Nanotubes and nanostructures in laser fields: From dynamical symmetry to selective high-order harmonic generation of soft x-rays

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Structure of the lecture

PART I. Dynamical symmetries of nanostructures in laser fields:

- What is dynamical symmetry (DS)?
- Symmetry properties of field-modified eigenstates and energy levels in quantum rings (QR's), thin crystals and single-walled carbon nanotubes (CNT's).

PART II. Selective generation of optical harmonics in high-intensity laser fields:

- What is high-order harmonic generation (HHG)?
- What is the role played by DS in the HHG phenomena?
- Selection rules (SR's) for the HHG spectra (HHGS) by atoms, molecules and CNT's.

PART III. Summary and conclusions.

Electron in a (hindered) quantum ring

$$\hat{H}(\varphi) = \frac{\hat{p}_\varphi^2}{2m\rho_0^2} + V_N(\varphi), \quad \hat{C}_N = \left(\varphi \to \varphi + \frac{2\pi}{N}\right).$$

$$\left[\hat{H}(\varphi), \hat{C}_N\right] = 0 \qquad \Longrightarrow$$

$$\begin{aligned} \hat{H}(\varphi)\psi_{E,p}(\varphi) &= E\psi_{E,p}(\varphi), \\ \hat{C}_N\psi_{E,p}(\varphi) &= e^{+i\frac{2\pi}{N}p}\psi_{E,p}(\varphi), \quad p = 0, 1, \ldots, N-1. \\ \psi_{E,p}(\varphi) &= e^{+ip\varphi}\phi(\varphi), \qquad \phi(\varphi) = \phi(\varphi + 2\pi) \end{aligned}$$

Degenerate states?

CASE I.

If $\left[\hat{H}(\varphi), \hat{\sigma}_y\right] = 0, \qquad \hat{\sigma}_y = (\varphi \to -\varphi)$

$$\hat{\sigma}_y\psi_{E,p}(\varphi) = \psi_{E,p}(-\varphi) = e^{-ip\varphi}\phi(-\varphi).$$

$$\psi_{E,p}(\varphi) \overset{?}{\Longleftrightarrow} \psi_{E,p}(-\varphi)$$

$$\hat{H}(\varphi)\psi_{E,p}(\varphi) = E\psi_{E,p}(\varphi) \Longrightarrow \hat{H}(\varphi)\psi_{E,p}(-\varphi) = E\psi_{E,p}(-\varphi)$$

$p \neq 0, N/2$: $\left\{\hat{\sigma}_y\psi_{E,p}, \psi_{E,p}\right\}$ are degenerate;
span a real 2D irrep,

$p = 0, N/2$: $\hat{\sigma}_y\psi_{E,p} \propto \psi_{E,p}$;
spans a real 1D irrep.

CASE II.

Otherwise, $\left[\hat{H}(\varphi), \hat{R}\right] = 0, \qquad \hat{R} = (t \to -t, *)$

$$\hat{R}\psi_{E,p}(\varphi) = \psi^*_{E,p}(\varphi) = e^{-ip\varphi}\phi^*(\varphi).$$

$$\psi_{E,p}(\varphi) \stackrel{?}{\Longleftrightarrow} \psi^*_{E,p}(\varphi)$$

$$\hat{H}(\varphi)\psi_{E,p}(\varphi) = E\psi_{E,p}(\varphi) \Longrightarrow \hat{H}(\varphi)\psi^*_{E,p}(\varphi) = E\psi^*_{E,p}(\varphi)$$

$p \neq 0, N/2:$ Γ_p is a complex 1D irrep $\Longrightarrow$
$\left\{\Gamma_p, \Gamma^*_p\right\}$ stick together;
$\left\{\hat{R}\psi_{E,p}, \psi_{E,p}\right\}$ are degenerate
(case **c** of Wigner's TRS test).

$p = 0, N/2:$ Γ_p is a real 1D irrep
$\hat{R}\psi_{E,p} \propto \psi_{E,p}$
(case **a** of Wigner's TRS test).

Electron in a (hindered) quantum ring exposed to circularly-polarized light

Time-periodic Hamiltonian:

$$\hat{H}(\varphi,t) = \frac{\hat{p}_\varphi^2}{2m\rho_0^2} + V_N(\varphi) + eE_0\rho_0 \cos(\varphi - \omega t)$$

$$\hat{H}(t) = \hat{H}(t+T),\ T = \frac{2\pi}{\omega}, \quad \hat{H}(t)\Psi(t) = i\hbar\frac{\partial}{\partial t}\Psi(t)$$

Floquet Theorem:

$$\Psi_\varepsilon(t) = e^{-\frac{i}{\hbar}\varepsilon t}\Phi_\varepsilon(t), \quad \Phi_\varepsilon(t) = \Phi_\varepsilon(t+T),$$

$$\hat{\mathcal{H}}_f(t)\Phi_\varepsilon(t) = \varepsilon\Phi_\varepsilon(t), \quad \hat{\mathcal{H}}_f(t) \equiv \hat{H}(t) - i\hbar\frac{\partial}{\partial t}$$

Why **quasi**energies (QE's)?

