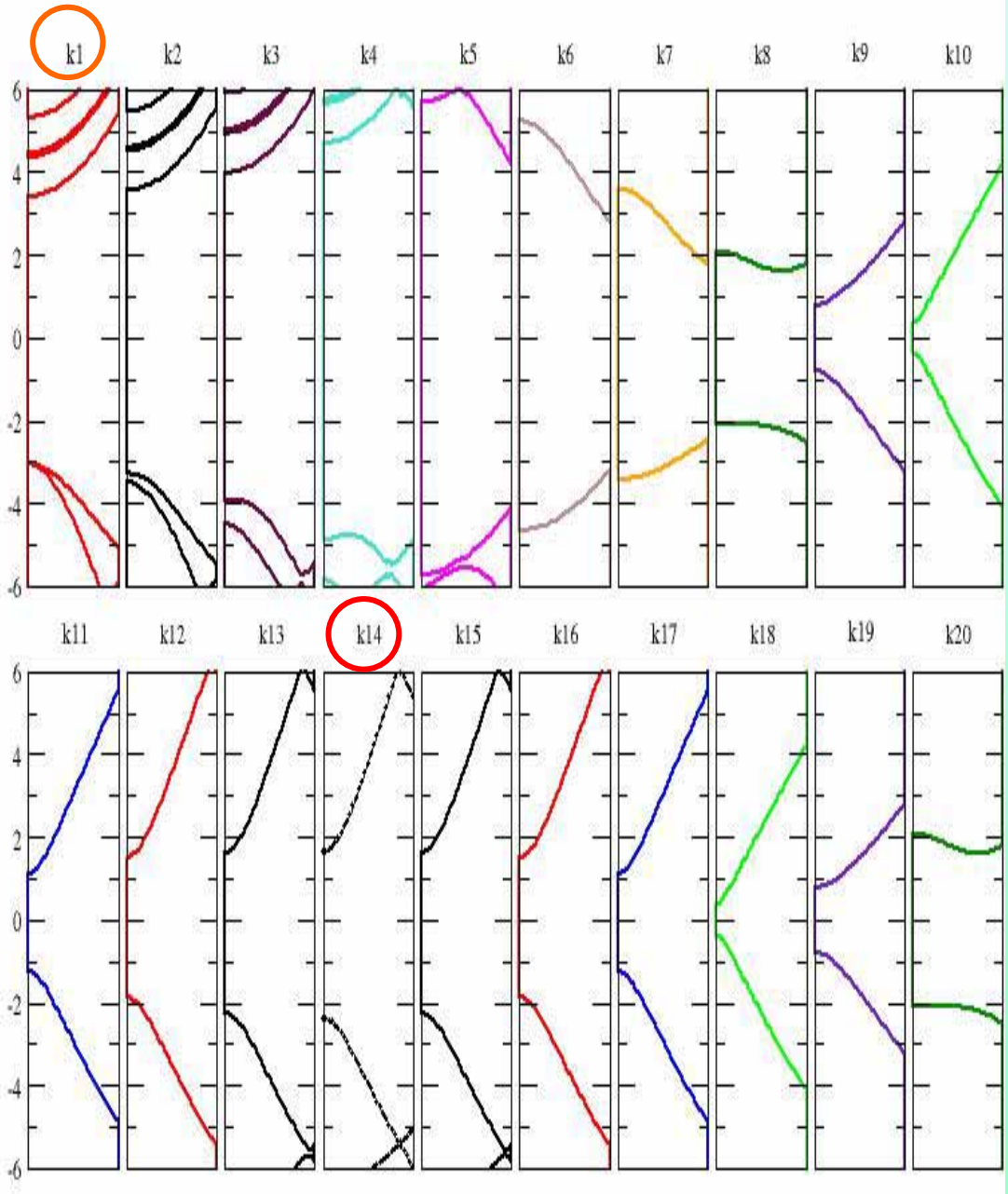

k21
k22
k23
k24
20
10
0
-10
-20

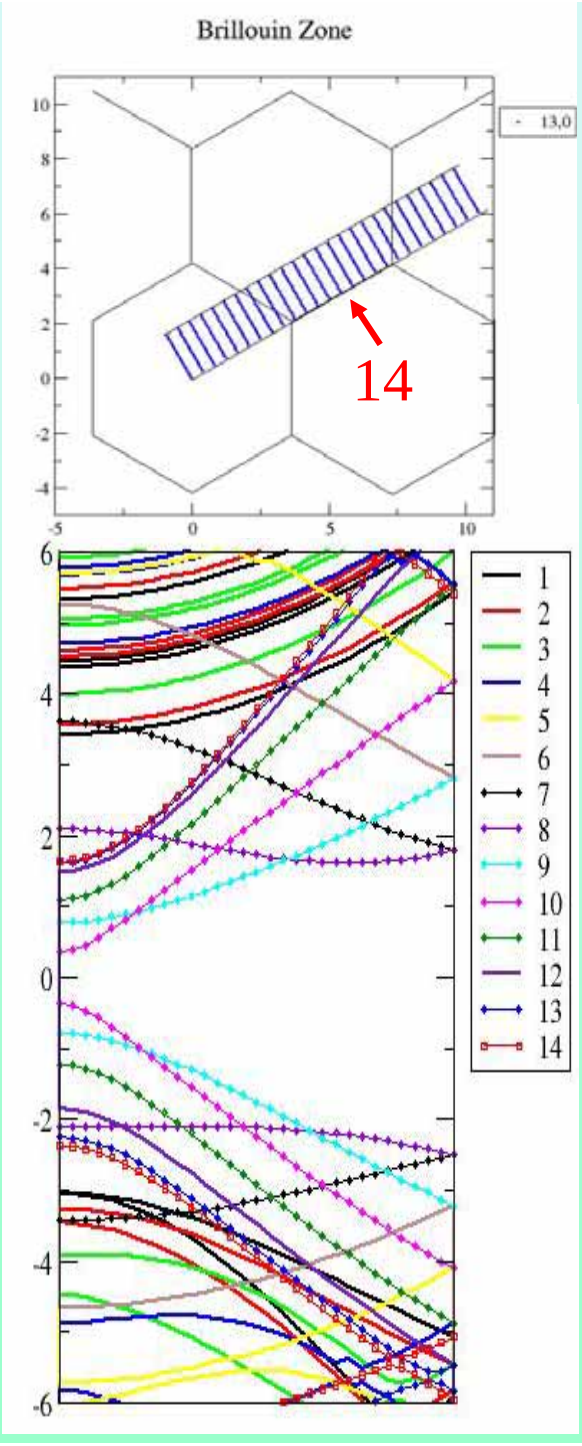

Brillouin Zone
13
12,12

(12,12)

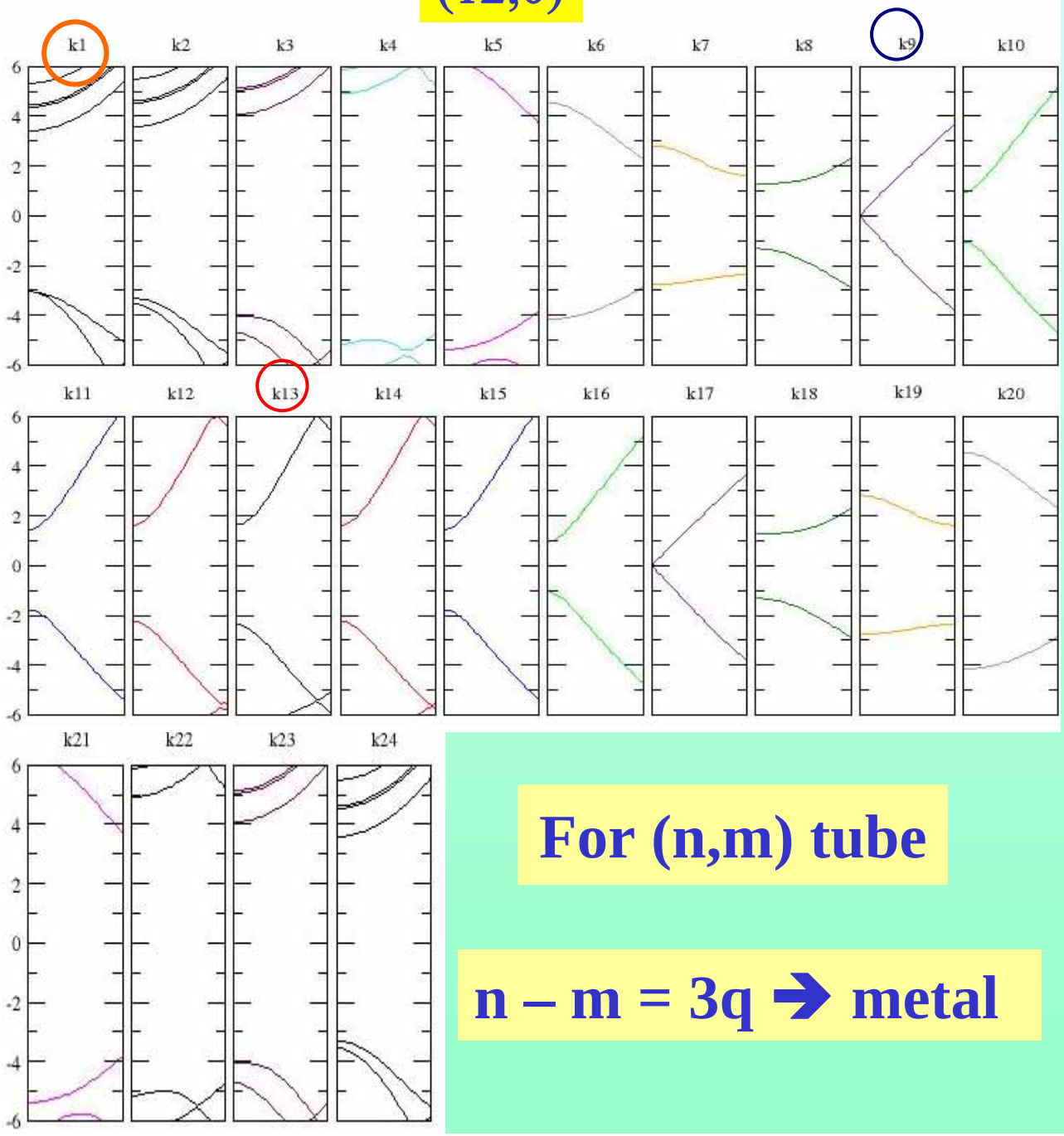

10
7.5
5
2.5
0
-2.5
-5
-7.5
-10
zf
lda

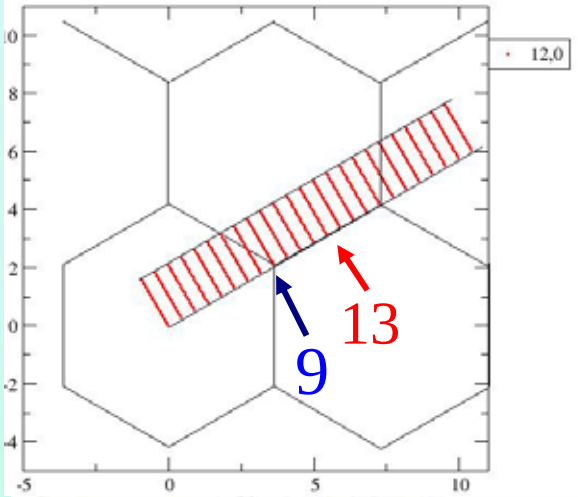
(13,0)

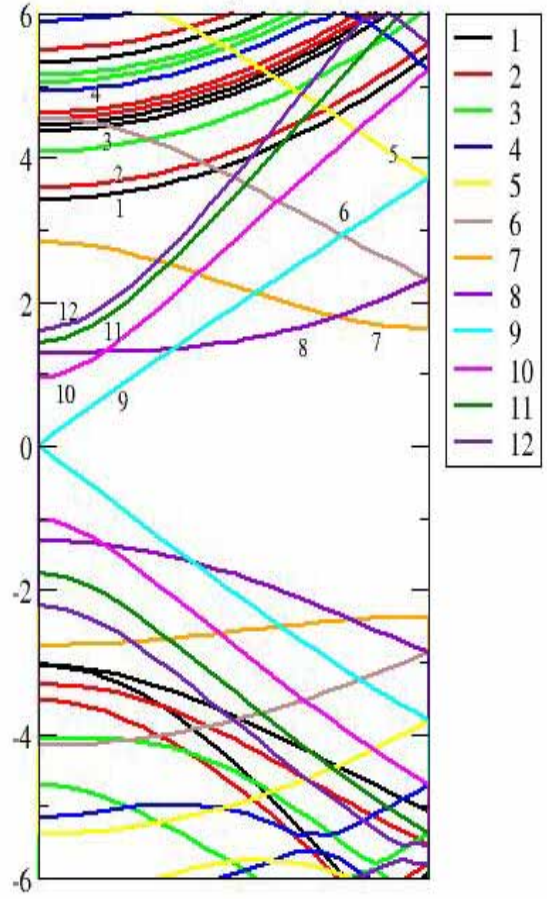

k1
k2
k3
k4
k5
k6
k7
k8
k9
k10
k11
k12
k13
k14
k15
k16
k17
k18
k19
k20


Brillouin Zone
13,0
14
1
2
3
4
5
6
7
8
9
10
11
12
13
14

(12,0)



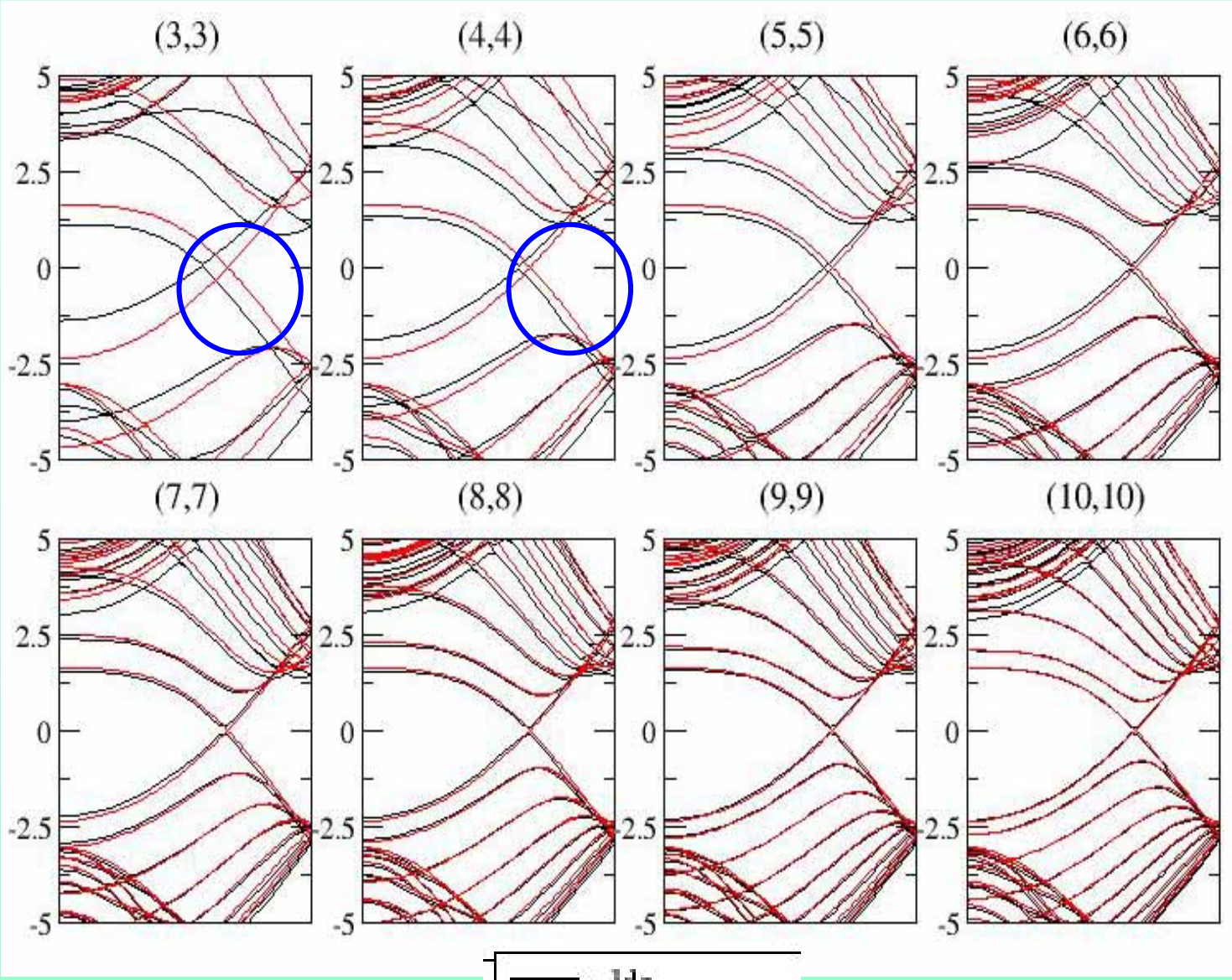




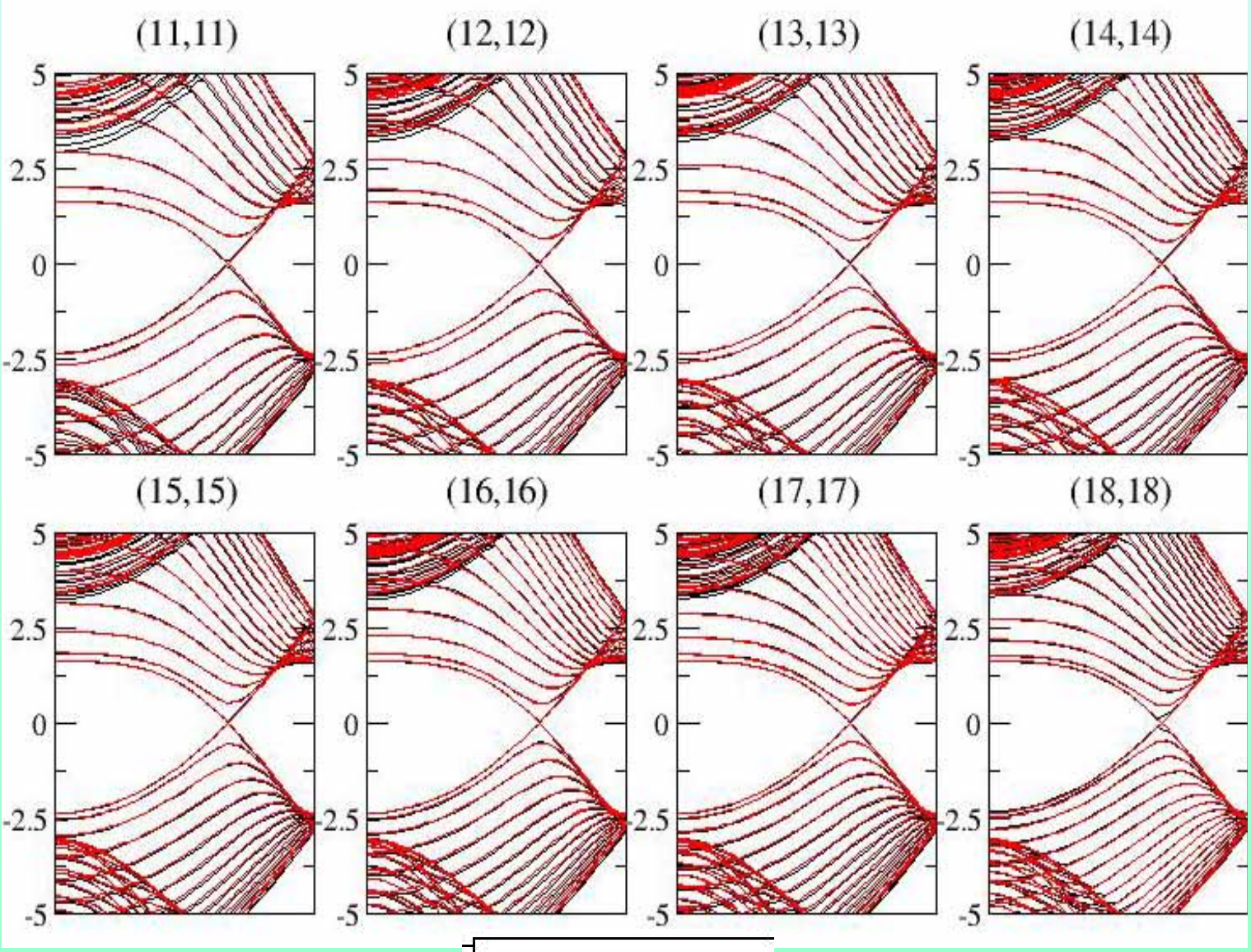
For (n,m) tube

n – m = 3q ➔ metal

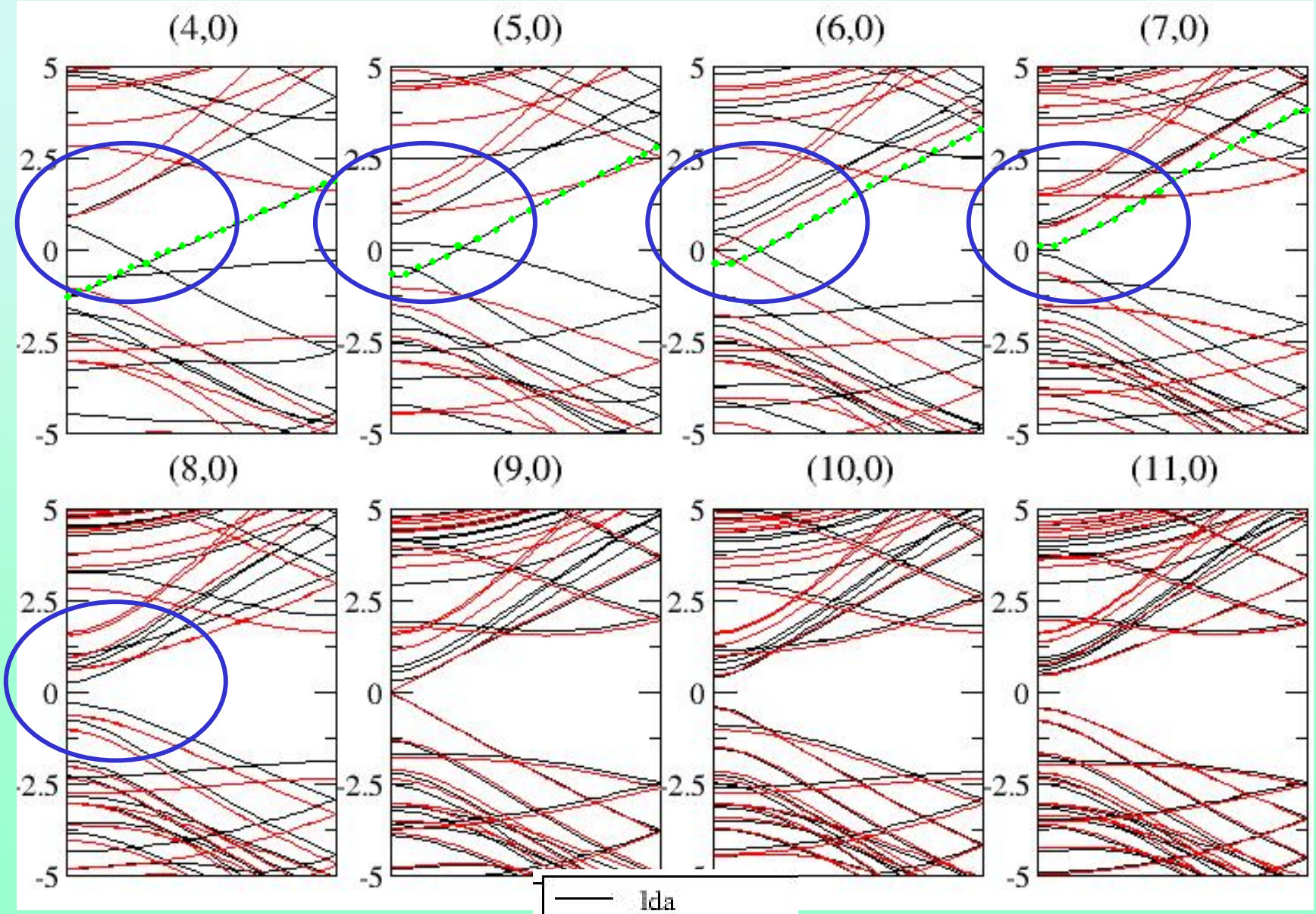
Band Structure of armchair tube (ZF *vs* real LDA calculation)