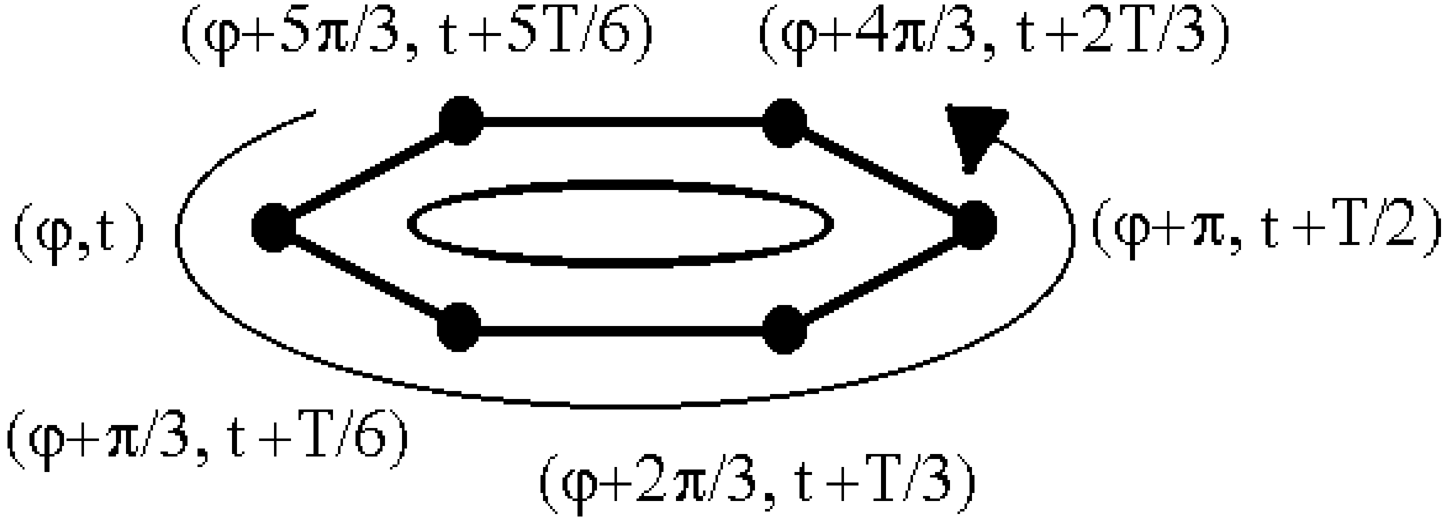
$$e^{-\frac{i}{\hbar}\varepsilon t}\Phi_\varepsilon(t) = e^{-\frac{i(\varepsilon+l\hbar\omega)t}{\hbar}} \cdot \left[e^{+l\omega t}\Phi_\varepsilon(t)\right] \equiv e^{-\frac{i\varepsilon_l t}{\hbar}}\Phi_{\varepsilon_l}(t),$$

where $\varepsilon_l = \varepsilon + l\hbar\omega, \quad l \in \mathcal{Z}$.

The N-th order dynamical symmetry (DS):

$$\left[\hat{\mathcal{H}}_f, \hat{P}_N\right] = 0, \qquad \hat{P}_N = \left(\varphi \to \varphi + \frac{2\pi}{N},\ t \to t + \frac{T}{N}\right)$$

$$\boxed{\hat{P}_N\Phi_{\varepsilon,p} = e^{+i\frac{2\pi}{N}p}\Phi_{\varepsilon,p}, \quad p = 0,\ldots,N-1.}$$



(φ+5π/3, t+5T/6)
(φ+4π/3, t+2T/3)
(φ,t)
(φ+π, t+T/2)
(φ+π/3, t+T/6)
(φ+2π/3, t+T/3)

Degenerate Floquet states?

$$\widehat{\mathcal{H}}_f(\varphi,t) = \frac{\widehat{p}_\varphi^2}{2m\rho_0^2} + V_N(\varphi) + eE_0\rho_0 \cos(\varphi - \omega t) - i\hbar\frac{\partial}{\partial t}$$

CASE I.

If $\left[\widehat{H}(\varphi), \widehat{\sigma}_y\right] \neq 0, \quad \widehat{\sigma}_y = (\varphi \to -\varphi) \qquad \Longrightarrow$

$\left[\widehat{\mathcal{H}}_f(\varphi,t), \widehat{R}\right] \neq 0, \quad \widehat{R} = (t \to -t, *).$

Cyclic, unitary DS group:

$$\mathcal{G}_N \equiv \left\{\widehat{P}_N,\ \widehat{P}_N^2,\ \ldots,\ \widehat{P}_N^{N-1},\ \widehat{P}_N^N = \mathbf{I}\right\}.$$

All irrep's $\Gamma_p\left(\widehat{P}_N\right)$, $p = 1, 2, \ldots, N-1$ are 1D.

$\Longrightarrow$ No non-accidental degeneracies.

CASE II.

If $\left[\widehat{H}(\varphi), \widehat{\sigma}_y\right] = 0 \qquad \Longrightarrow$

Generalized time-reversal symmetry:

$$\widehat{R}_{\varphi t} = (\varphi \to -\varphi,\ t \to -t,\ *)$$

Non-unitary DS group:

$$\mathcal{G}_N \otimes \left\{\mathbf{I}, \widehat{R}_{\varphi t}\right\}$$

A simple argument:

$$\Phi_{\varepsilon,p}(\varphi,t) \overset{?}{\Longleftrightarrow} \hat{R}_{\varphi t}\Phi_{\varepsilon,p}(\varphi,t)$$

$$\hat{P}_N\left[\Phi_{\varepsilon,p}(\varphi,t)\right] \overset{?}{\Longleftrightarrow} \hat{P}_N\left[\hat{R}_{\varphi t}\Phi_{\varepsilon,p}(\varphi,t)\right]$$

Unlike the field-free case $\left(\hat{C}_N\hat{R} = \hat{R}\hat{C}_N\right)$,

$$\hat{P}_N\hat{R}_{\varphi t} = \hat{R}_{\varphi t}\hat{P}_N^{-1}$$

$$\begin{aligned} \hat{P}_N\left[\Phi_{\varepsilon,p}\right] &= e^{+i\frac{2\pi}{N}p}\left[\Phi_{\varepsilon,p}\right] \quad \Longrightarrow \\ \hat{P}_N\left[\hat{R}_{\varphi t}\Phi_{\varepsilon,p}\right] &= \hat{R}_{\varphi t}\left[\hat{P}_N^{-1}\Phi_{\varepsilon,p}\right] = e^{+i\frac{2\pi}{N}p}\left[\hat{R}_{\varphi t}\Phi_{\varepsilon,p}\right] \\ \Phi_{\varepsilon,p} &\propto \hat{R}_{\varphi t}\Phi_{\varepsilon,p}, \quad \underline{\text{for all } p.} \end{aligned}$$

$\Longrightarrow$ No new degeneracy is introduced.

A (more) rigorous argument: Wigner's theory of anti-unitary symmetries (corepresantations):

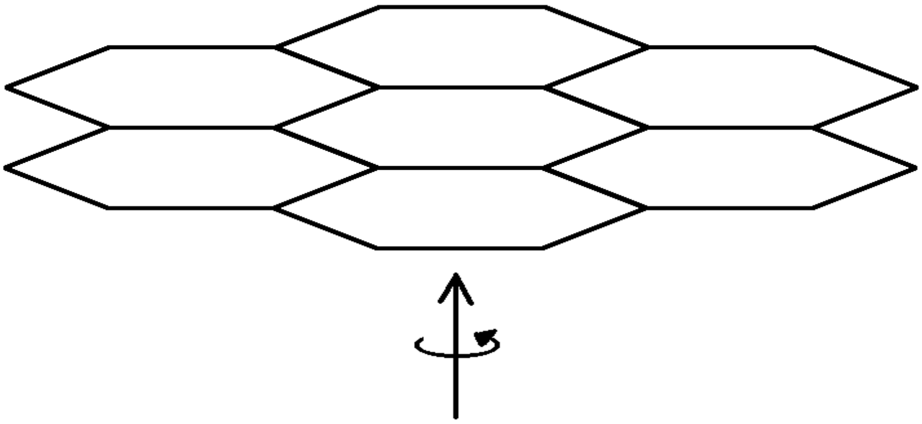
$$\bar{\Gamma}_p\left(\hat{P}_N\right) \equiv \Gamma_p^*\left(\hat{R}_{\varphi t}^{-1}\hat{P}_N\hat{R}_{\varphi t}\right) \overset{?}{\Longleftrightarrow} \Gamma_p\left(\hat{P}_N\right)$$

$$\bar{\Gamma}_p\left(\hat{P}_N\right) = \Gamma_p^*\left(\hat{R}_{\varphi t}^{-1}\hat{P}_N\hat{R}_{\varphi t}\right) = \Gamma_p^*\left(\hat{P}_N^{-1}\right) = \Gamma_p\left(\hat{P}_N\right)$$

(case **a** of Wigner's anti-unitary (TRS) test)

$\Longrightarrow$ No new degeneracy is introduced.