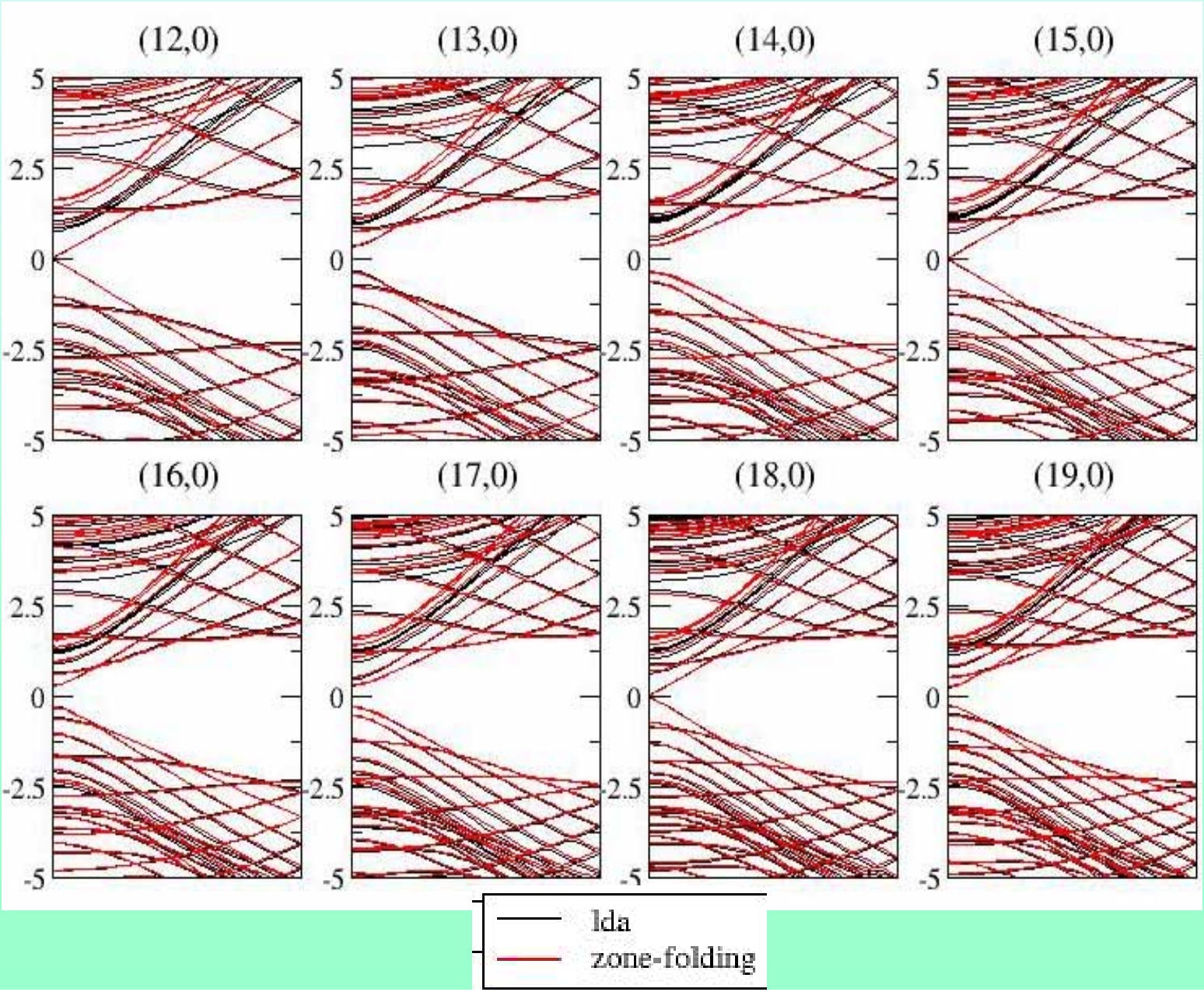
Band Structure of armchair tube (ZF *vs* real LDA calculation)



Band Structure of zigzag tube (ZF *vs* real LDA calculation)

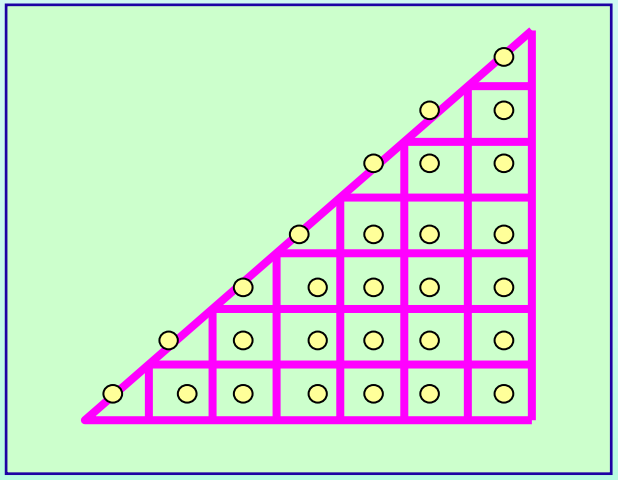


Band Structure of zigzag tube (ZF *vs* real LDA calculation)

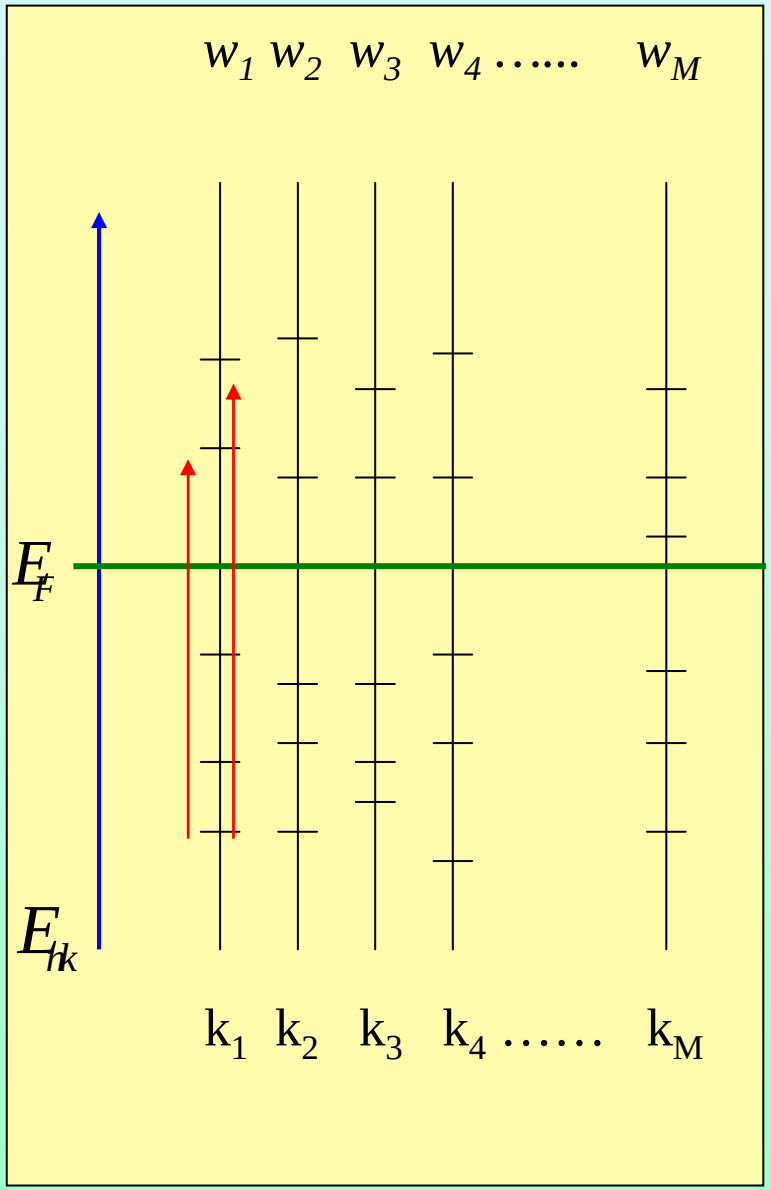


Optical Properties of Graphene and SWCNTs

Imaginary part of dielectric function

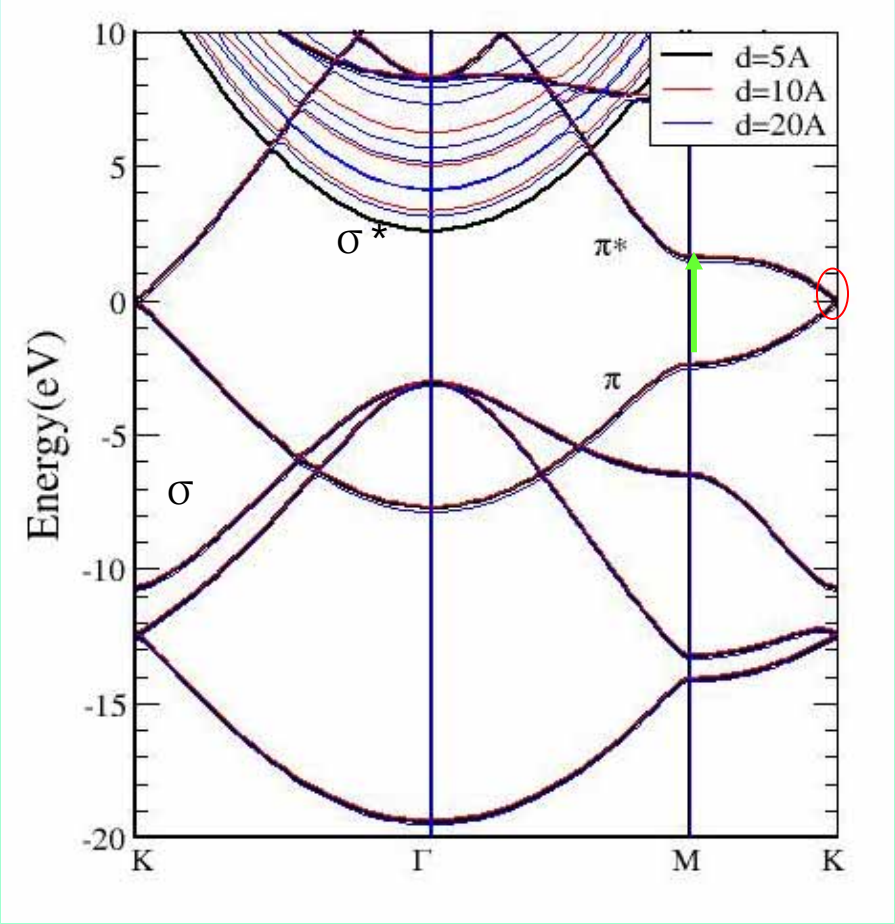


$$\vec{M}_{cv} = \left\langle \phi_c \mid \vec{p} \mid \phi_v \right\rangle$$

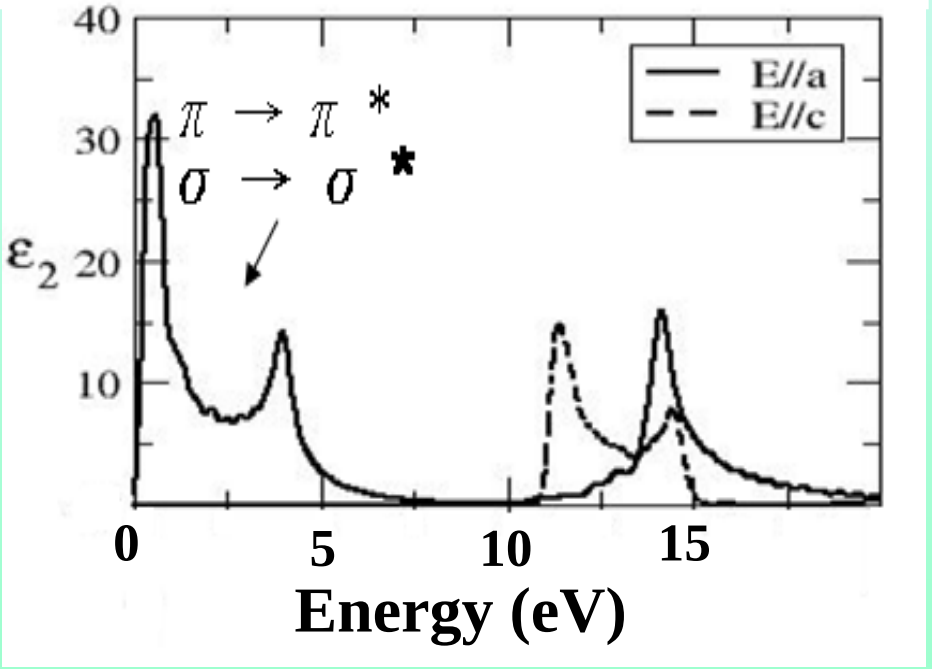


$$\varepsilon_2(\omega) = \frac{4\pi e^2}{m^2\omega^2}\frac{V}{(2\pi)^3}\sum_v\sum_c\int_{Bz} d^3k \left|\vec{\varepsilon}\cdot\vec{M}_{cv}(\vec{k})\right|^2 \delta\left[E_c(\vec{k}) - E_v(\vec{k}) - \hbar\omega\right]$$

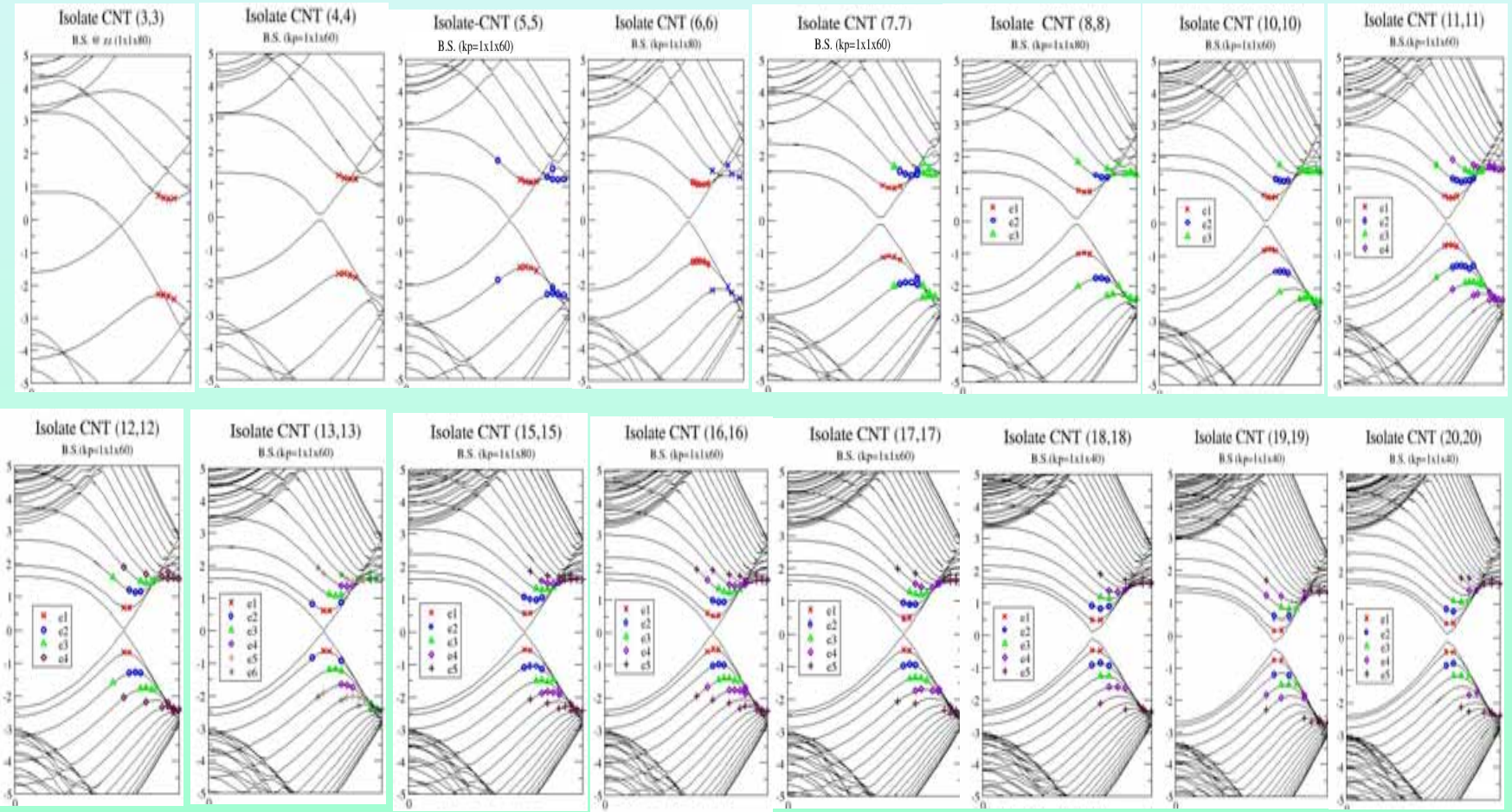
Band structure and optical properties of Graphene



The imaginary part of the dielectric function ε_2

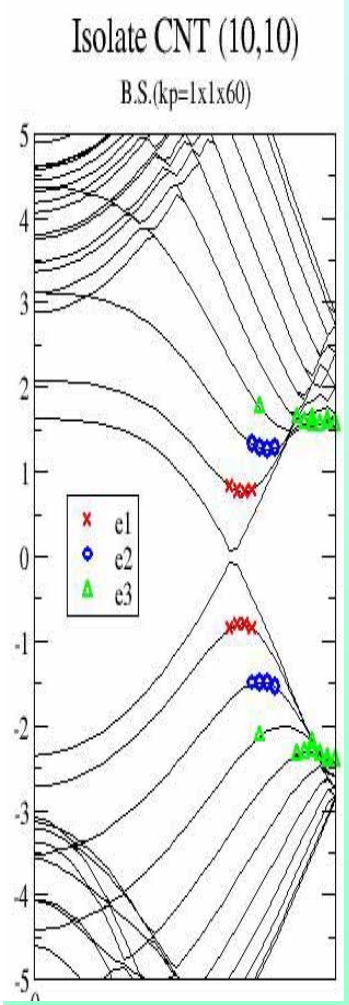


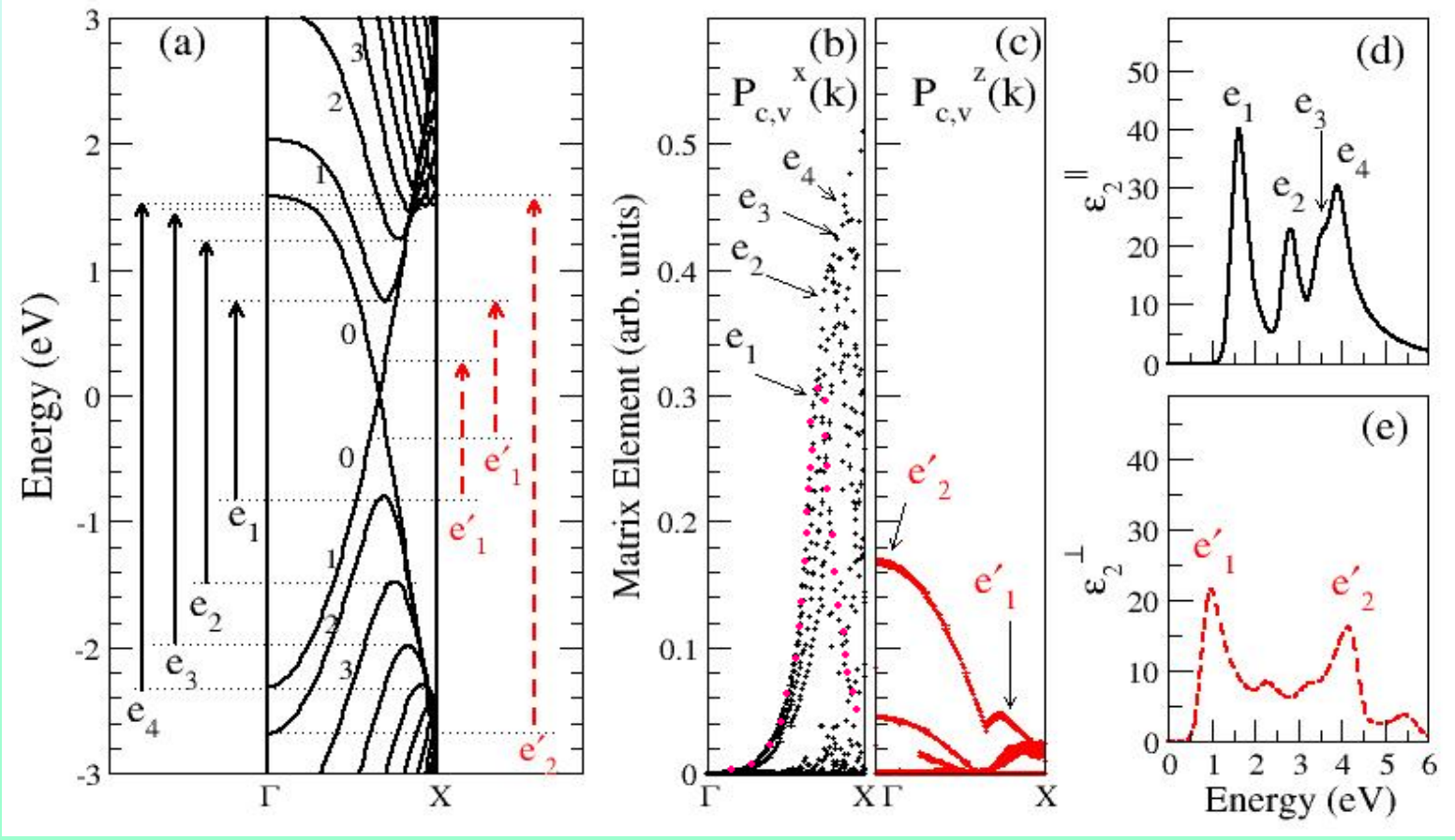
Armchair Tubes



Van Hove singularity appears in the joint density of states and the matrix elements between the corresponding valence and conduction states have a nonzero value, a corresponding peak will show up in the dielectric function spectrum. ⇒ $E_{\mu\mu}$ **transition**

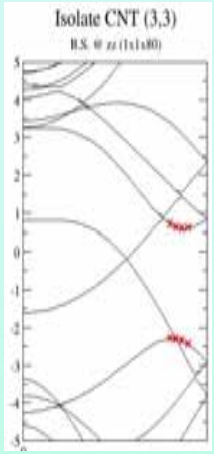
The band structures, matrix elements, and the ε_2 spectrum for (10,10) carbon nanotube

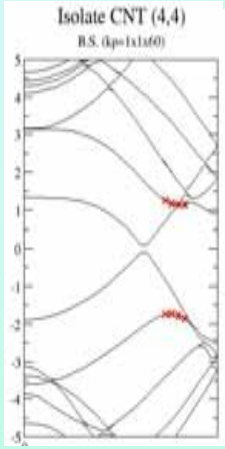


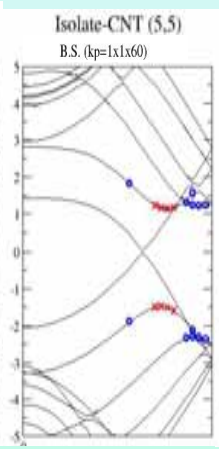


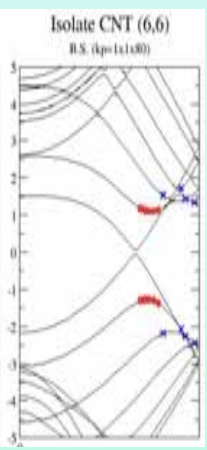
$E_{\mu\mu}$ **transition**

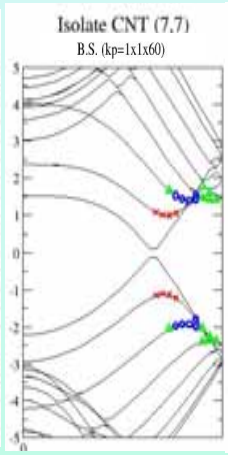
$$p_{cv}^{x} = \left| \langle \phi_c \mid p_x \mid \phi_v \rangle \right|^2$$

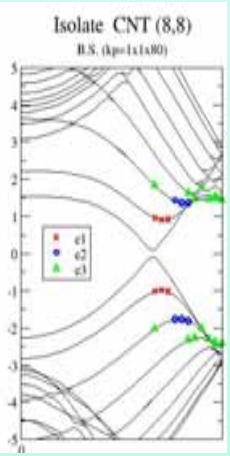

Isolate CNT (3,3)

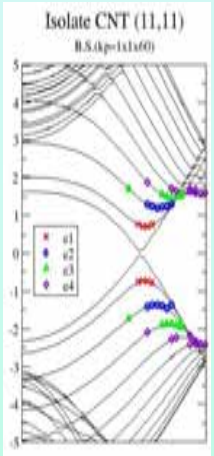

Isolate CNT (4,4)

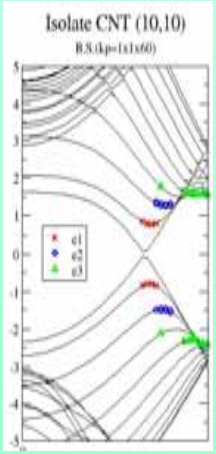

Isolate-CNT (5,5)
B.S. (kp=1x1x60)


Isolate CNT (6,6)


Isolate CNT (7.7)
B.S. (kp=1x1x60)

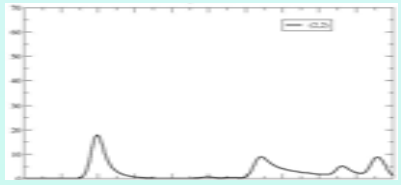

Isolate CNT (8,8)


Isolate CNT (11,11)
B.S.(kp=1x1x60)

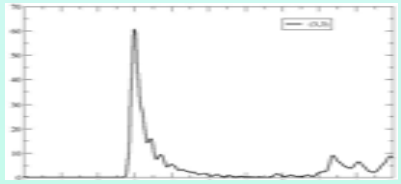

Isolate CNT (10,10)
B.S.(kp=1x1x60)

ε_2 at zz direction

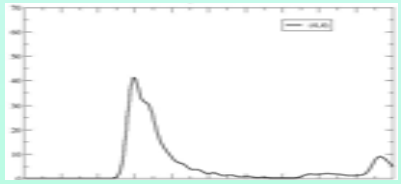
(2,2)



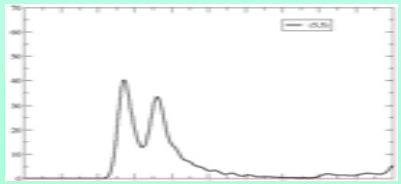
(3,3)



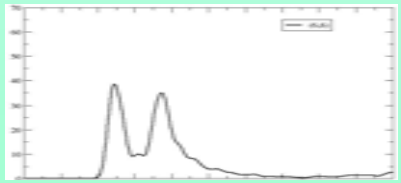
(4,4)



(5,5)



(6,6)



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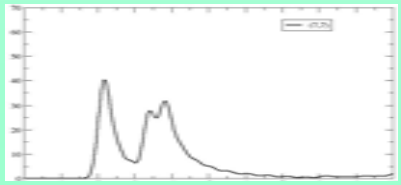

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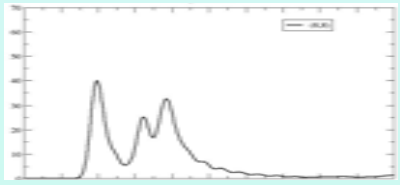

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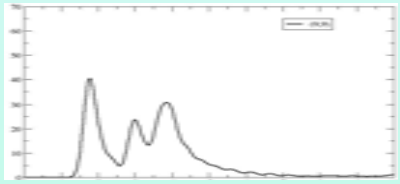

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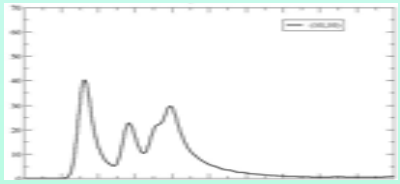

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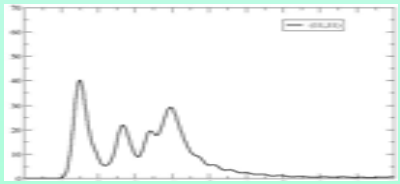

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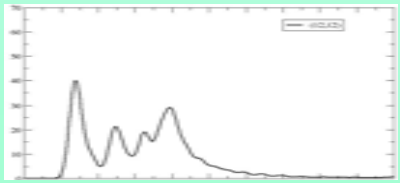

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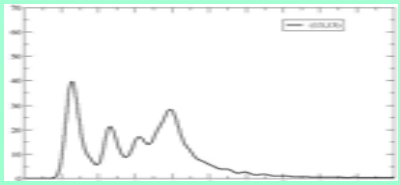

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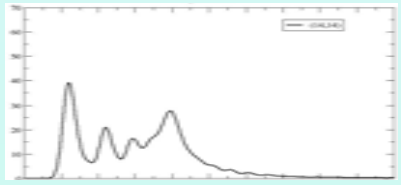

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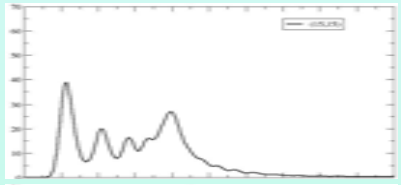

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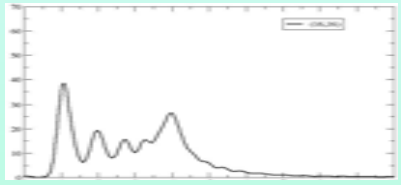

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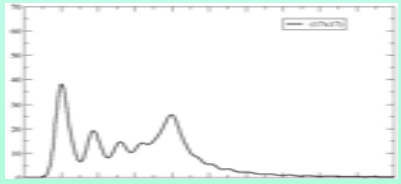

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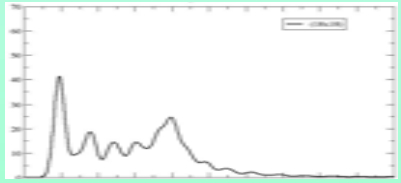

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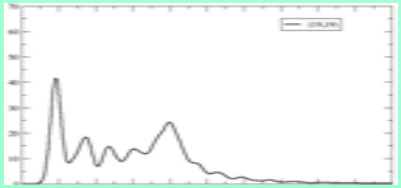

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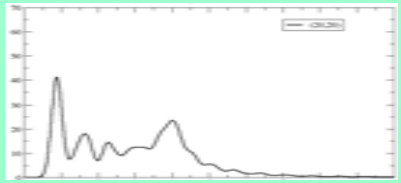
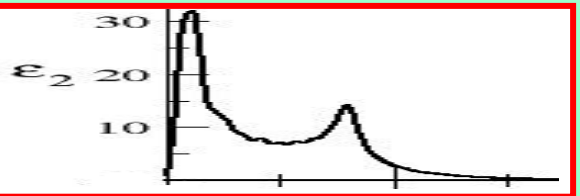
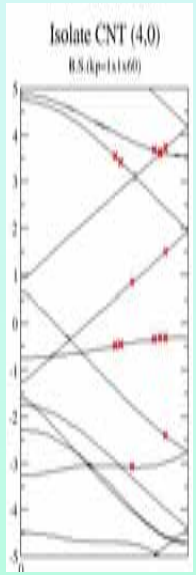
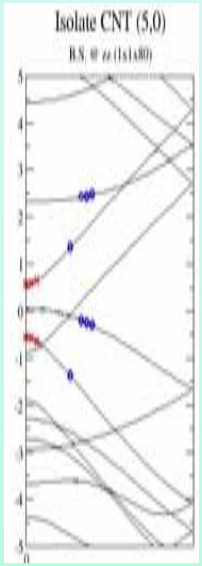


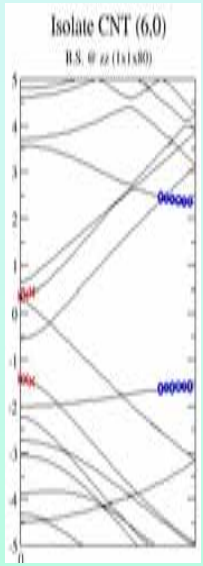
Graphene

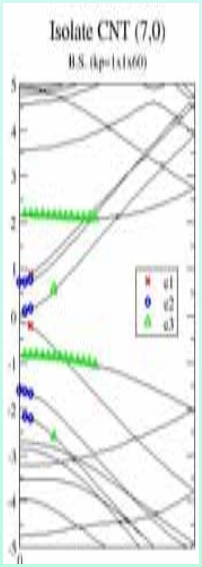


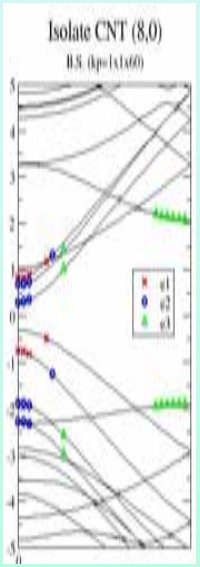
Zigzag Tubes

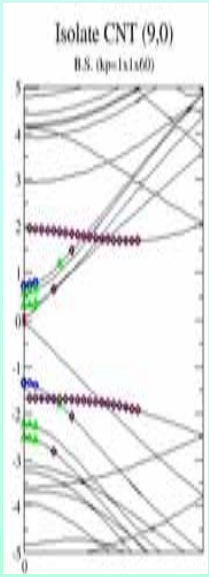


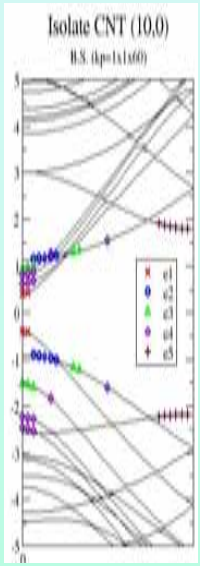


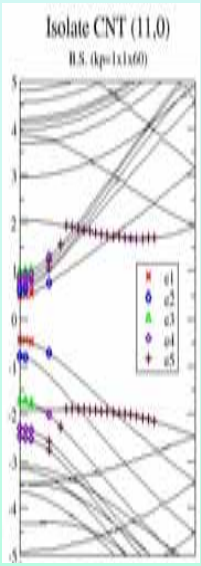


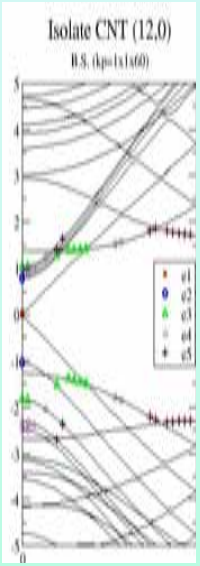


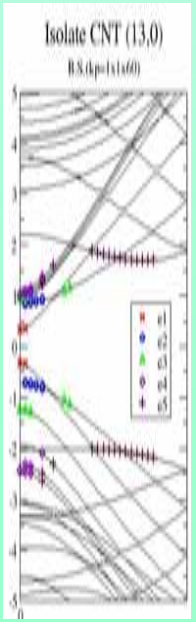


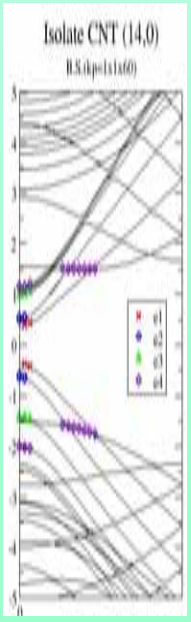


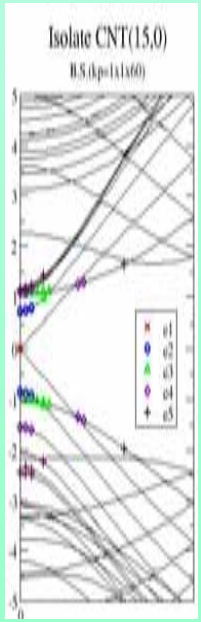


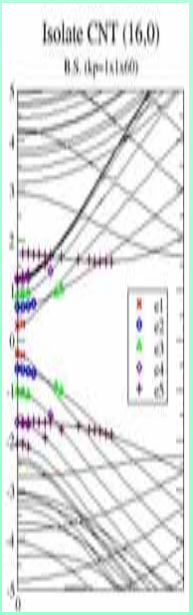


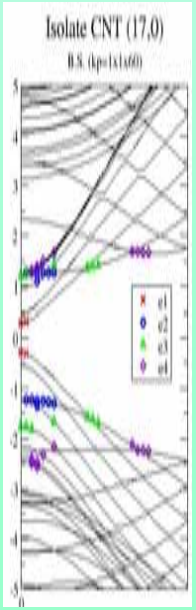


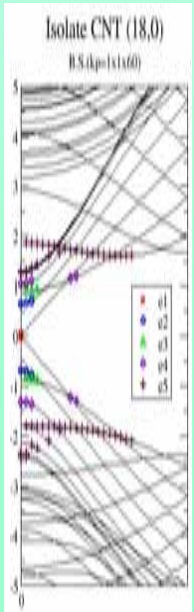


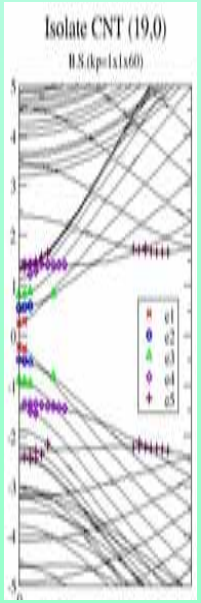


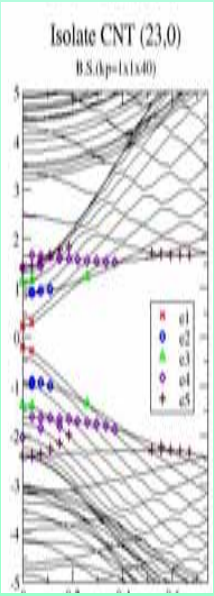




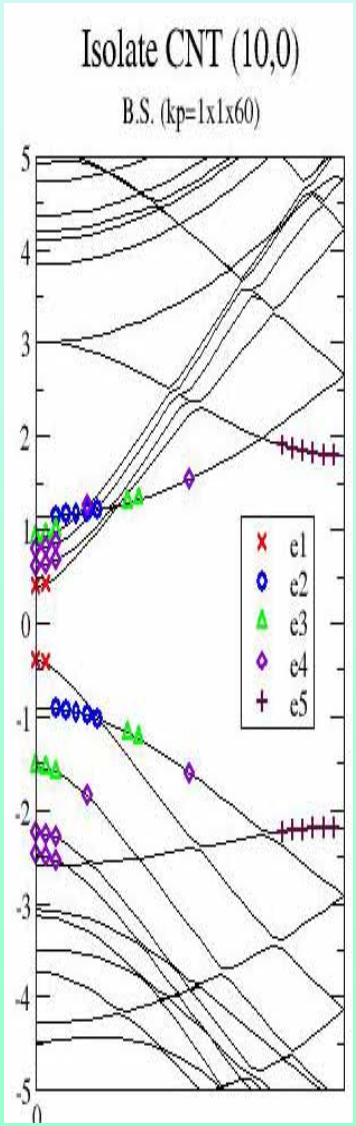


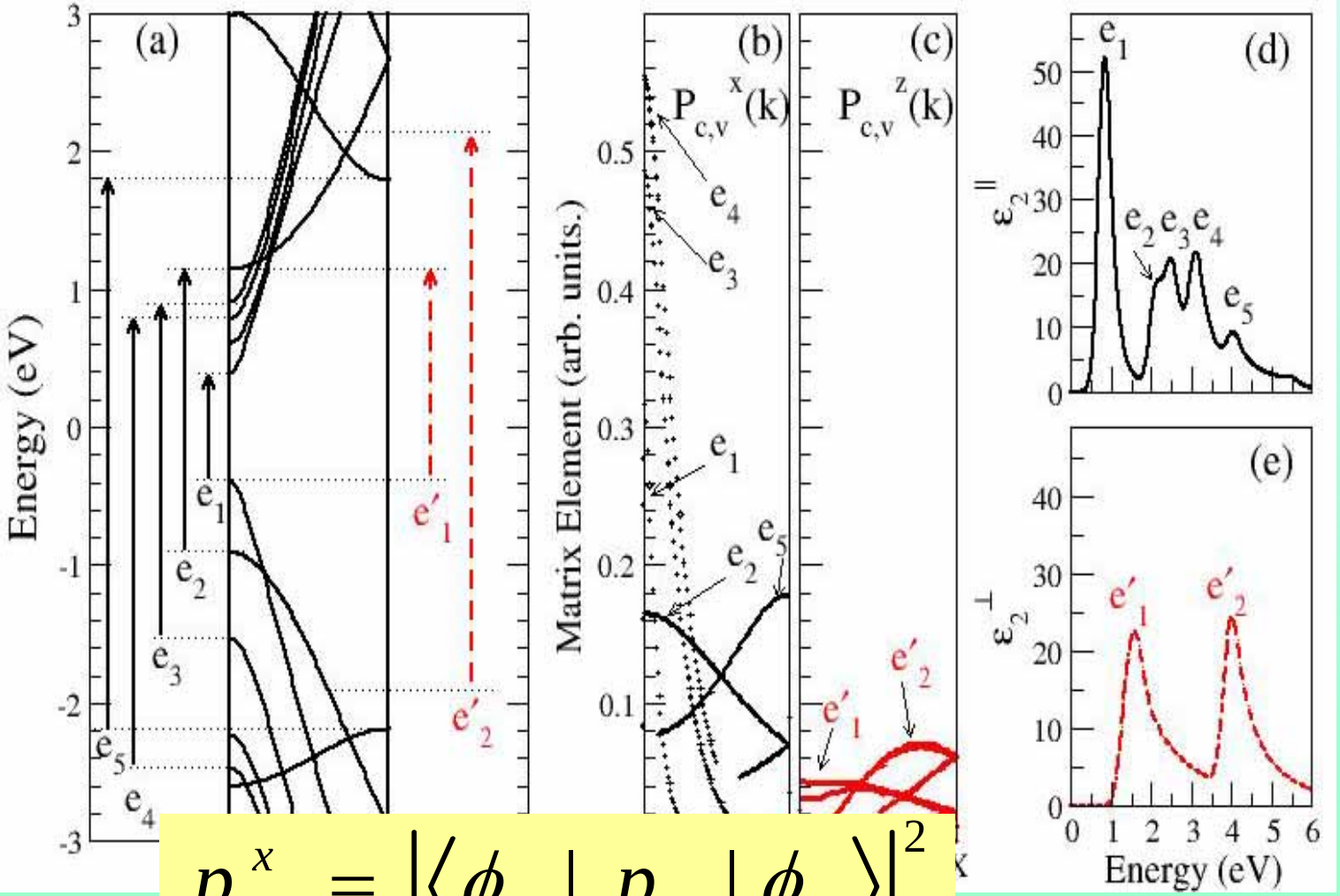






The band structures, matrix elements, and the ε_2 spectrum for (10,0) carbon nanotube

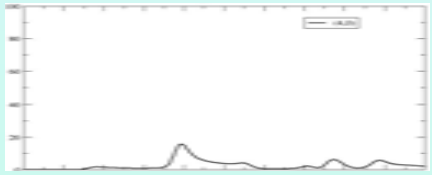




$$p_{cv}^{x} = \left| \langle \phi_c \mid p_x \mid \phi_v \rangle \right|^2$$

ε_2 at zz direction

(4,0)



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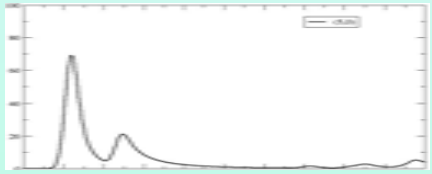

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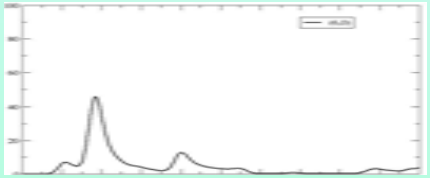

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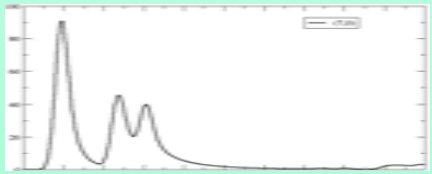

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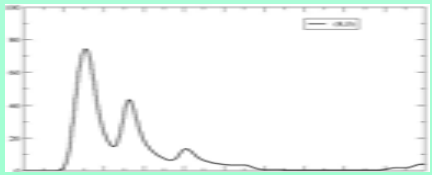

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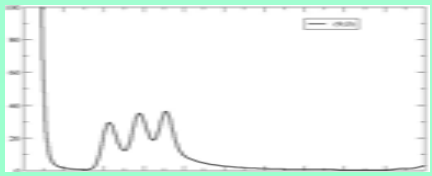

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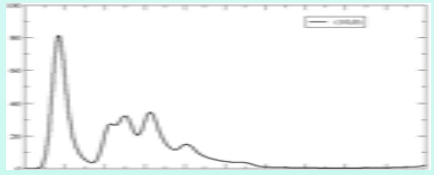

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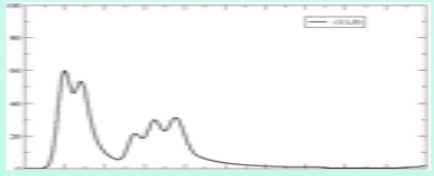

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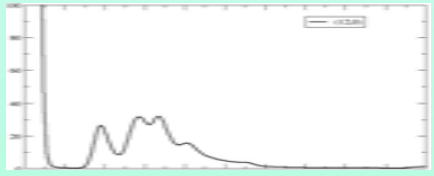

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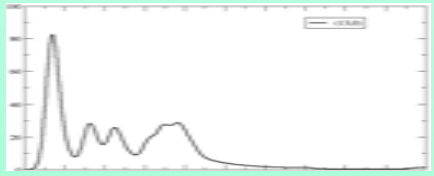

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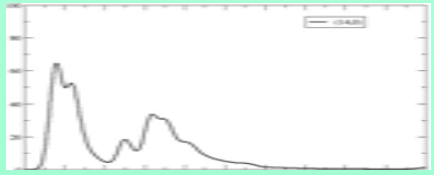