A thin crystal in CP-light



$$\widehat{H}_{cr}(\mathbf{r}) = \frac{\widehat{p}_x^2 + \widehat{p}_y^2}{2m} + V_{cr}(\mathbf{r}), \quad \left[\widehat{H}_{cr}(\mathbf{r}), \widehat{C}_N\right] = \left[\widehat{H}_{cr}(\mathbf{r}), \widehat{\sigma}_y\right] = 0$$

$$\widehat{\mathcal{H}}_f(\mathbf{r},t) = \widehat{H}_{cr}(\mathbf{r}) - \frac{eE_0}{m\omega}\left[\widehat{p}_x \cos(\omega t) + \widehat{p}_y \sin(\omega t)\right] - i\hbar\frac{\partial}{\partial t}$$

$$\left[\widehat{p}_x \cos(\omega t) + \widehat{p}_y \sin(\omega t)\right] \sim \cos(\varphi - \omega t)$$

Floquet-Bloch (FB) states:

$$\begin{aligned} \Phi_{\varepsilon(\mathbf{k}),\mathbf{k}}(\mathbf{r},t) &= e^{+i\mathbf{k}\mathbf{r}}\phi_{\varepsilon(\mathbf{k}),\mathbf{k}}(\mathbf{r},t), \\ \widehat{\mathcal{H}}_f(\mathbf{r},t)\Phi_{\varepsilon(\mathbf{k}),\mathbf{k}}(\mathbf{r},t) &= \varepsilon(\mathbf{k})\Phi_{\varepsilon(\mathbf{k}),\mathbf{k}}(\mathbf{r},t). \end{aligned}$$

Symmetry properties of the QEB's

$$\widehat{P}_N\Phi_{\varepsilon(\mathbf{k}),\mathbf{k}}(\mathbf{r},t) \equiv e^{+i(C_N^{-1}\mathbf{k})\cdot\mathbf{r}}\phi_{\varepsilon(\mathbf{k}),\mathbf{k}}(C_N\mathbf{r}, t+T/N) \equiv \Phi_{\varepsilon(\mathbf{k}),C_N^{-1}\mathbf{k}}(\mathbf{r},t)$$

$$\Longrightarrow \quad \varepsilon(C_N\mathbf{k}) = \varepsilon(\mathbf{k})$$

$$\widehat{R}_{\varphi t}\Phi_{\varepsilon(\mathbf{k}),\mathbf{k}}(\mathbf{r},t) \equiv e^{+i(-k_x x + k_y y)}\phi^*_{\varepsilon(\mathbf{k}),\mathbf{k}}(\sigma_y\mathbf{r}, -t) \equiv \Phi_{\varepsilon(\mathbf{k}),\sigma_x\mathbf{k}}(\mathbf{r},t)$$

$$\Longrightarrow \quad \varepsilon(\sigma_x\mathbf{k}) = \varepsilon(\mathbf{k})$$