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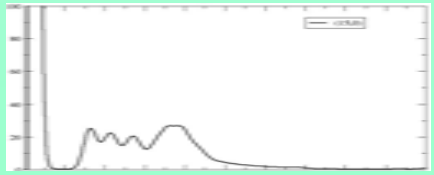

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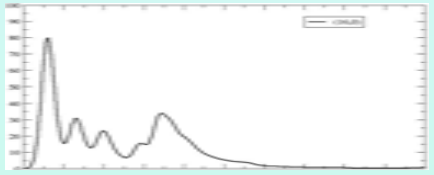

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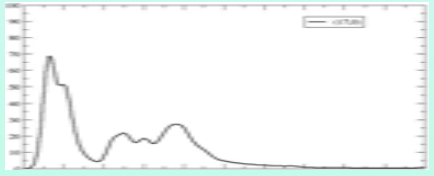

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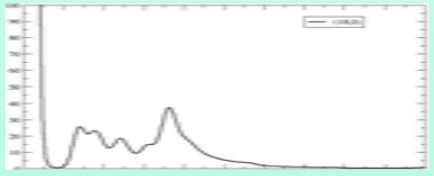

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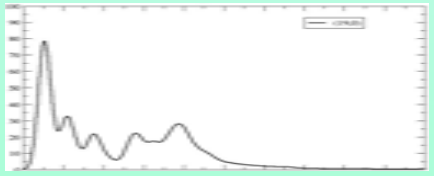

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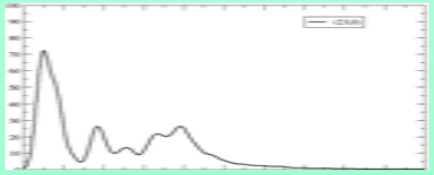
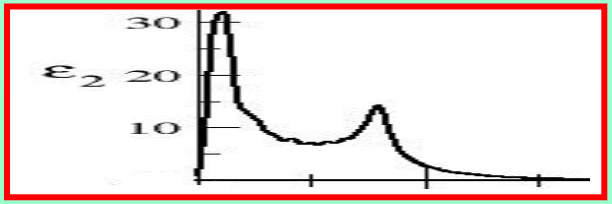
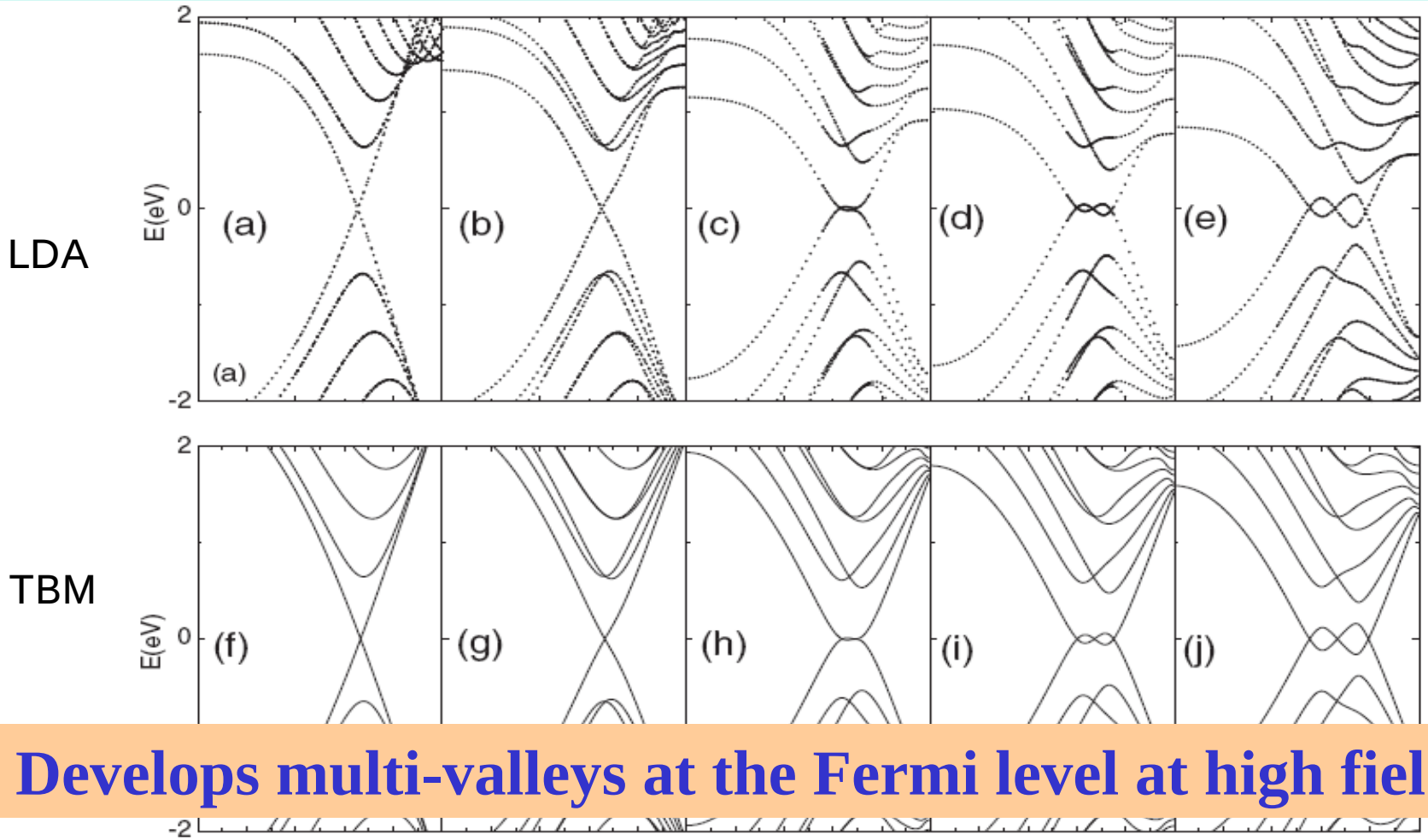


Graphene



Electronic and Optical Properties of SWCNTs under External Transverse Electric Field

Band Structure of (12,12) SWCNT under Different External Field

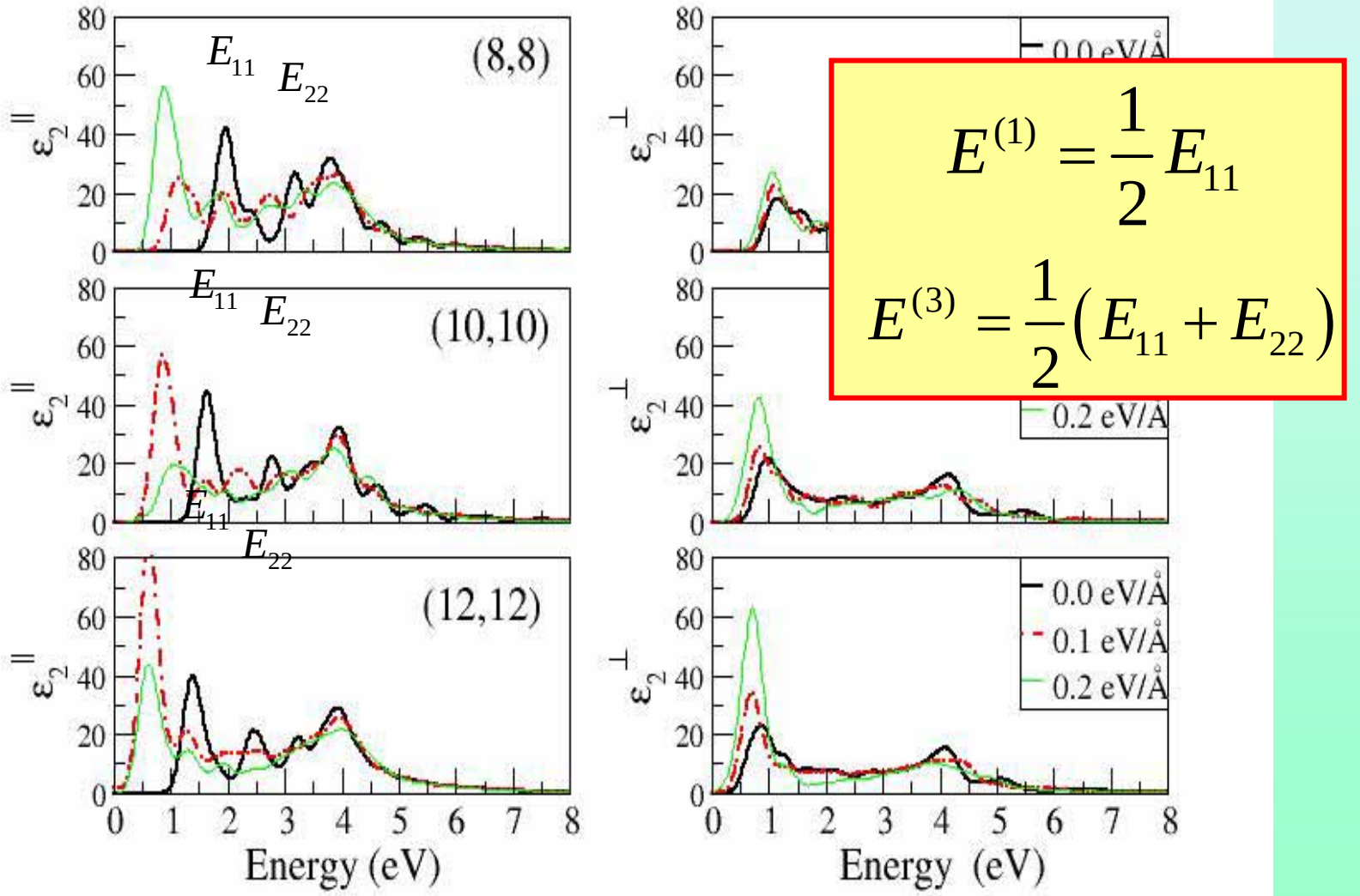


Develops multi-valleys at the Fermi level at high fields

0.0 V / ºA 0.05 V / ºA 0.10 V / ºA 0.12 V / ºA 0.15 V / ºA

Wei Ren, T. H. Cho, T. C. Leung, and C. T. Chan Appl. Phys. Lett. 93, 142102 (2008)

The imaginary part of the dielectric function of armchair nanotubes under different external field

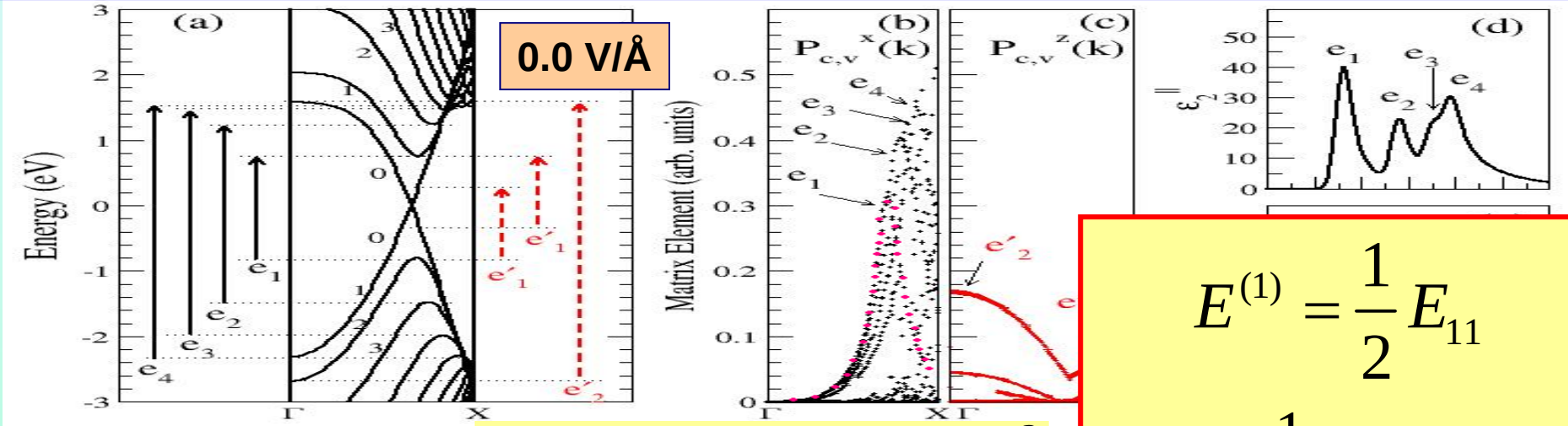


$$E^{(1)} = \frac{1}{2} E_{11}$$

$$E^{(3)} = \frac{1}{2}\left(E_{11} + E_{22}\right)$$

T. H. Cho, W. S. Su, T. C. Leung, Wei Ren, and C. T. Chan Phys. Rev. B 79, 235123 (2009)

The band structures, matrix elements, and the ε_2 spectrum for (10,10) carbon nanotube under different external field

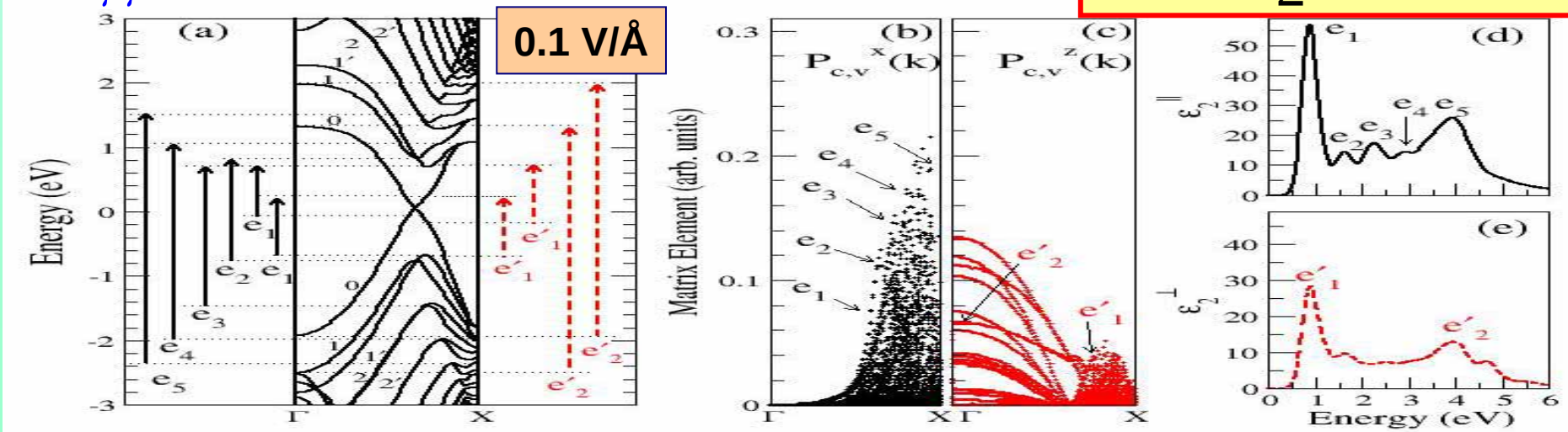


$E_{\mu\mu}$ transition

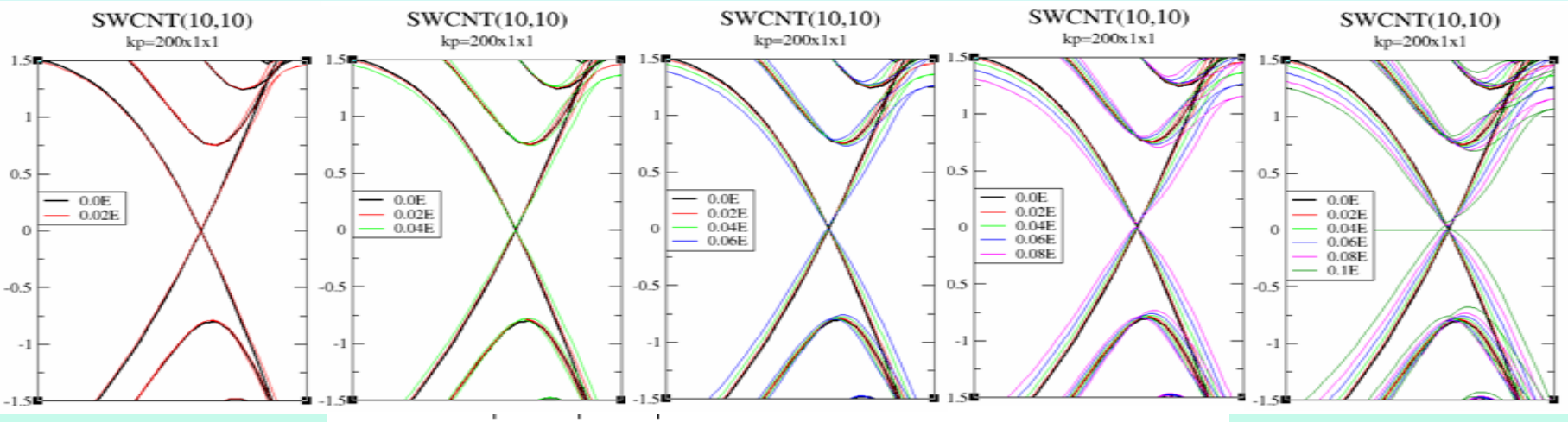
$$p_{cv}^{x} = \left| \langle \phi_c \mid p_x \mid \phi_v \rangle \right|^2$$

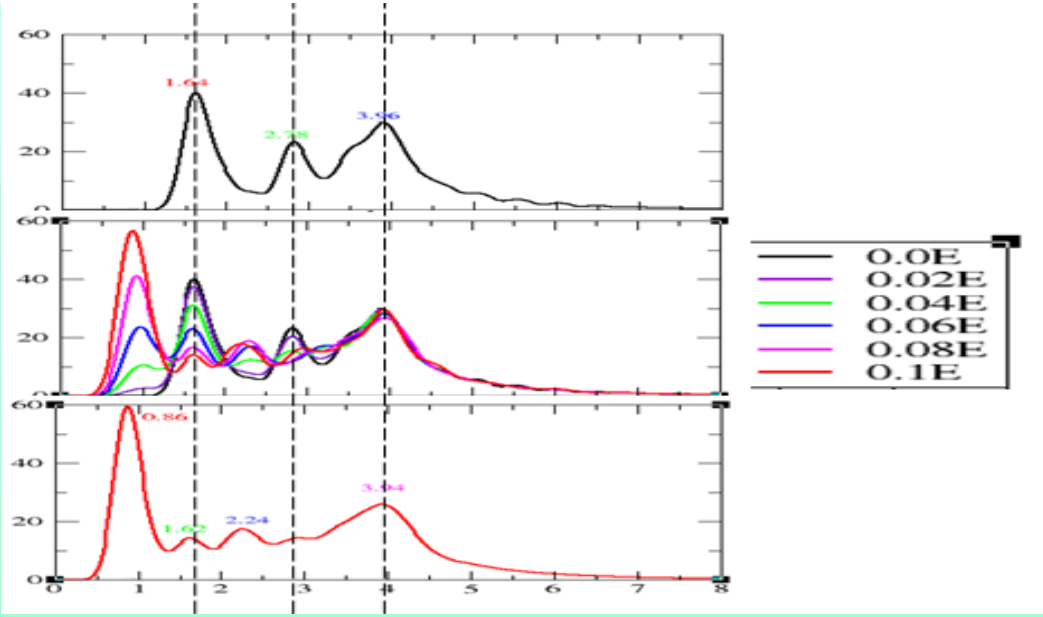
$$E^{(1)} = \frac{1}{2} E_{11}$$

$$E^{(3)} = \frac{1}{2}\left(E_{11} + E_{22} \right)$$



$E_{\mu\mu}$ and $E_{\mu\mu\pm1}$

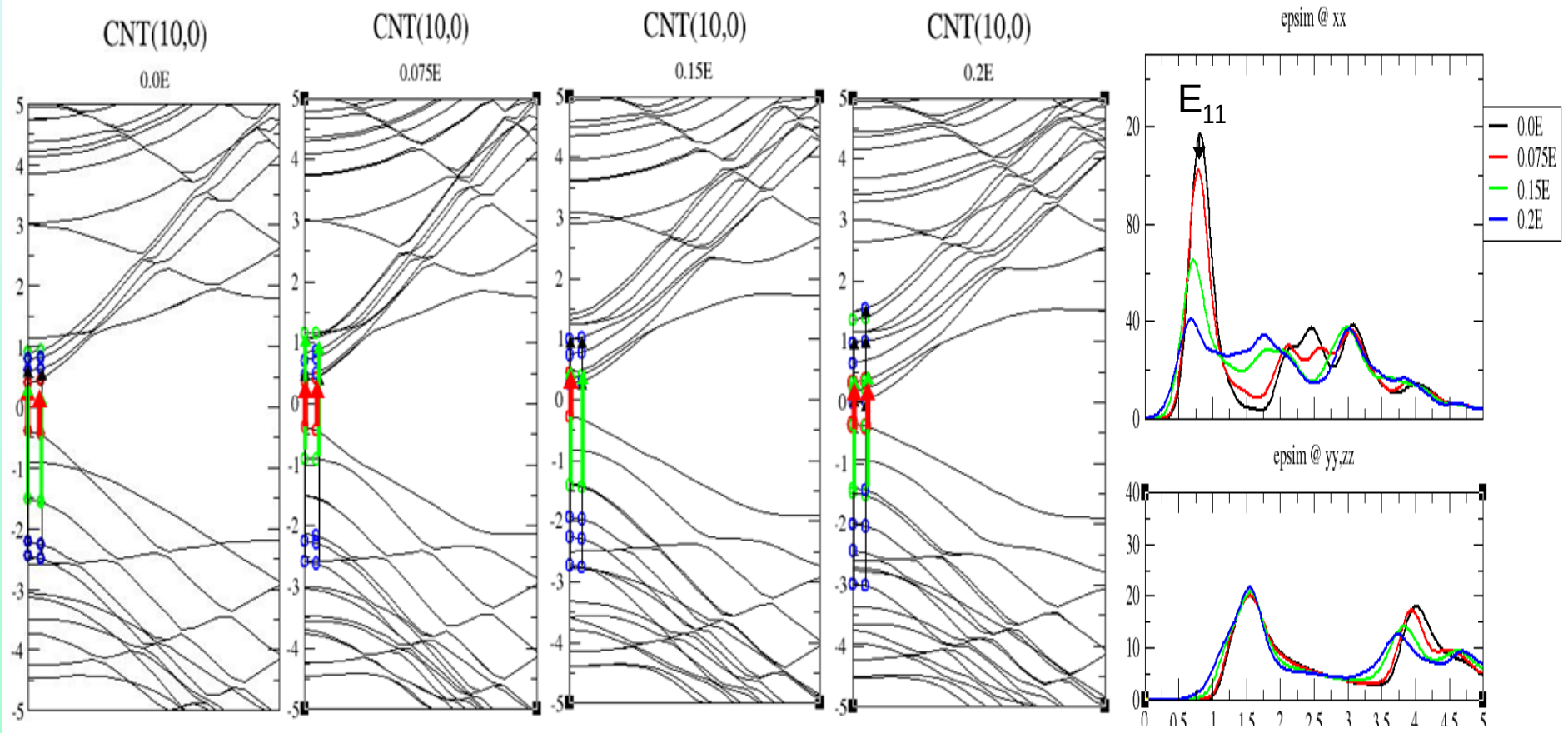




The high sensitivity of the optical spectrum to an applied field make it a plausible material for optoelectronic applications.

Zigzag SWCNTs under External Transverse Electric Field

Band Structure of (10,0) SWCNT under Different External Field



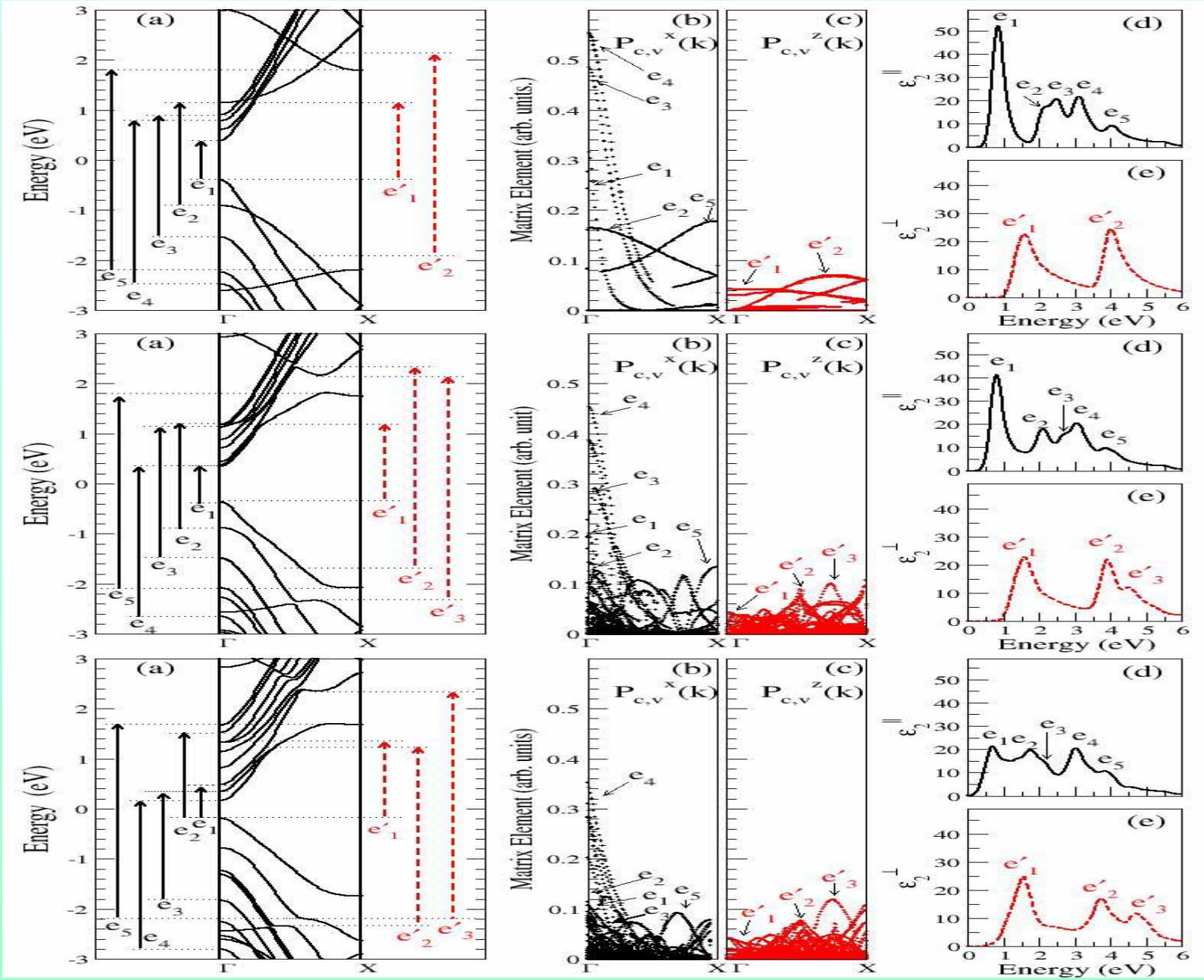
0.0 V / ºA *0.075 V / ºA* *0.15 V / ºA* *0.20 V / ºA*

The gap is reduced significantly; however, the effect of the electric field on the dielectric function is less pronounced.

0.0 V/Å

0.1 V/Å

0.2 V/Å



The band structures, matrix elements, and the ε_2 spectrum for (10,0) carbon nanotube under different external field.

The imaginary part of the dielectric function of zigzag nanotubes under different external field

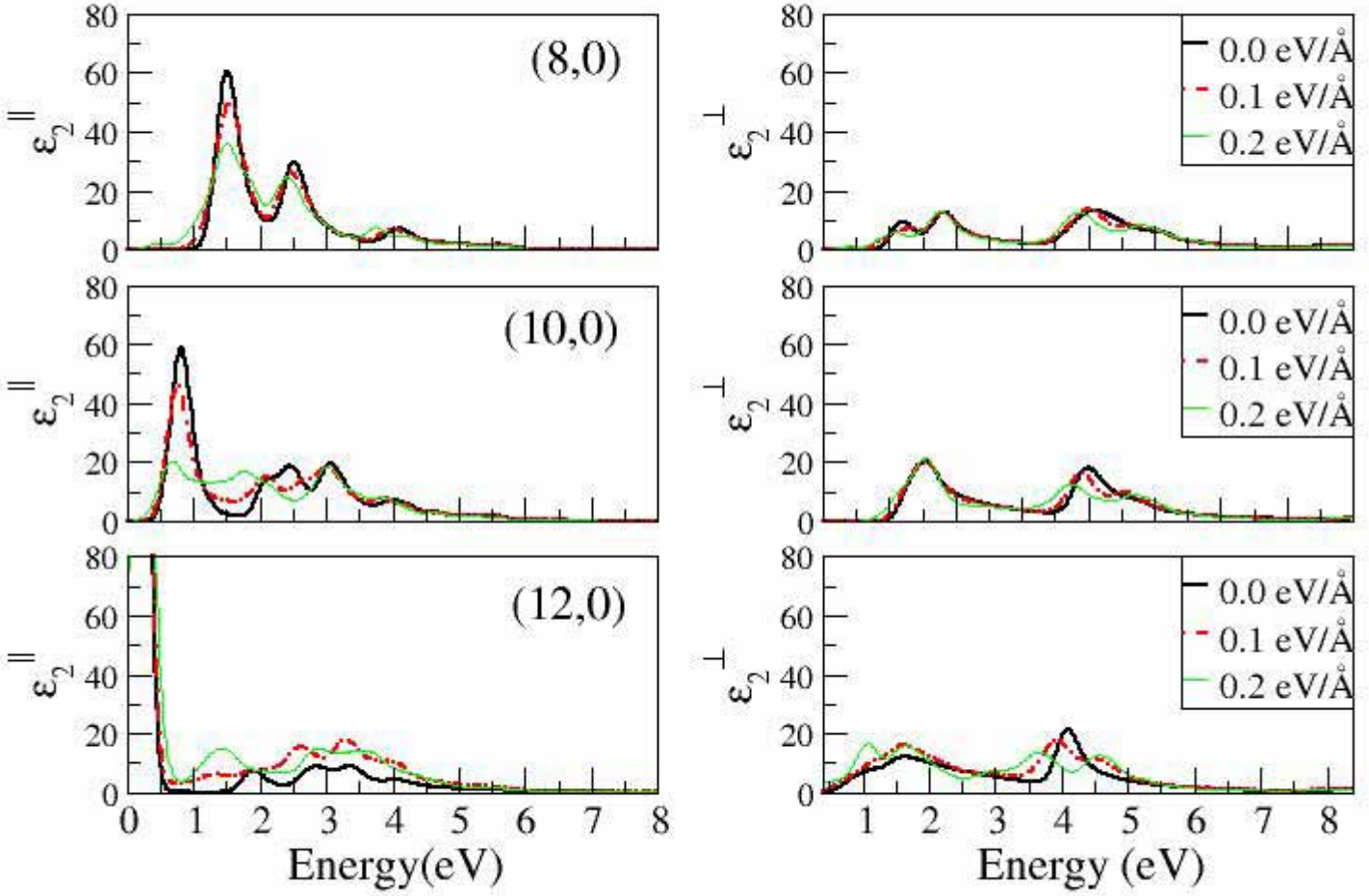
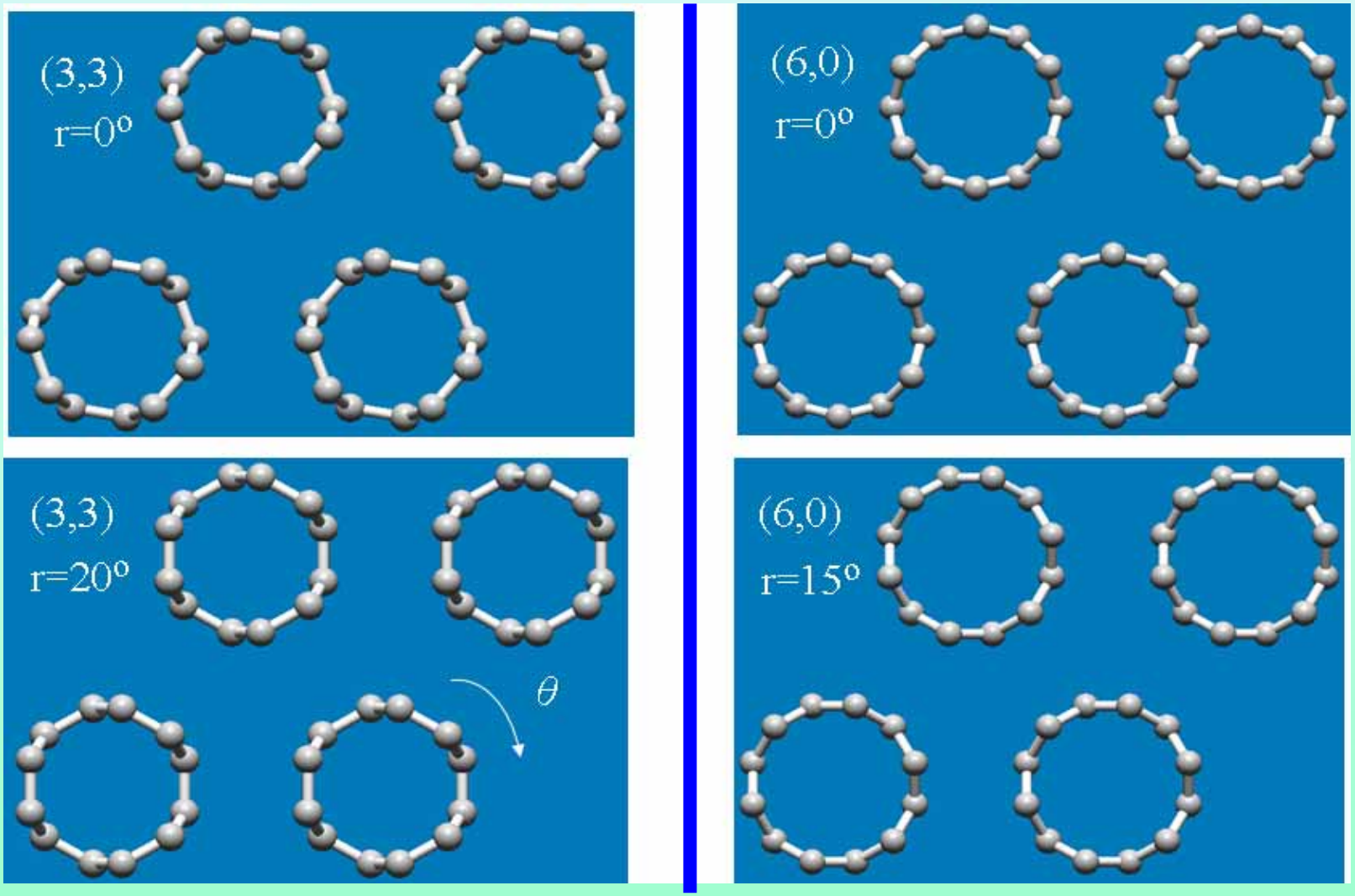
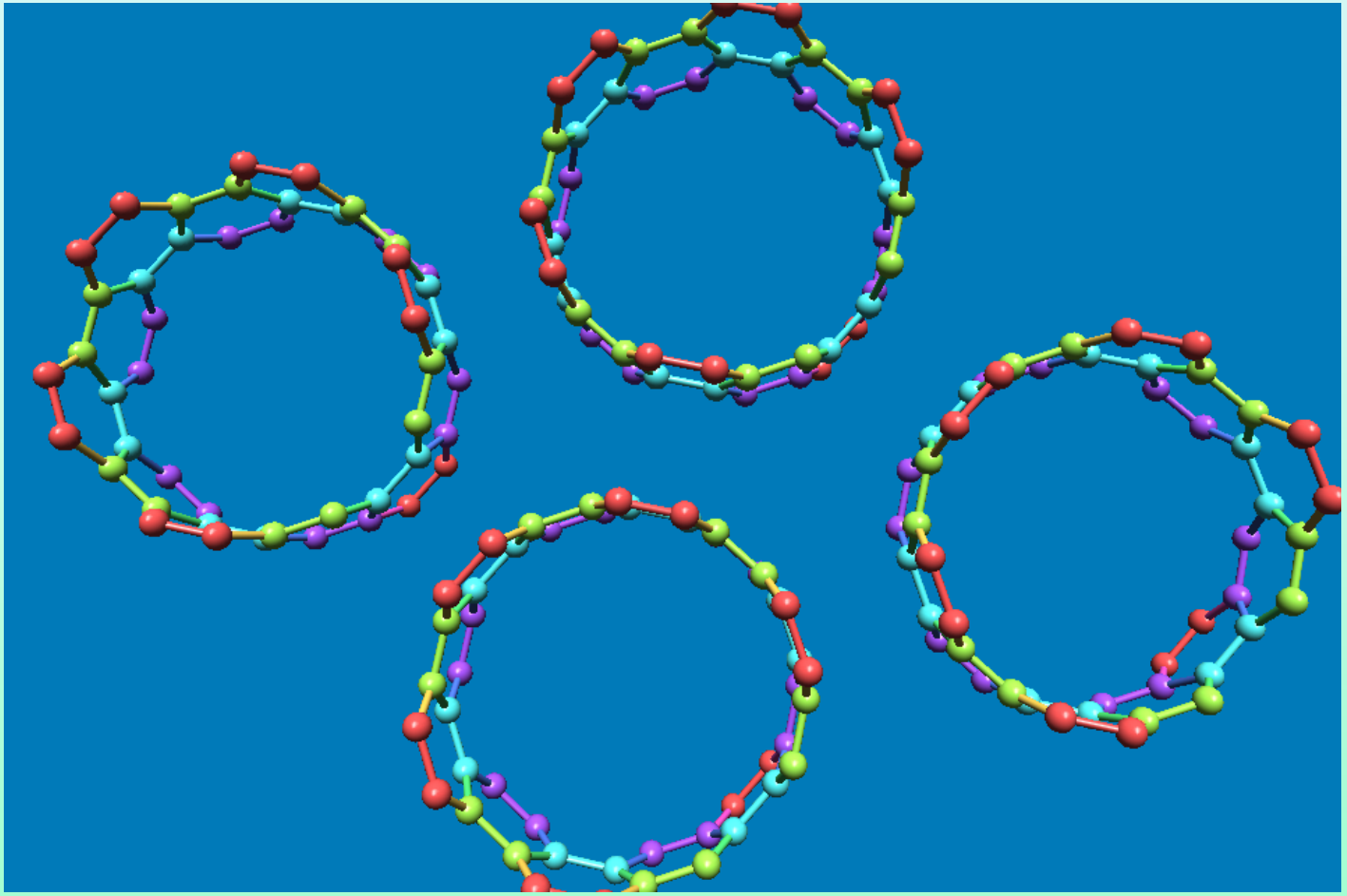


FIG. 8: (Color online) The imaginary part of the dielectric function of zigzag nanotubes for light polarized parallel (left panel) and perpendicular (right panel) to the nanotubes axis under external electric field $\vec{E}$ of 0.0, 0.1, and 0.2 V/$\mathring{A}$.

Electronic and Optical Properties of SWCNT Bundles

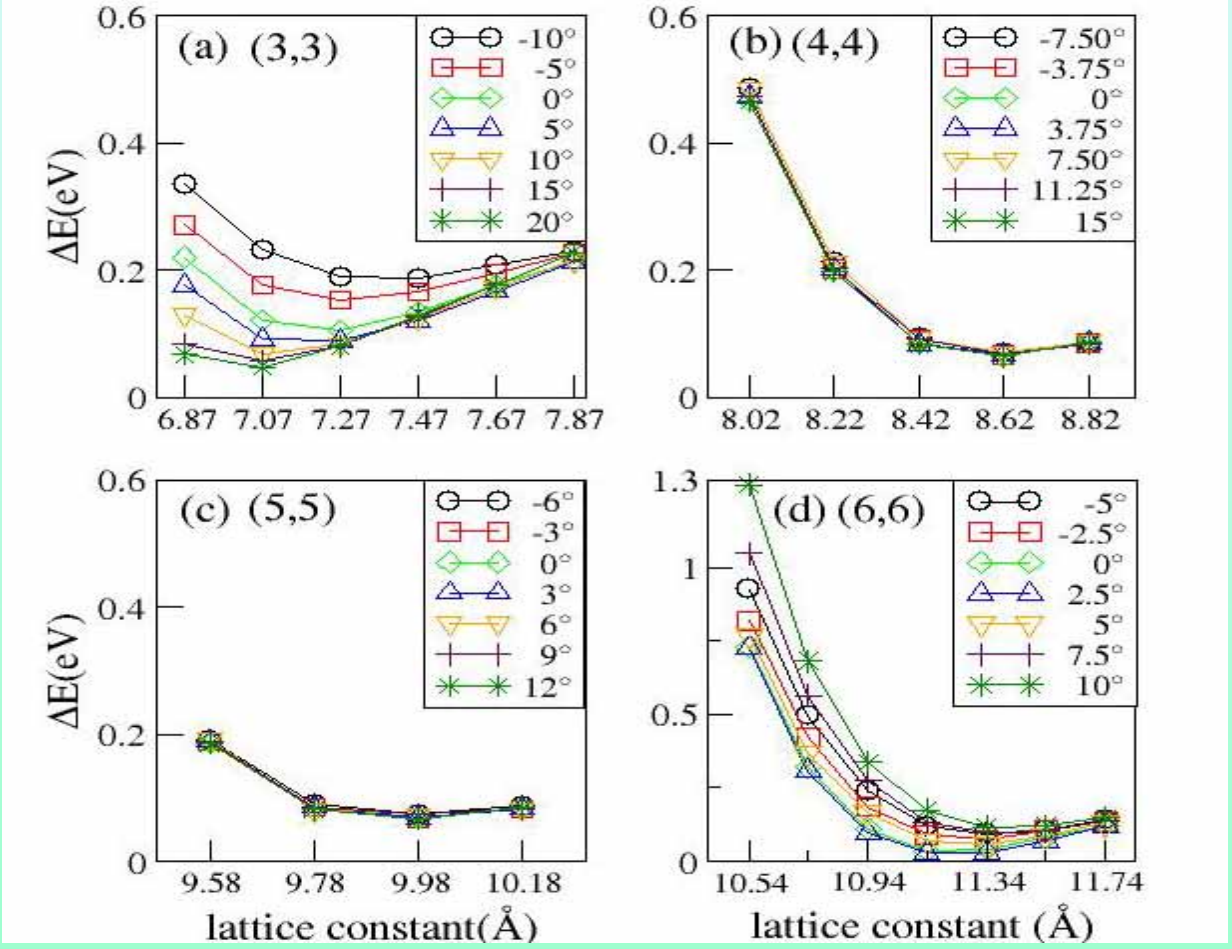


Schematic of the nanotubes in (a) (3,3) bundle with rotation 0°, (b) the (3,3) bundle with rotation 20°, (c) (6,0) bundle with rotation 0°, and (d) (6,0) bundle with rotation 15°.



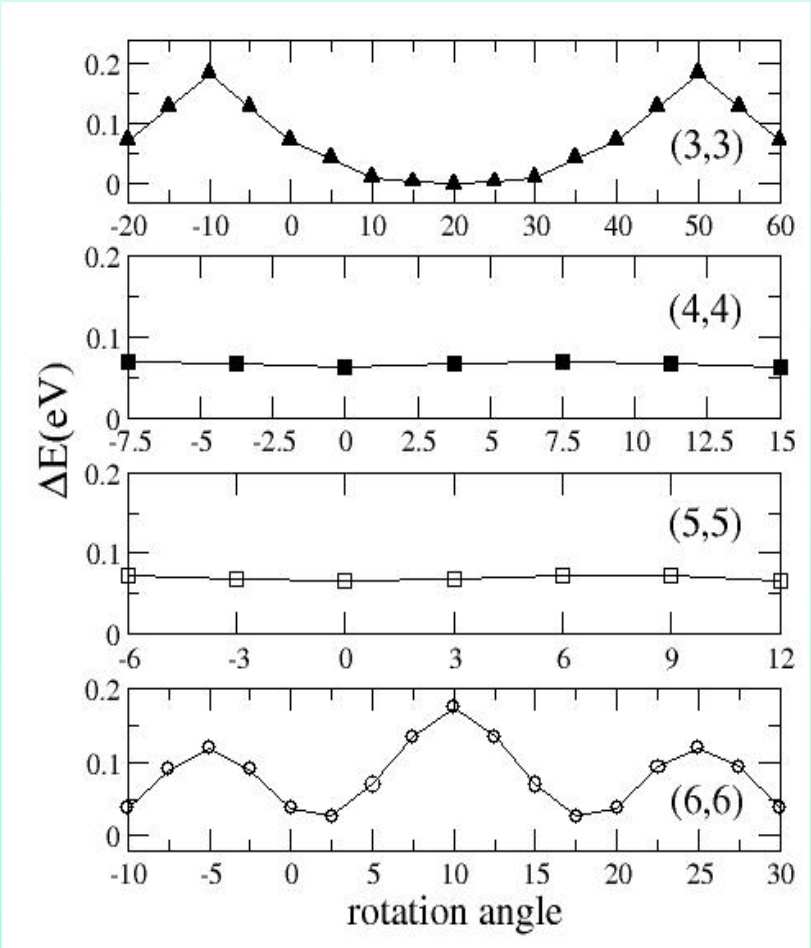
The tubes are arranged in a close-packing array

The total energy *vs* tube-tube distance with different rotational angles for the (3,3) , (4,4) , (5,5) , and (6,6) bundles



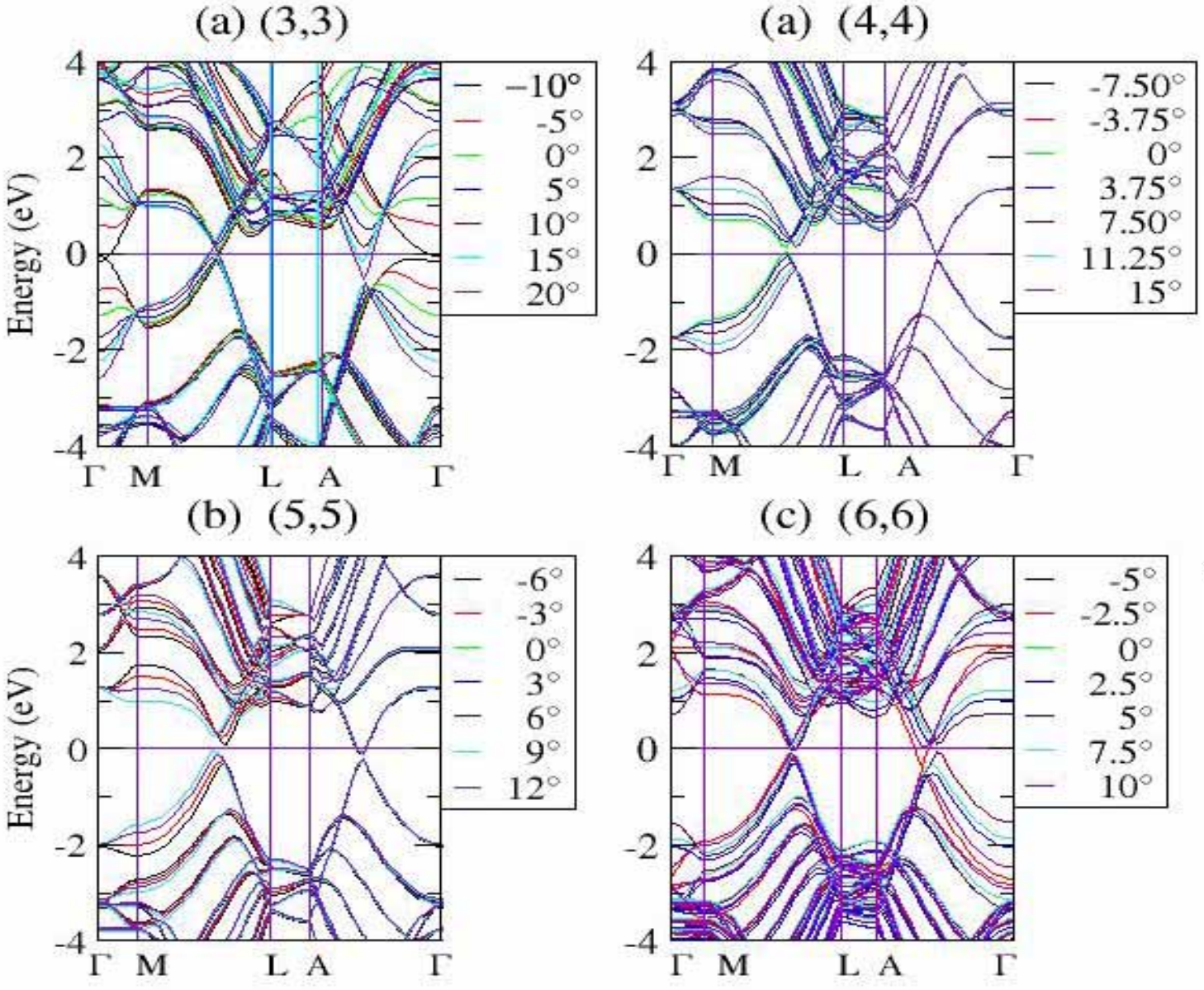
The total energy show strong dependence on the orientation for the n = 3q (n,n) bundles, while those of the n ≠ 3q bundles are nearly independent of such orientation.