plane-group	point-group	EB-sym.	DS pl. group	DS pt. group	QEB-sym.	GTRS
$p1$	$\mathcal{C}_1$	$\mathcal{C}_2$	$g1$	$\mathcal{G}_1$	$\mathcal{C}_1$	NO
$pm[x]$	$\mathcal{C}_s[x]$	$\mathcal{C}_{2v}$	$g1$	$\mathcal{G}_1$	$\mathcal{C}_s[y]$	$\hat{R}_{\varphi t}$
$pg[x]$	$\mathcal{C}_s[x]$	$\mathcal{C}_{2v}$	$g1$	$\mathcal{G}_1$	$\mathcal{C}_s[y]$	$\hat{R}_{gt}$
$cm[x]$	$\mathcal{C}_s[x]$	$\mathcal{C}_{2v}$	$g1$	$\mathcal{G}_1$	$\mathcal{C}_s[y]$	$\hat{R}_{\varphi t}$
$p2$	$\mathcal{C}_2$	$\mathcal{C}_2$	$g2$	$\mathcal{G}_2$	$\mathcal{C}_2$	NO
$p2mg$	$\mathcal{C}_{2v}$	$\mathcal{C}_{2v}$	$g2$	$\mathcal{G}_2$	$\mathcal{C}_{2v}$	$\hat{R}_{gt}$
$p2gg$	$\mathcal{C}_{2v}$	$\mathcal{C}_{2v}$	$g2$	$\mathcal{G}_2$	$\mathcal{C}_{2v}$	$\hat{R}_{gt}$
$p2mm$	$\mathcal{C}_{2v}$	$\mathcal{C}_{2v}$	$g2$	$\mathcal{G}_2$	$\mathcal{C}_{2v}$	$\hat{R}_{\varphi t}$
$c2mm$	$\mathcal{C}_{2v}$	$\mathcal{C}_{2v}$	$g2$	$\mathcal{G}_2$	$\mathcal{C}_{2v}$	$\hat{R}_{\varphi t}$
$p3$	$\mathcal{C}_3$	$\mathcal{C}_6$	$g3$	$\mathcal{G}_3$	$\mathcal{C}_3$	NO
$p3m1[x]$	$\mathcal{C}_{3v}[x]$	$\mathcal{C}_{6v}$	$g3$	$\mathcal{G}_3$	$\mathcal{C}_{3v}[y]$	$\hat{R}_{\varphi t}$
$p31m[x]$	$\mathcal{C}_{3v}[x]$	$\mathcal{C}_{6v}$	$g3$	$\mathcal{G}_3$	$\mathcal{C}_{3v}[y]$	$\hat{R}_{\varphi t}$
$p4$	$\mathcal{C}_4$	$\mathcal{C}_4$	$g4$	$\mathcal{G}_4$	$\mathcal{C}_4$	NO
$p4gm$	$\mathcal{C}_{4v}$	$\mathcal{C}_{4v}$	$g4$	$\mathcal{G}_4$	$\mathcal{C}_{4v}$	$\hat{R}_{gt}$
$p4mm$	$\mathcal{C}_{4v}$	$\mathcal{C}_{4v}$	$g4$	$\mathcal{G}_4$	$\mathcal{C}_{4v}$	$\hat{R}_{\varphi t}$
$p6$	$\mathcal{C}_6$	$\mathcal{C}_6$	$g6$	$\mathcal{G}_6$	$\mathcal{C}_6$	NO
$p6mm$	$\mathcal{C}_{6v}$	$\mathcal{C}_{6v}$	$g6$	$\mathcal{G}_6$	$\mathcal{C}_{6v}$	$\hat{R}_{\varphi t}$

- $pm[x]$ - reflections are parallel to the x-axis.
- (unitary) DS plane-groups are symmorphic:

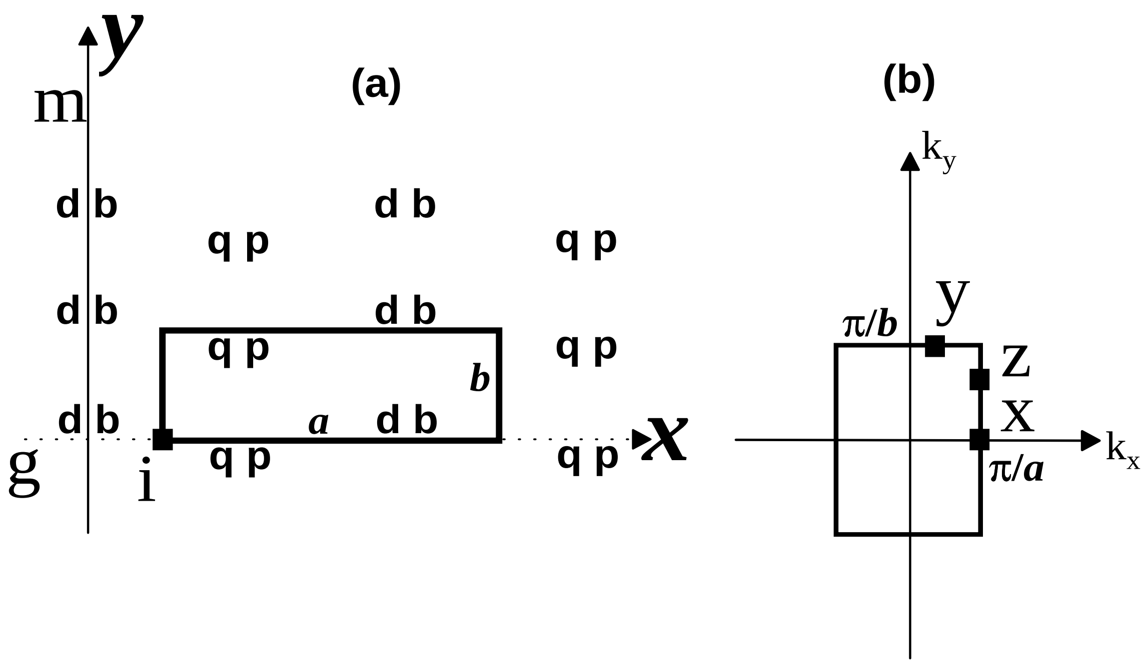
$$p4gm \implies g4 = \text{(2D translations)} \wedge \mathcal{G}_4.$$

Degenerate FB-states at specific k's?

Case A: The $\Gamma(k = 0)$-point ($\equiv$ (hindered) quantum ring) $\implies$ No non-accidental degeneracies.

Case B: The edge of the BZ;
$\implies$ Sticking together of QEB's?

A p2mg-lattice in CP-light



(a) A 2D motif possessing the $p2mg$ symmetry.

(b) The corresponding rectangular 1-st BZ.

The glide reflection symmetry:

$$\hat{g} = \left(x \to x + \frac{a}{2}, y \to -y \right)$$

Generalized time-reversal symmetry:

$$\hat{\mathcal{H}}_f(\mathbf{r}, t) = \hat{H}_{cr}(\mathbf{r}) - \frac{eE_0}{m\omega} [\hat{p}_x \cos(\omega t) + \hat{p}_y \sin(\omega t)] - i\hbar \frac{\partial}{\partial t}$$

$$\hat{R}_{gt} = \left(x \to x + \frac{a}{2}, y \to -y, \ t \to -t, \ * \right)$$

The edge Z of the BZ:

$$\Phi_{\varepsilon(\mathbf{Z}),\mathbf{Z}}(\mathbf{r},t) = e^{+i(\frac{\pi}{a}x+k_y y)}\phi_{\varepsilon(\mathbf{Z}),\mathbf{Z}}(\mathbf{r},t),$$
$$k_Z = \left(\frac{\pi}{a}, k_y\right), k_y \in \left(\frac{\pi}{b}, \frac{\pi}{b}\right]$$

The effect of $\hat{R}_{gt}$ on $\Phi_{\varepsilon(\mathbf{Z}),\mathbf{Z}}(\mathbf{r},t)$:

$$\hat{R}_{gt} = \left(x \to x + \frac{a}{2}, y \to -y,\ t \to -t,\ *\right)$$

$$\hat{R}_{gt}\Phi_{\varepsilon(\mathbf{Z}),\mathbf{Z}}(\mathbf{r},t) = \hat{R}_{gt} e^{+i(\frac{\pi}{a}x+k_y y)}\phi_{\varepsilon(\mathbf{Z}),\mathbf{Z}}(\mathbf{r},t) =$$
$$= -ie^{-i(\frac{\pi}{a}x-k_y y)}\phi^*_{\varepsilon(\mathbf{Z}),\mathbf{Z}}\left(x+\frac{a}{2}, -y, -t\right)$$

$$\hat{R}_{gt}\Phi_{\varepsilon(\mathbf{Z}),\mathbf{Z}}(\mathbf{r},t) \propto \Phi_{\varepsilon(\mathbf{Z}),\mathbf{Z}}(\mathbf{r},t)?$$