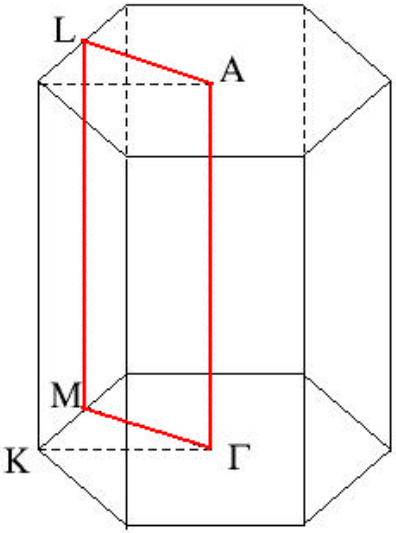
Total energies vs. the rotational angle for the (3,3) , (4,4) , (5,5) and (6,6) bundles



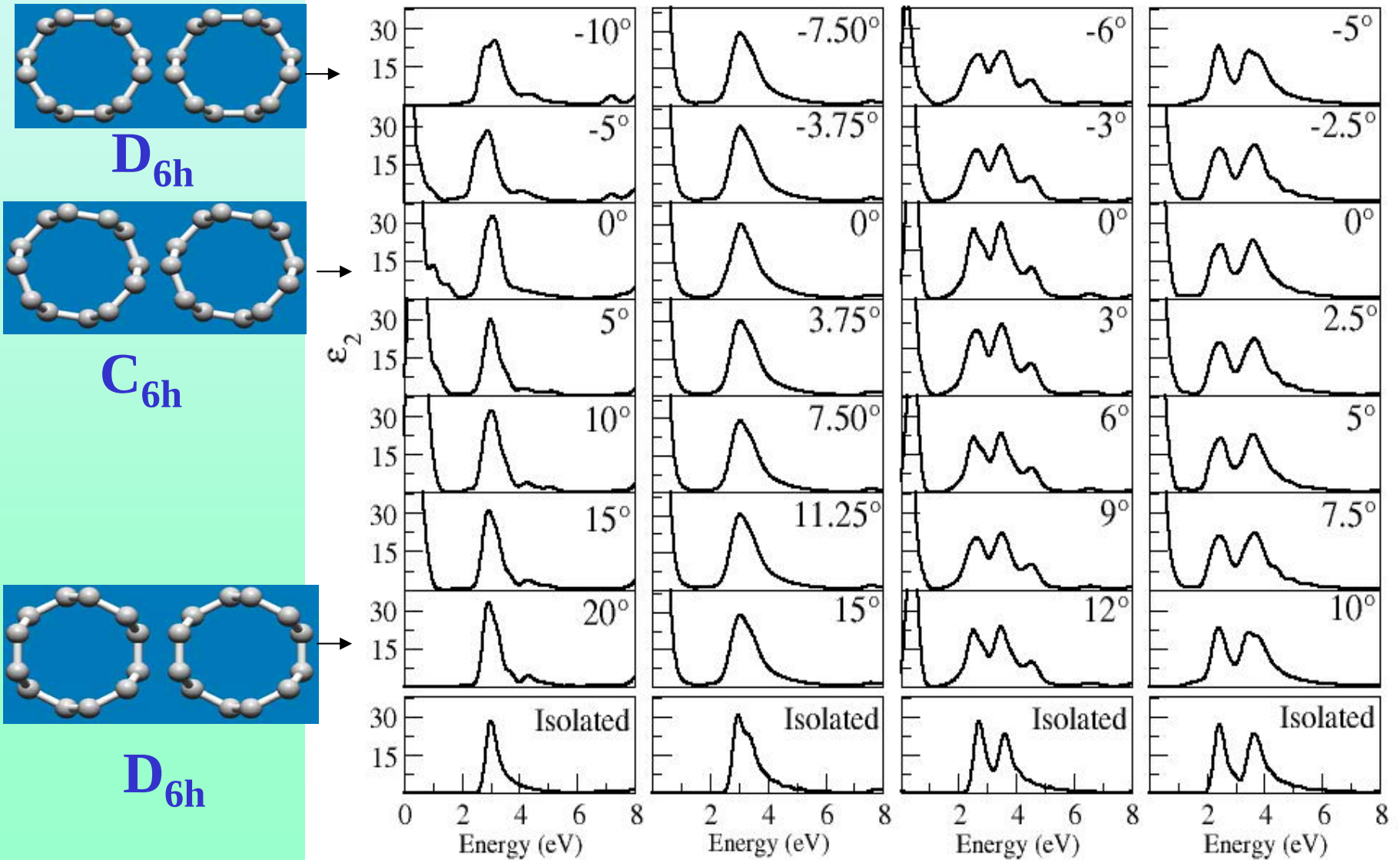
The total energy show strong dependence on the orientation for the n = 3q (n,n) bundles, while those of the n ≠ 3q bundles are nearly independent of such orientation.

The band structure of the (3,3), (4,4), (5,5), and (6,6) bundles for the different orientations

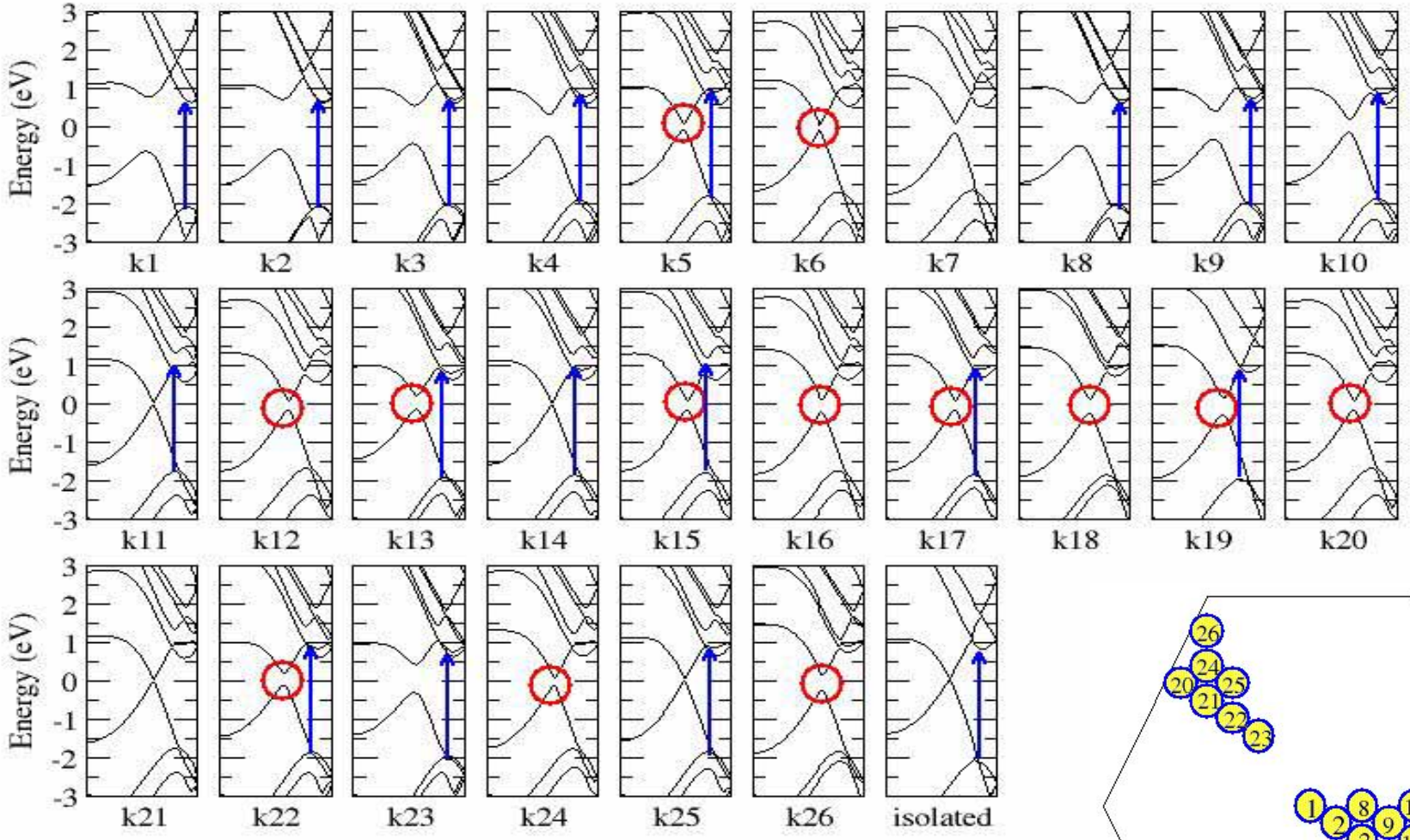


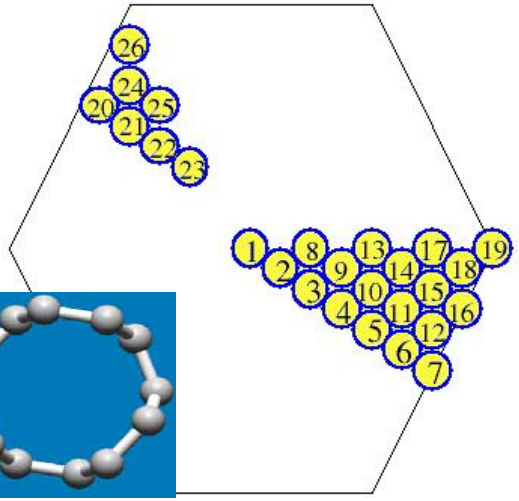


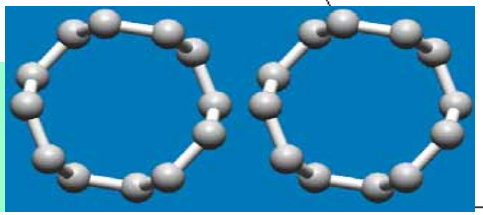
The spectrum of the isolated and bundled (3,3), (4,4), (5,5) and (6,6) SWCNTs for different rotational angles



The band structure along the k_z direction for different k_n points of the (3,3) bundles with rotational angle 0°

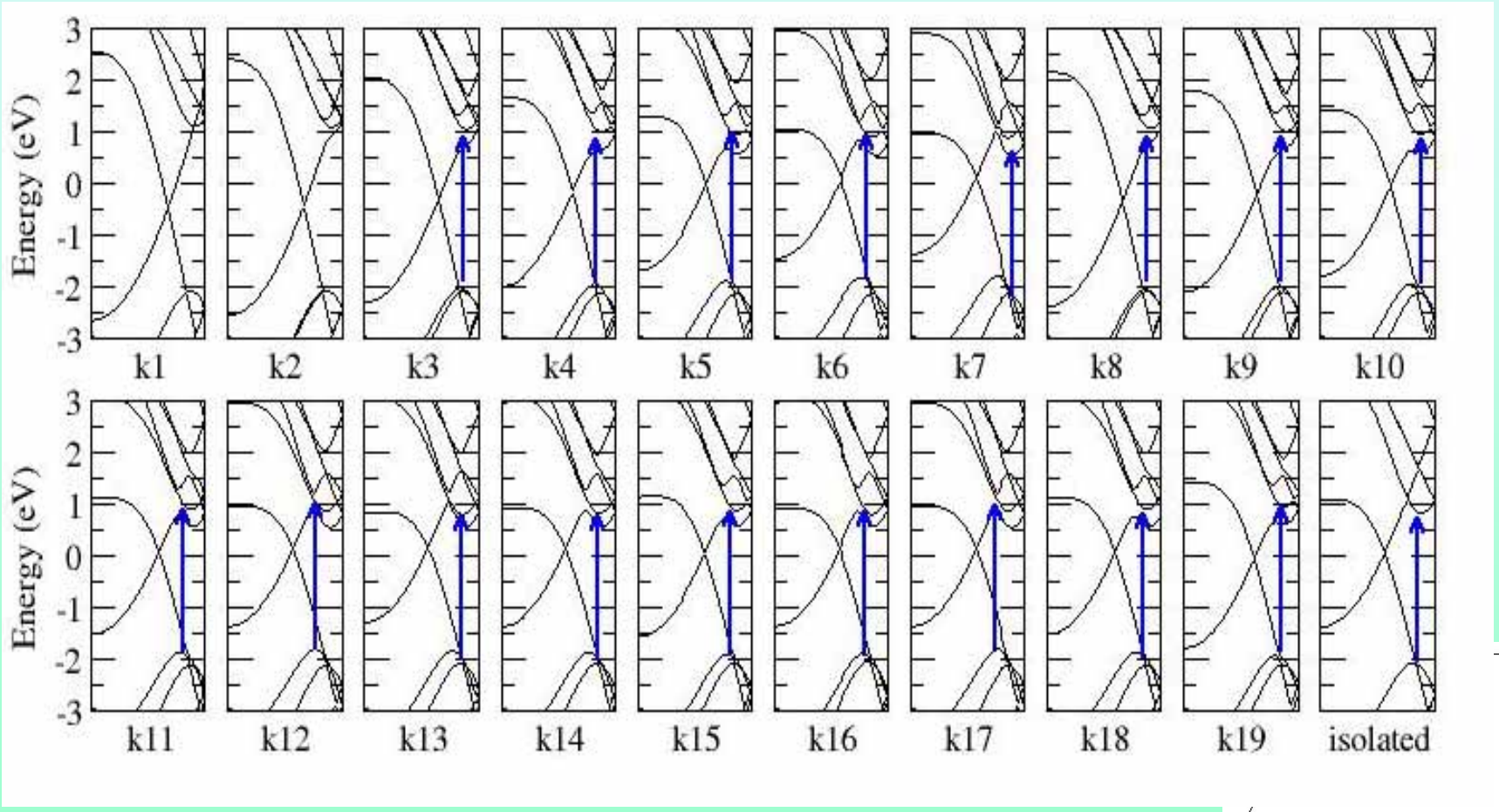




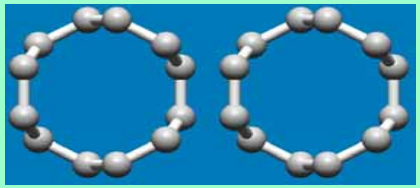


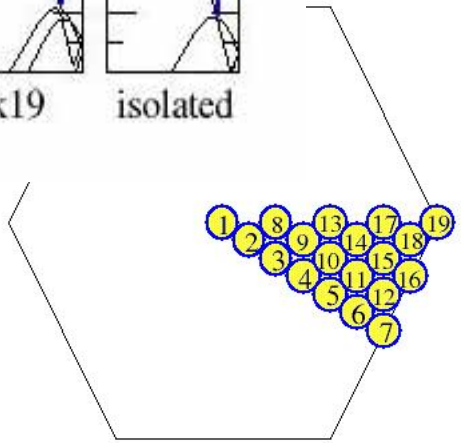
C_{6h}

The band structure along the k_z direction for different k_n points of the (3,3) bundles with rotational angle 20°



D_{6h}

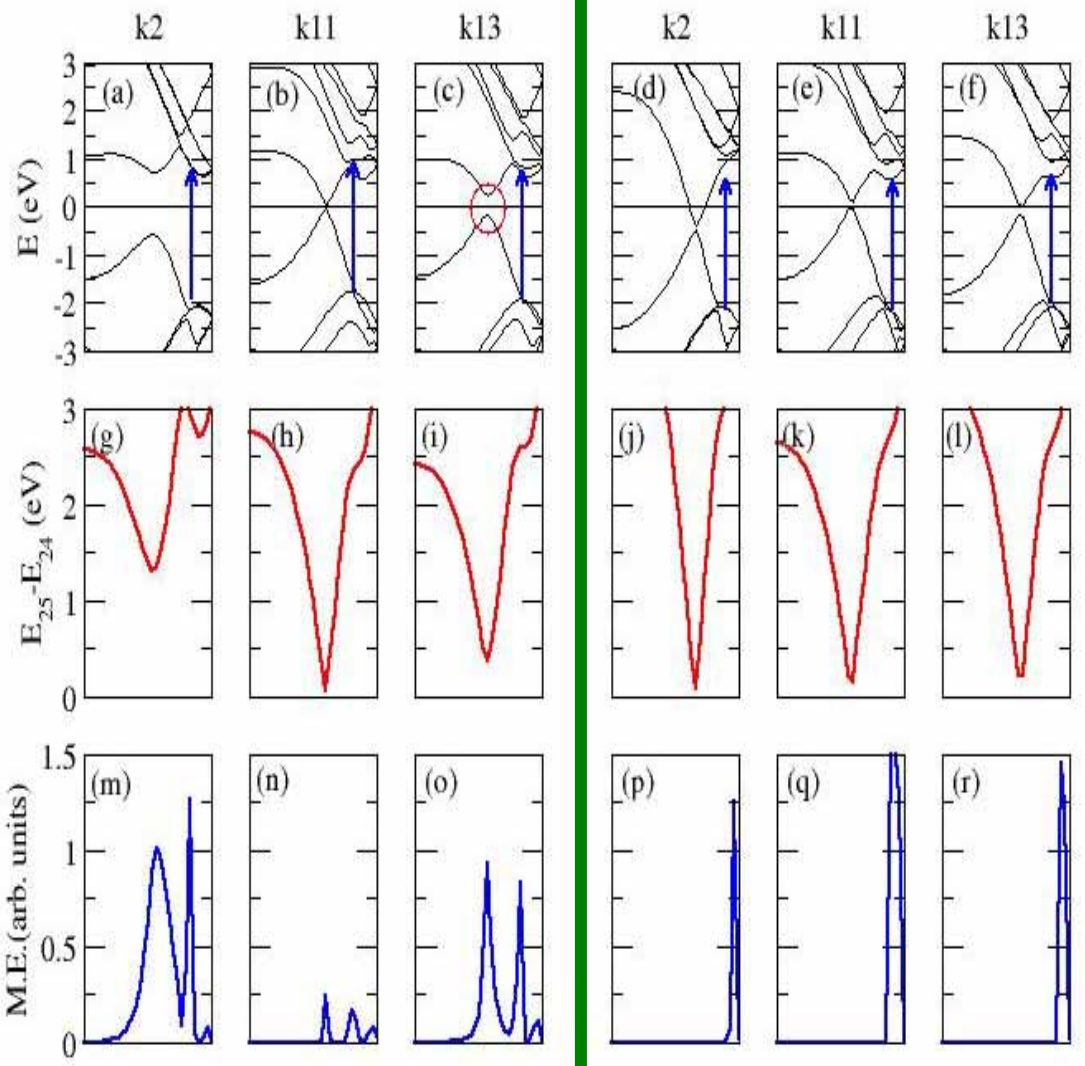


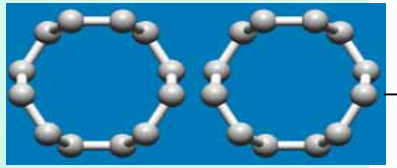


(3,3) bundles for k_2, k_{11}, and k_{13}

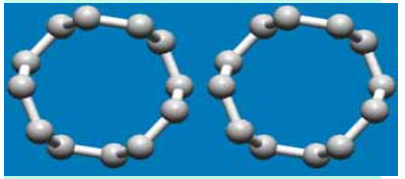
angle 0°

angle 20°

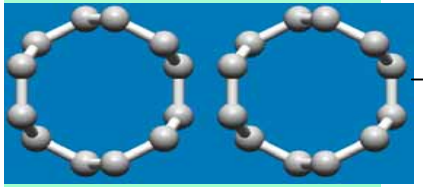




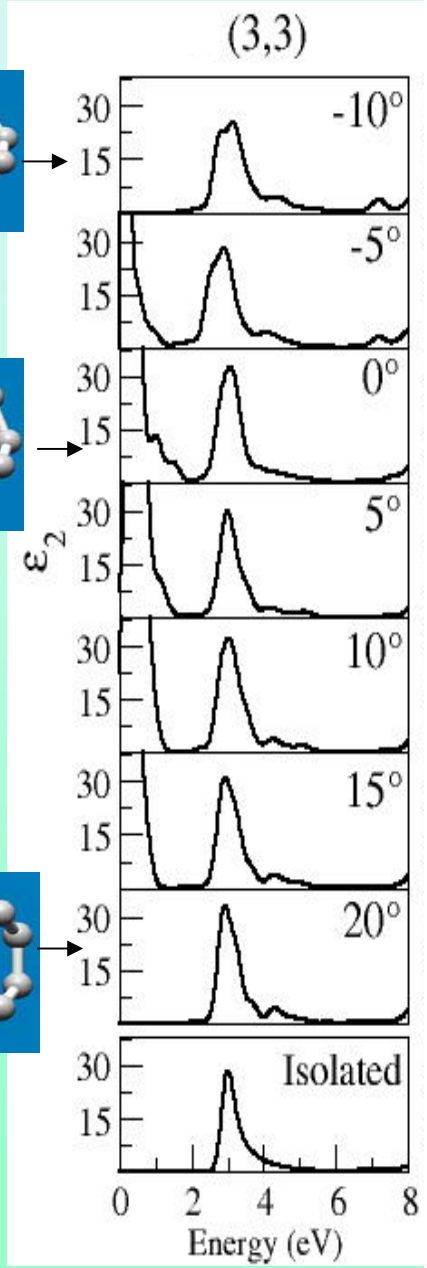
D_{6h}



C_{6h}

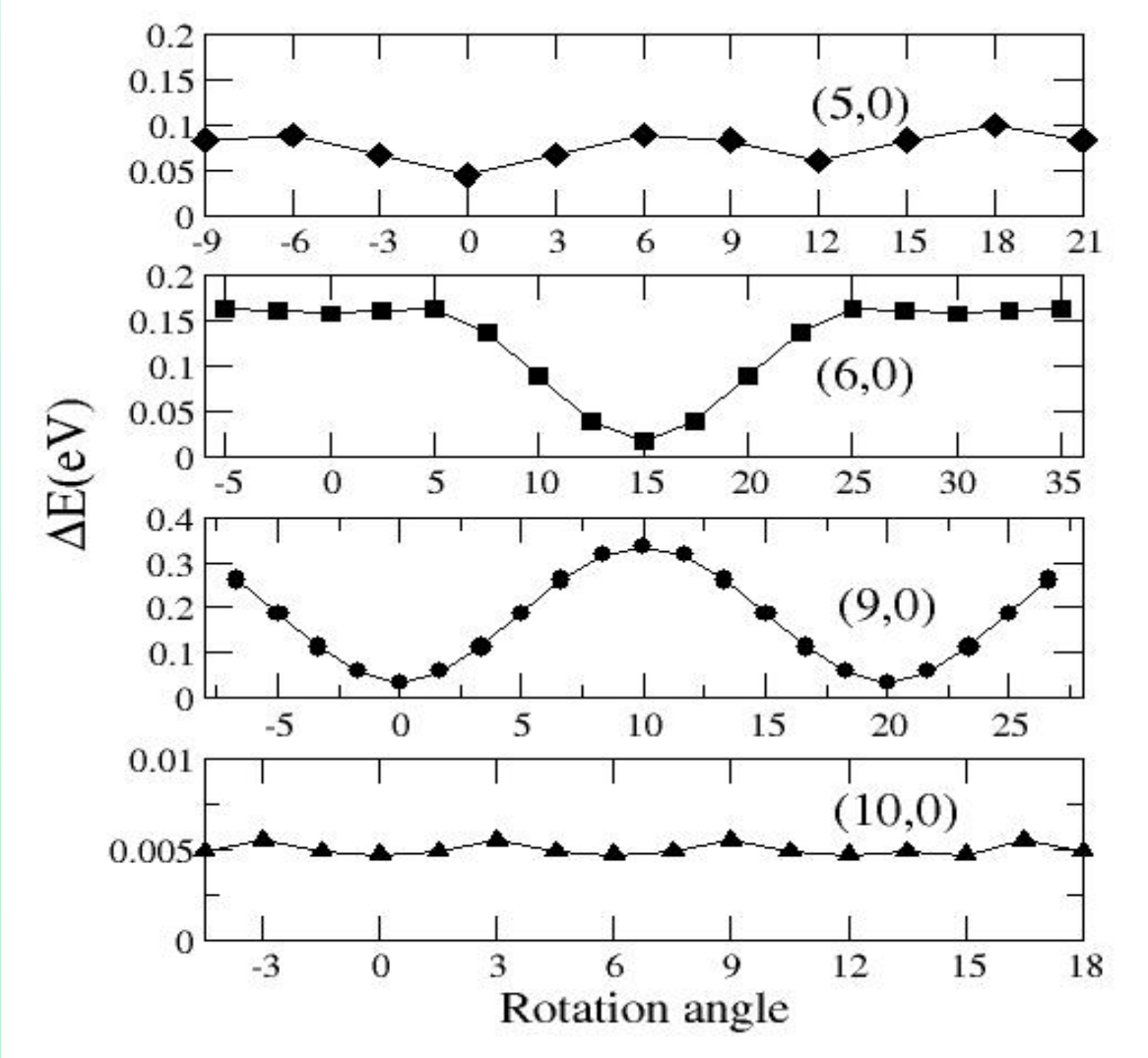


D_{6h}

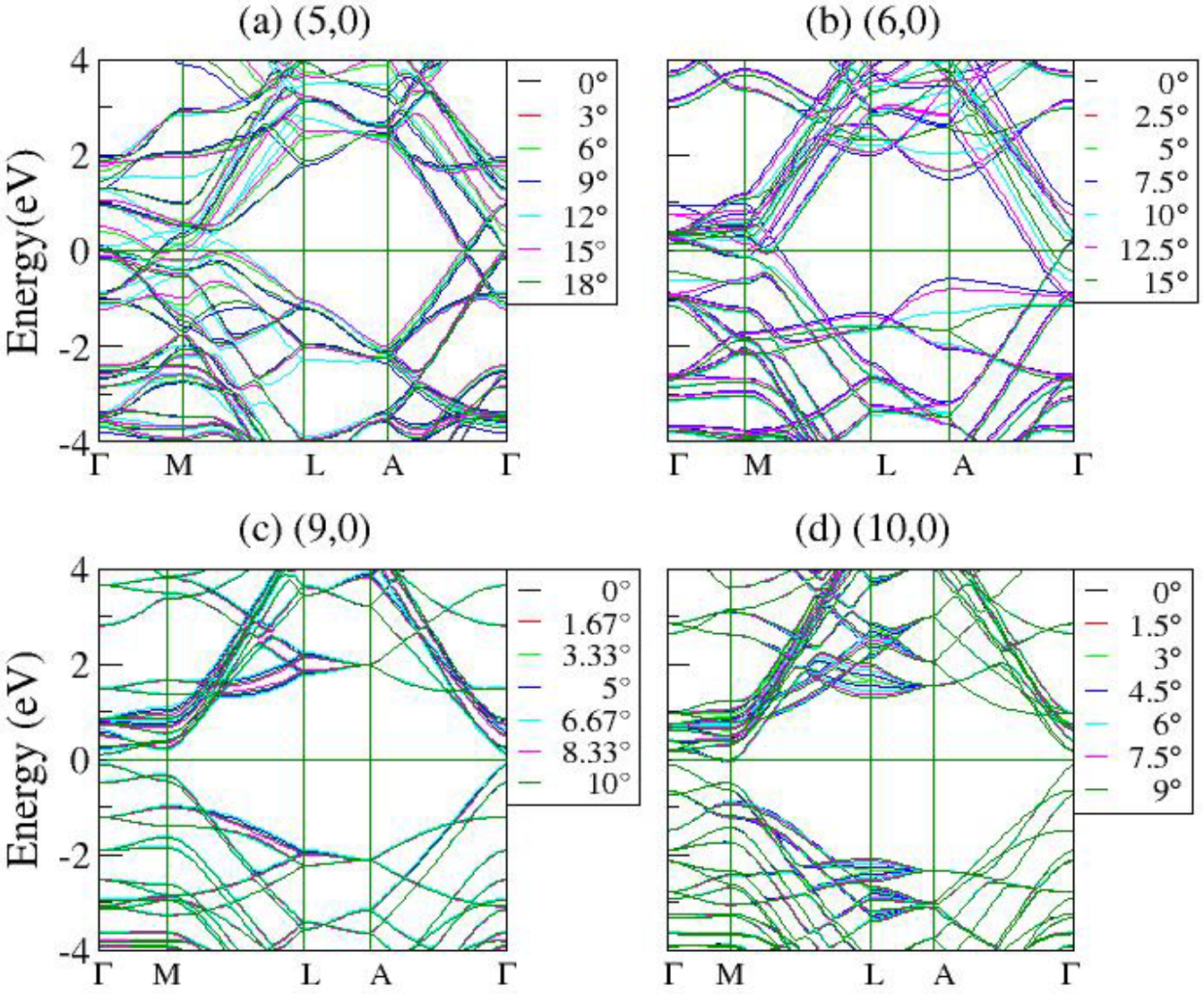


Zigzag SWCNT Bundles

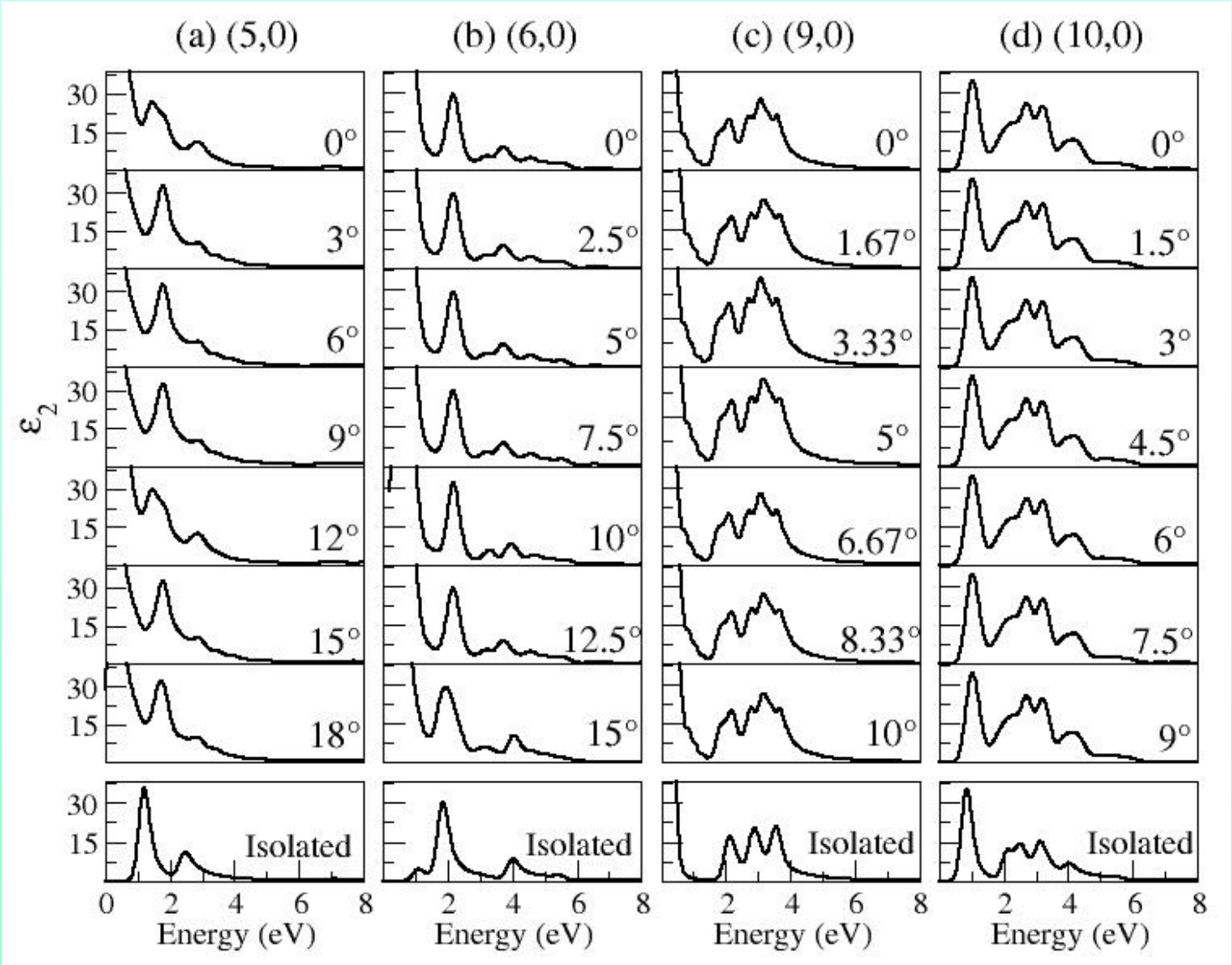
Total energies vs. the rotational angle for the (5,0) , (6,0) , (9,0) and (10,0) bundles



The band structure of the (5,0), (6,0), (9,0), and (10,0) bundles for the different orientations

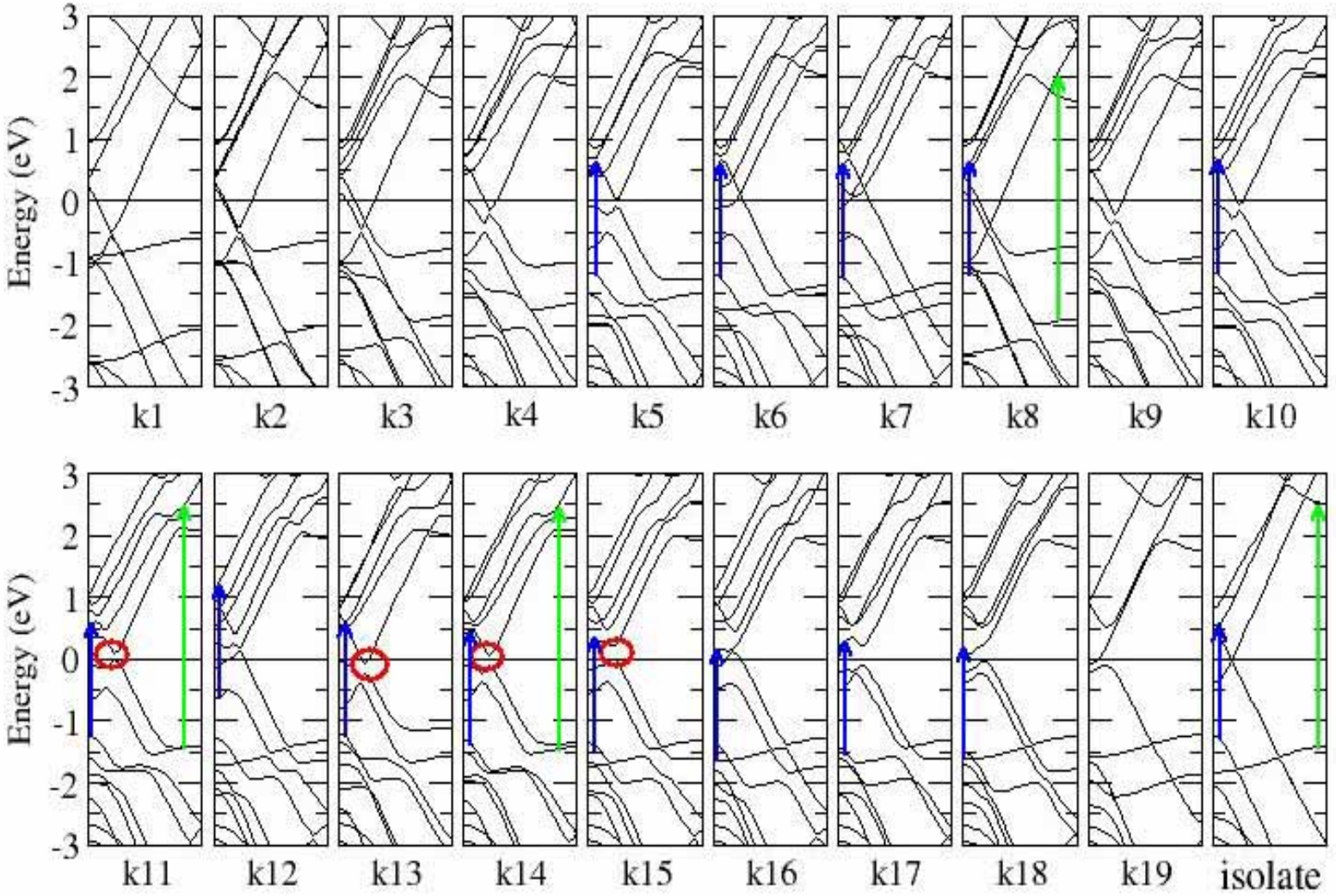


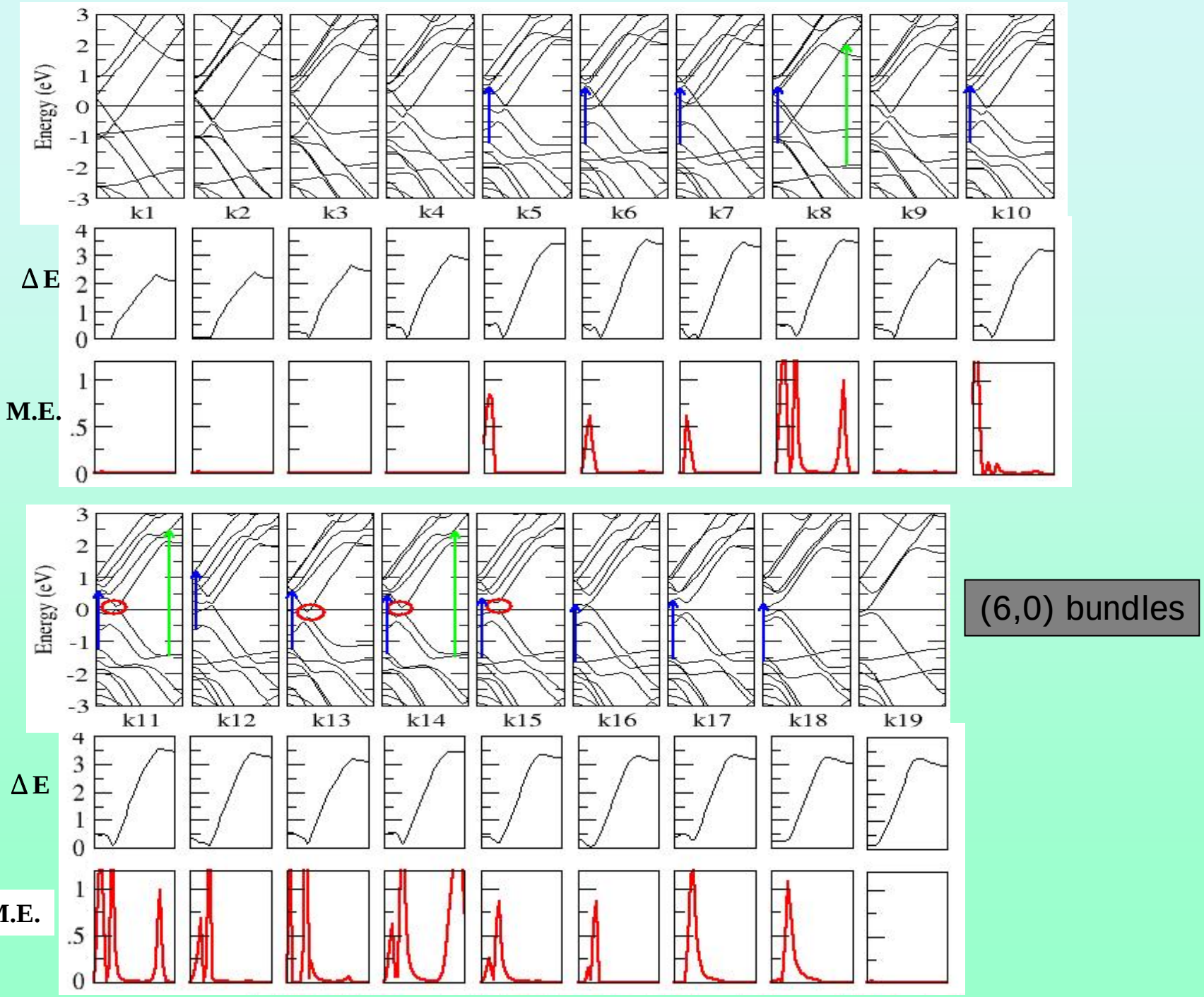
The spectrum of the isolated and bundled (5,0), (6,0), (9,0) and (10,0) SWCNTs for different rotational angles



Bundled CNT (6,0)

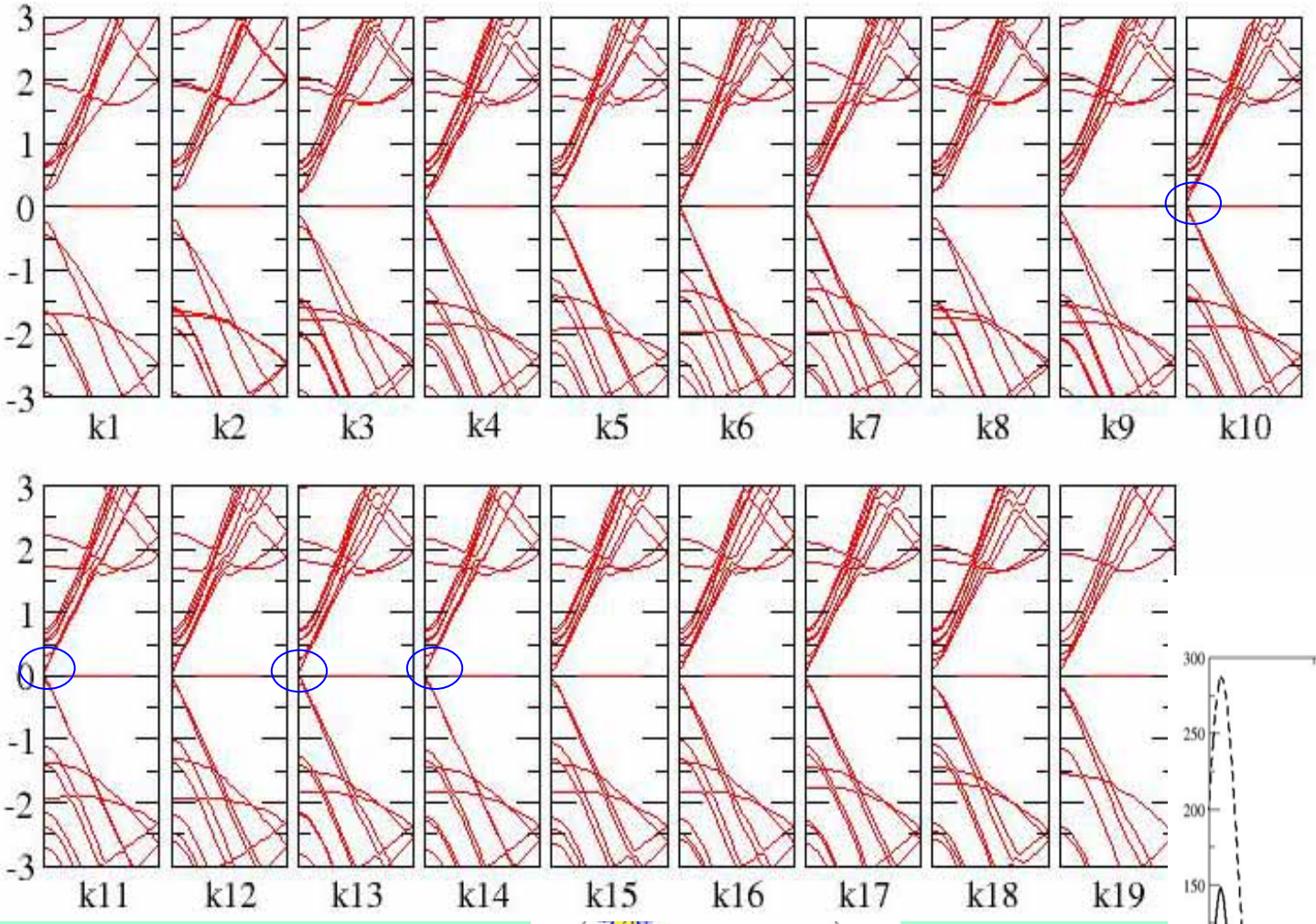
The band structure along the k_z direction for different k_n points of the (6,0) bundles with rotational angle 0^o