Suppose that,

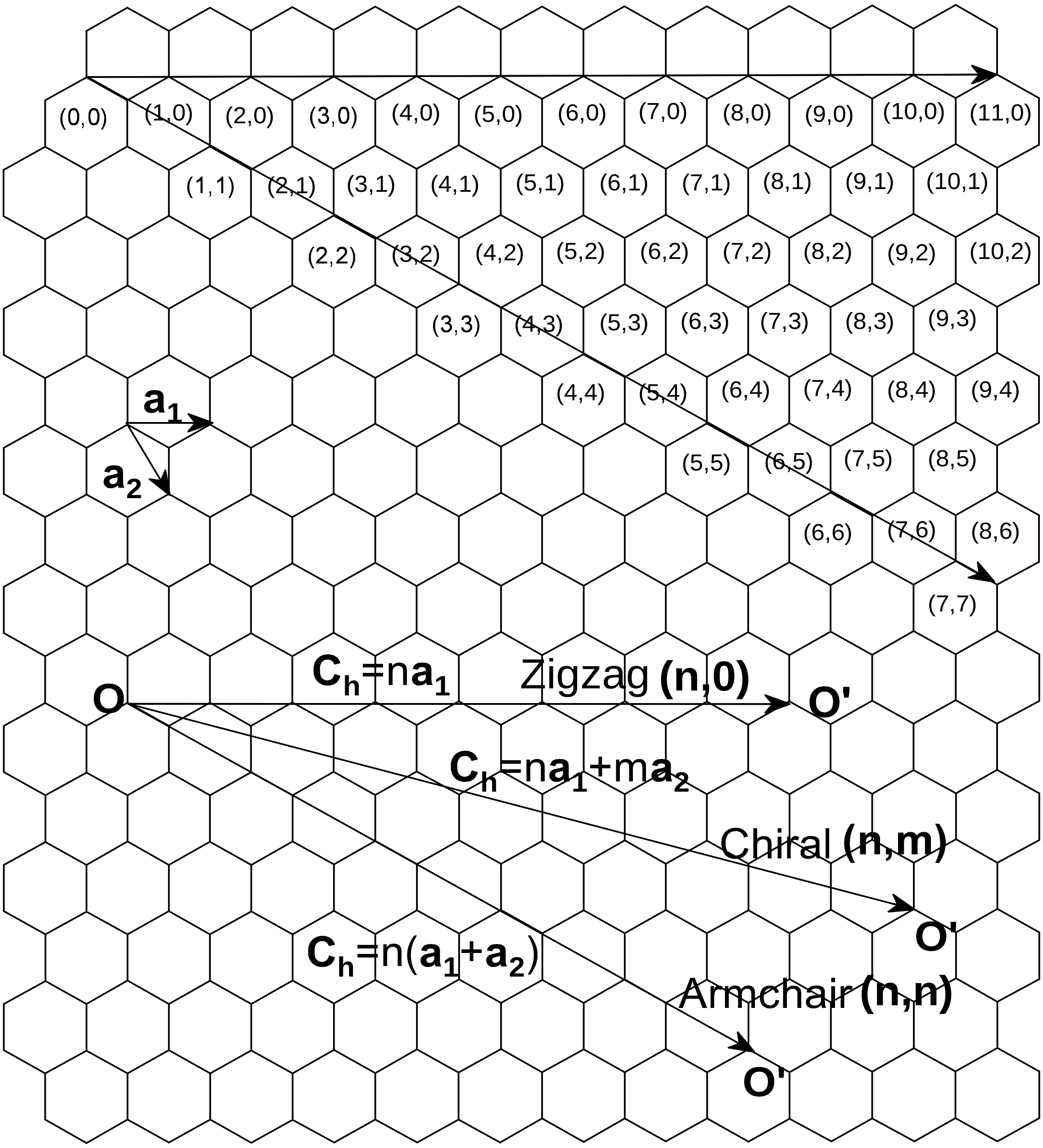
$$\hat{R}_{gt}\Phi_{\varepsilon(Z),Z}(\mathbf{r},t) = \beta\Phi_{\varepsilon(Z),Z}(\mathbf{r},t),$$

$$\left[\hat{R}_{gt}\hat{R}_{gt}\right]\Phi_{\varepsilon(Z),Z} = (x \to x + a)\,\Phi_{\varepsilon(Z),Z} = -\Phi_{\varepsilon(Z),Z}\,,$$

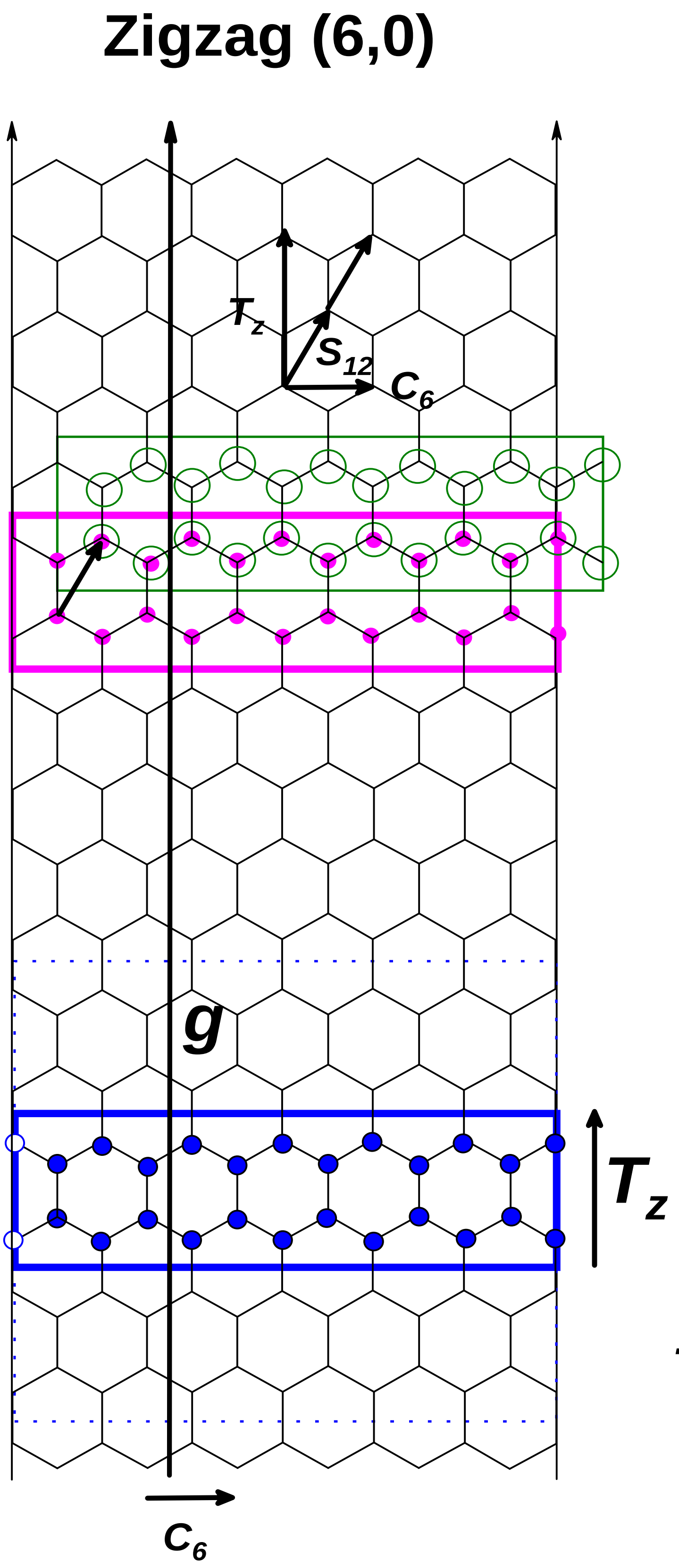
$$\hat{R}_{gt}\left[\hat{R}_{gt}\Phi_{\varepsilon(Z),Z}\right] = \hat{R}_{gt}\beta\Phi_{\varepsilon(Z),Z} = \beta^*\beta\Phi_{\varepsilon(Z),Z}\,.$$