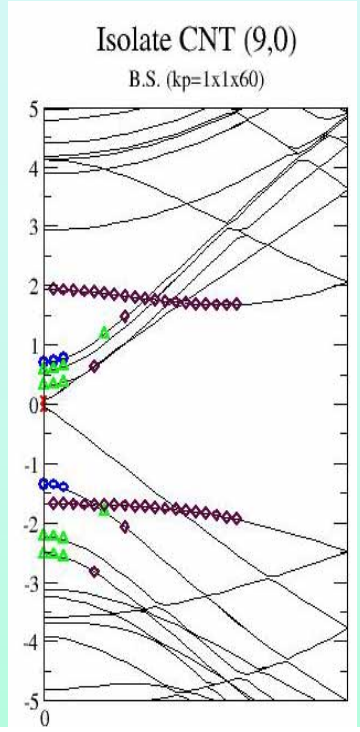


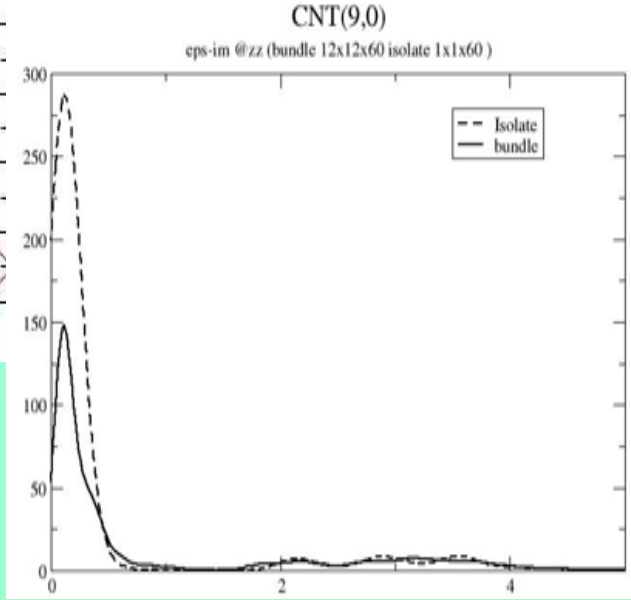

Energy (eV)
k1
k2
k3
k4
k5
k6
k7
k8
k9
k10
ΔE
M.E.
k11
k12
k13
k14
k15
k16
k17
k18
k19
(6,0) bundles

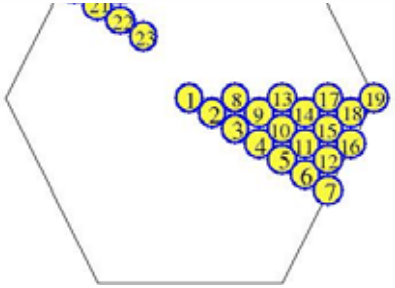
Bundled CNT (9,0)

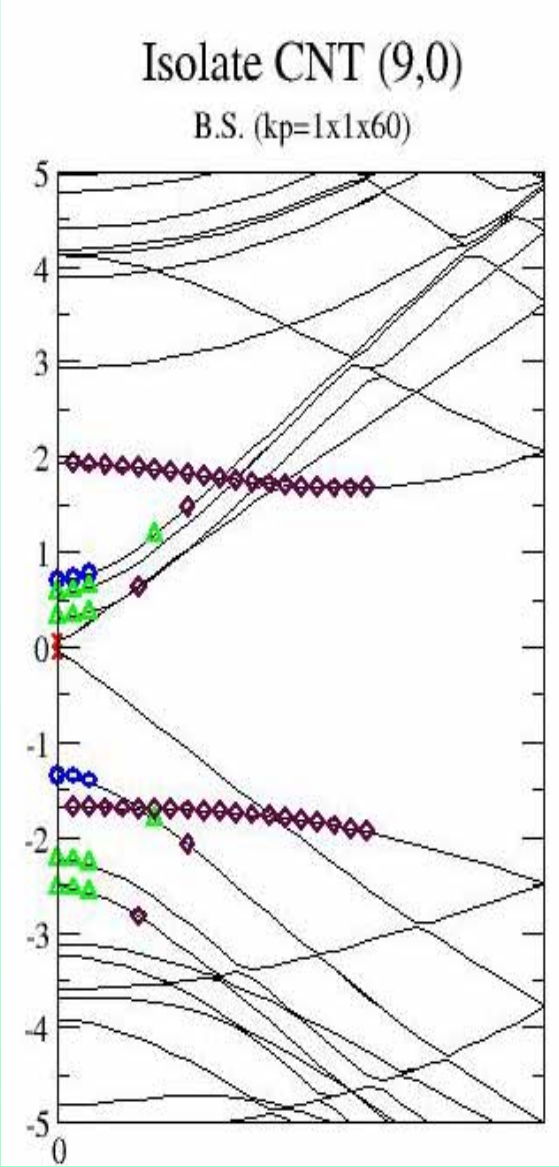
Bundled CNT (9,0) Kpt-1 ~ Kpt-19



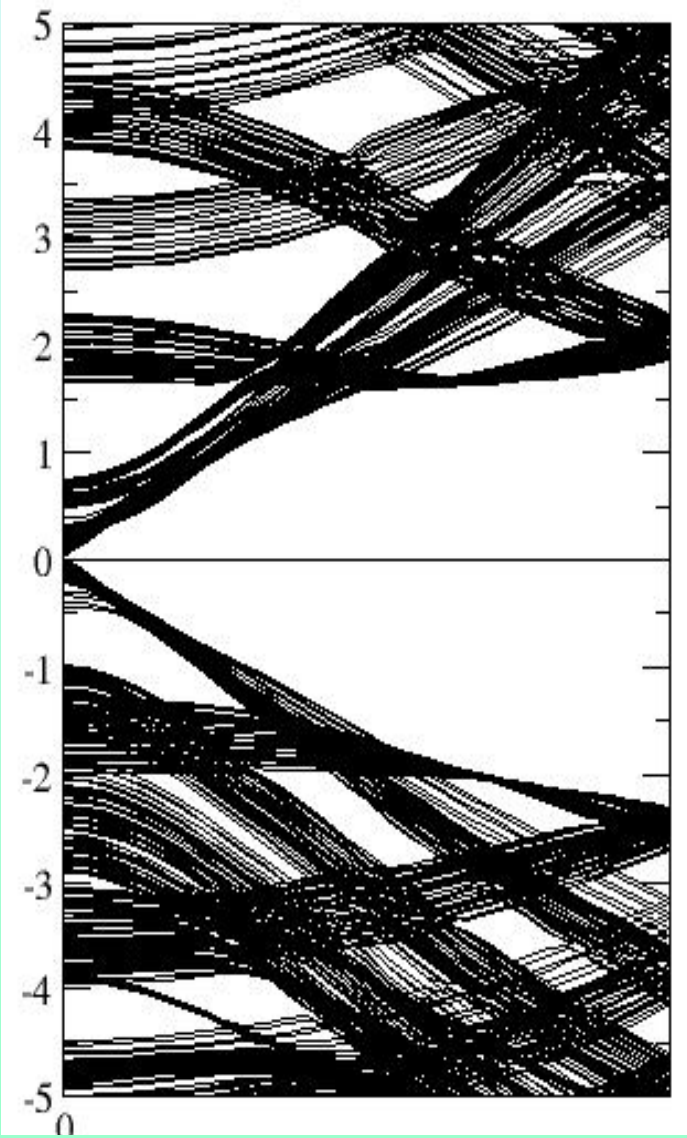






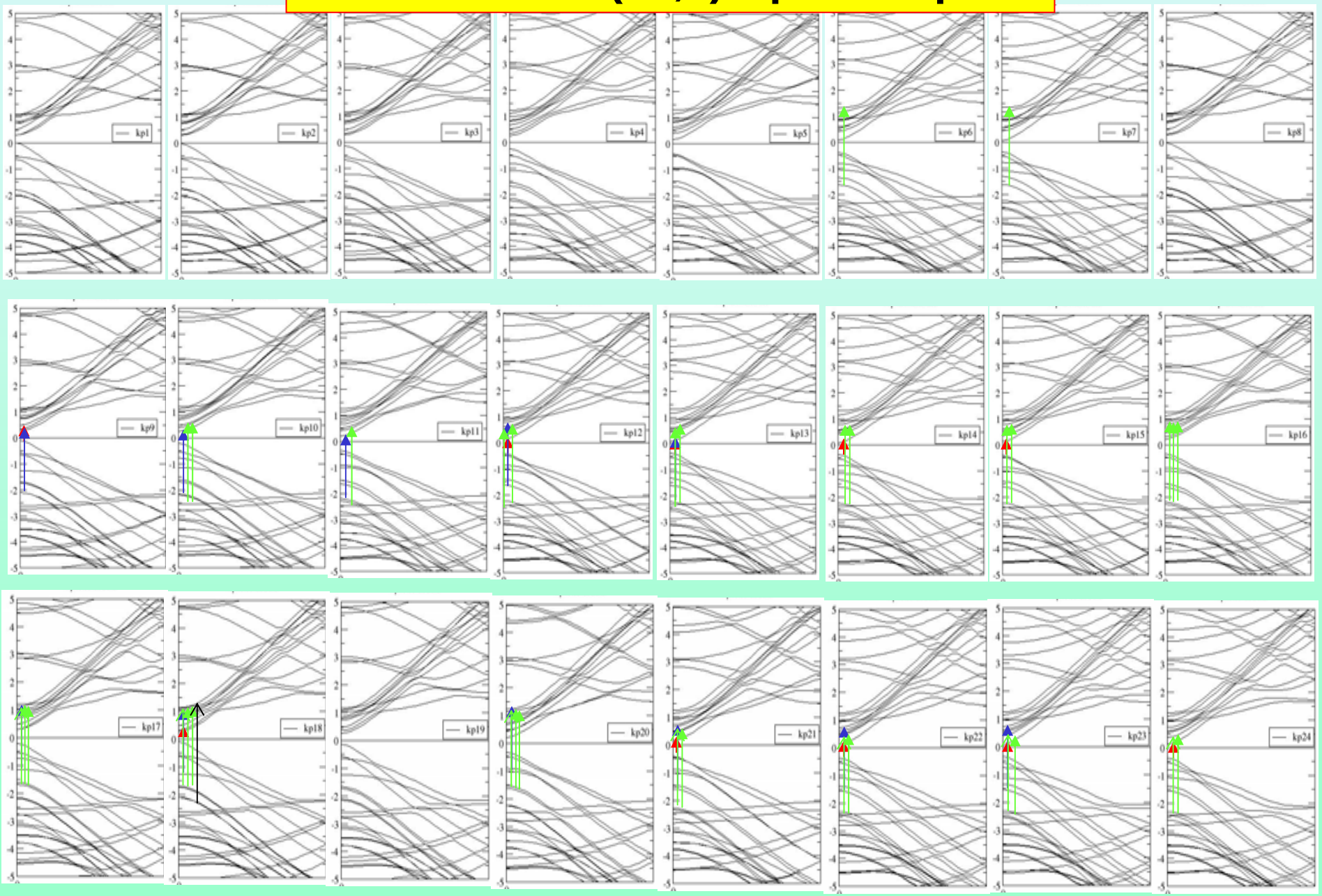


Bundled CNT (9,0)

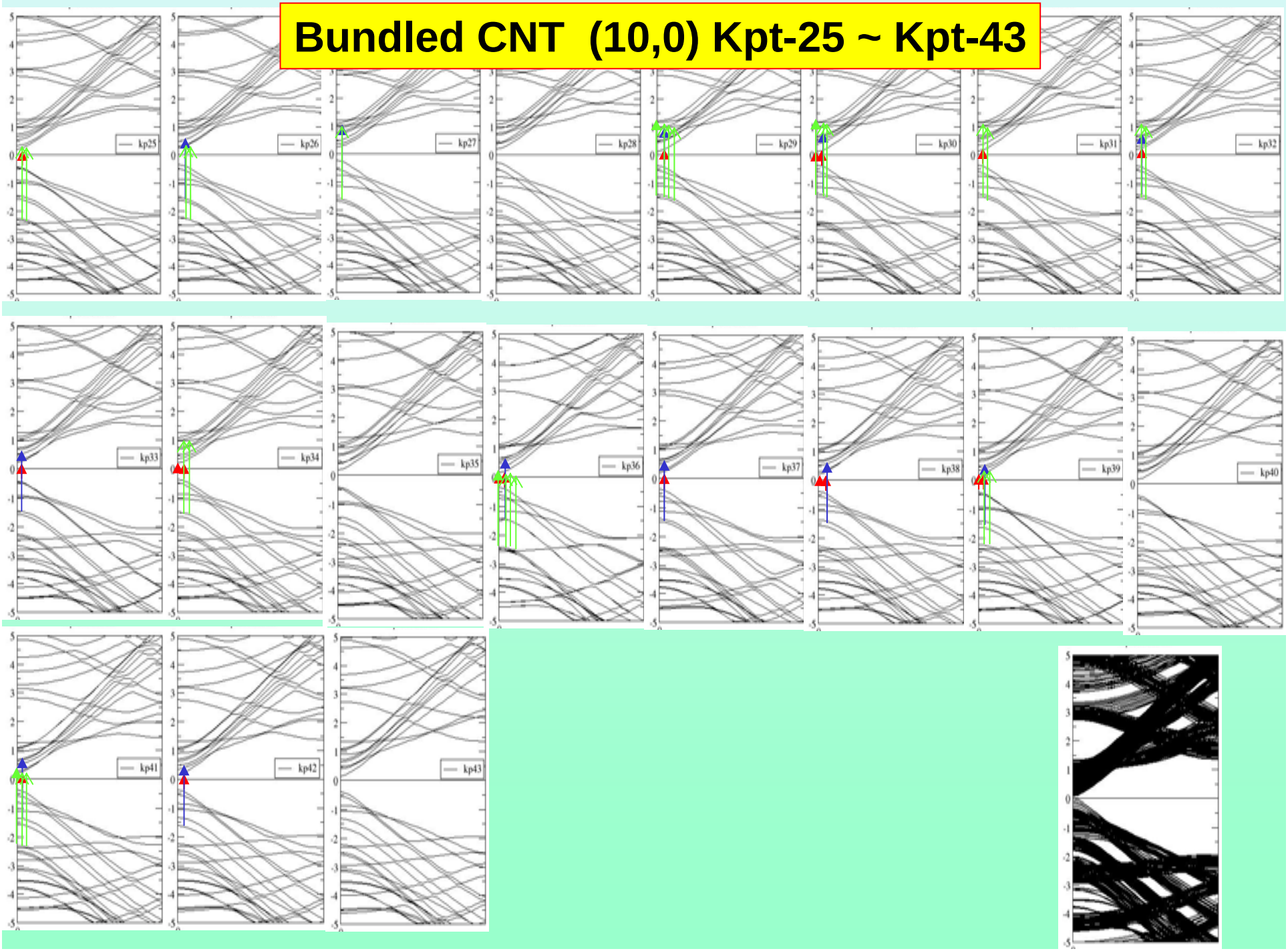


Bundled CNT (10,0)

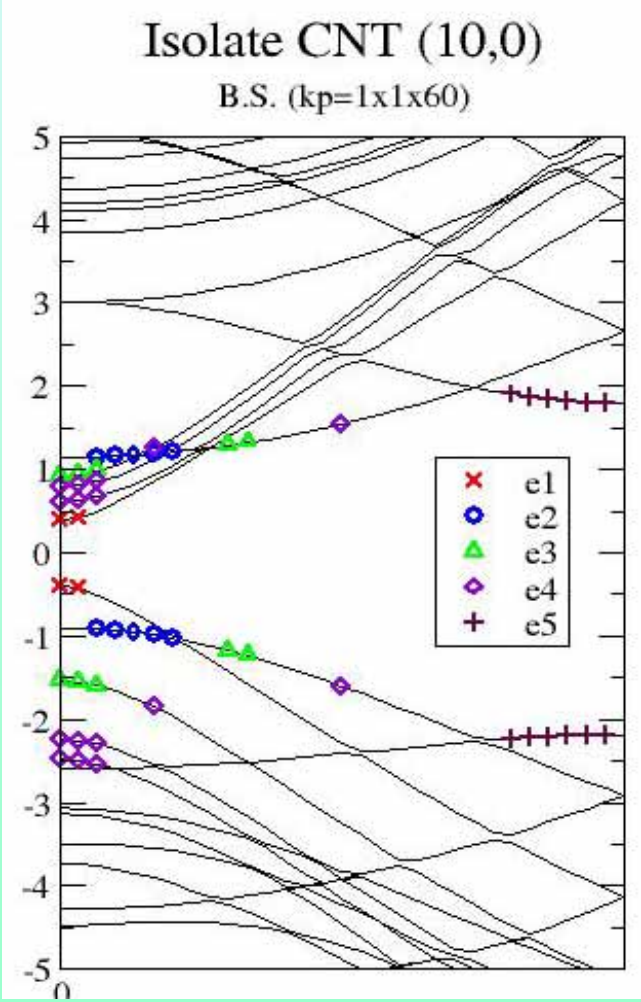
Bundled CNT (10,0) Kpt-1 ~ Kpt-24

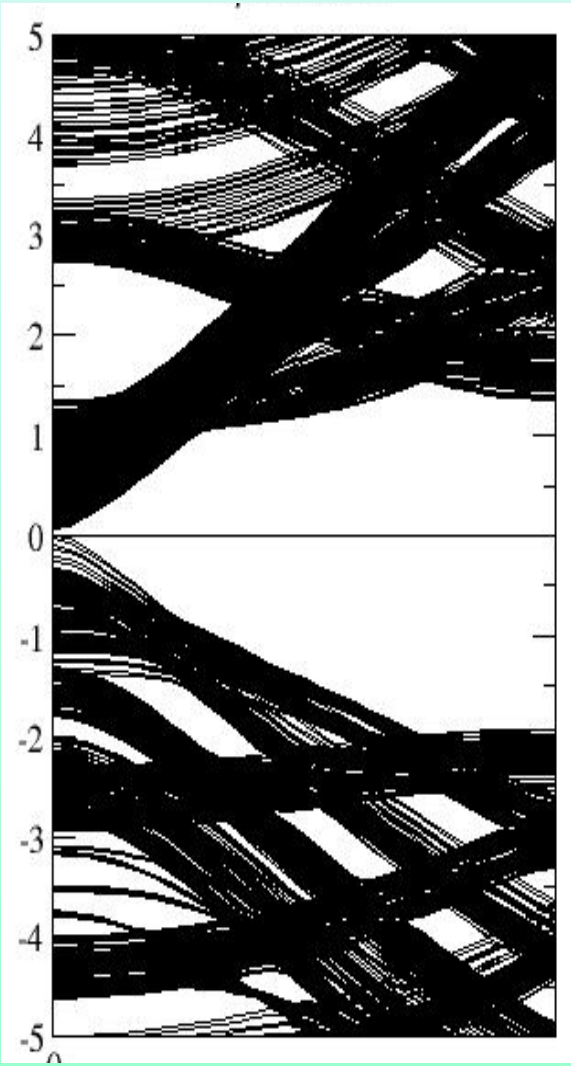


Bundled CNT (10,0) Kpt-25 ~ Kpt-43

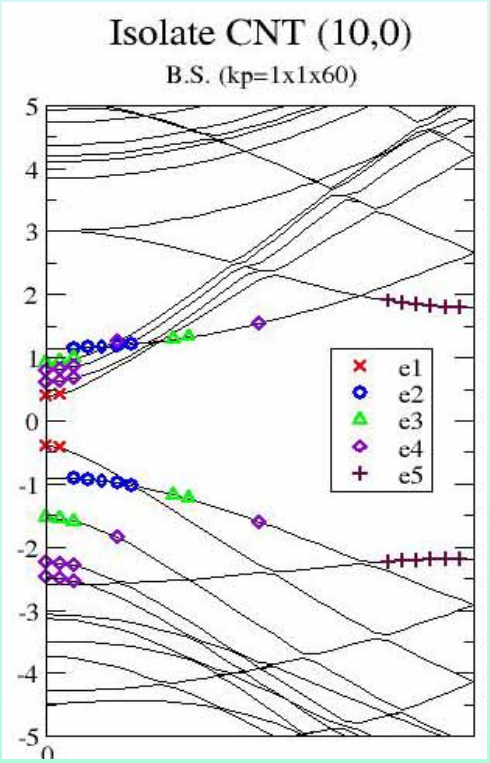


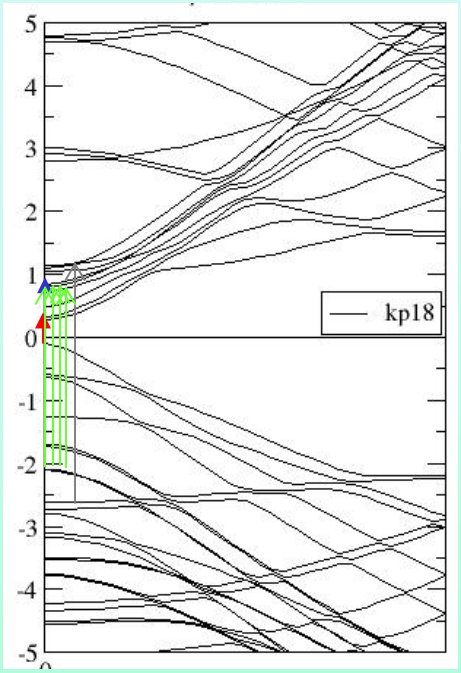
Bundled CNT (10,0)

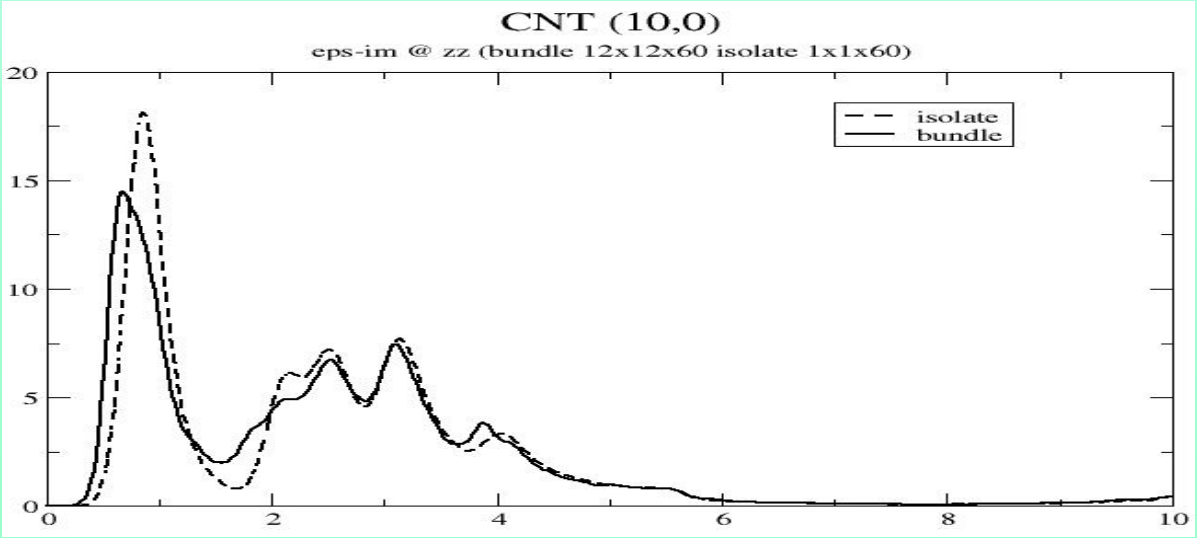




Bundled CNT (10,0) Kpt-18







Summary

1. The armchair tube remains metallic under external electric field, but the dielectric function are modified significantly owing to the subband mixing caused by the external field.

2. In the case of zigzag tubes, the gap is reduced significantly ; however, the effect of the electric field on the dielectric function is less pronounced.

3. The total energy and electronic dispersion show strong dependence on the orientation of the tube in the bundles for the $n = 3q$ (n,n) and (n,0) bundles, while those of the $n \neq 3q$ bundles are nearly independent of such orientation.

4. The optical properties are strongly influence by the orientation of the tube in the bundles.

5. For (n,n) bundle a δ-type peak at about 0.1 eV appears for orientation such that the D_{6h} symmetry is broken.

6. For the case of the (5,0) and (6,0) bundles, the δ-type peak appears at about 0.1 eV due to a pseudogap opening at the Fermi level.

7. The optical spectrum of the (9,0) bundle is also similar to that of the isolated (9,0) tube.

8. The gap of the (10,0) bundles is reduced significantly by the bundling effect, but the modification of the optical spectrum was found to be less pronounced.

Thanks

Bundled CNT (9,0) Kpt-1 ~ Kpt-19