However, $\beta^*\beta \neq -1 \quad \Longrightarrow$
Sticking together of QEB's along Z!

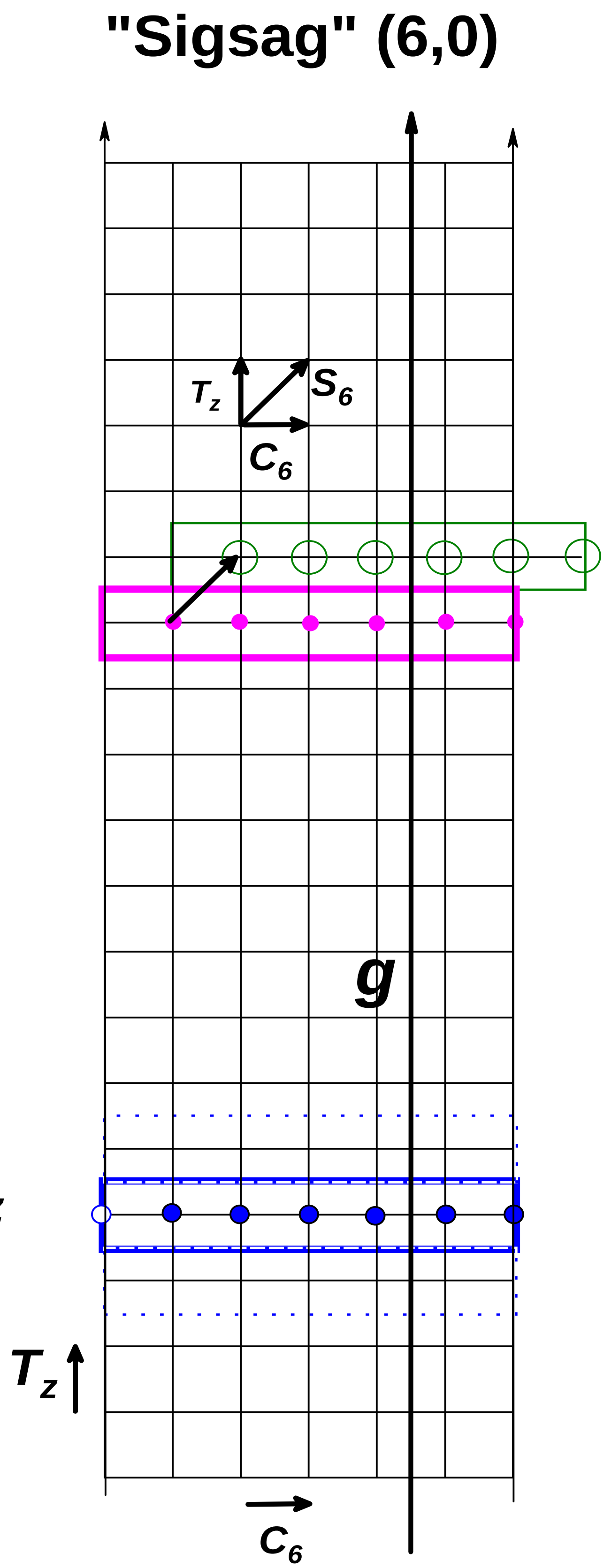
Carbon nanotubes (CNT's): Rolling the graphite sheet



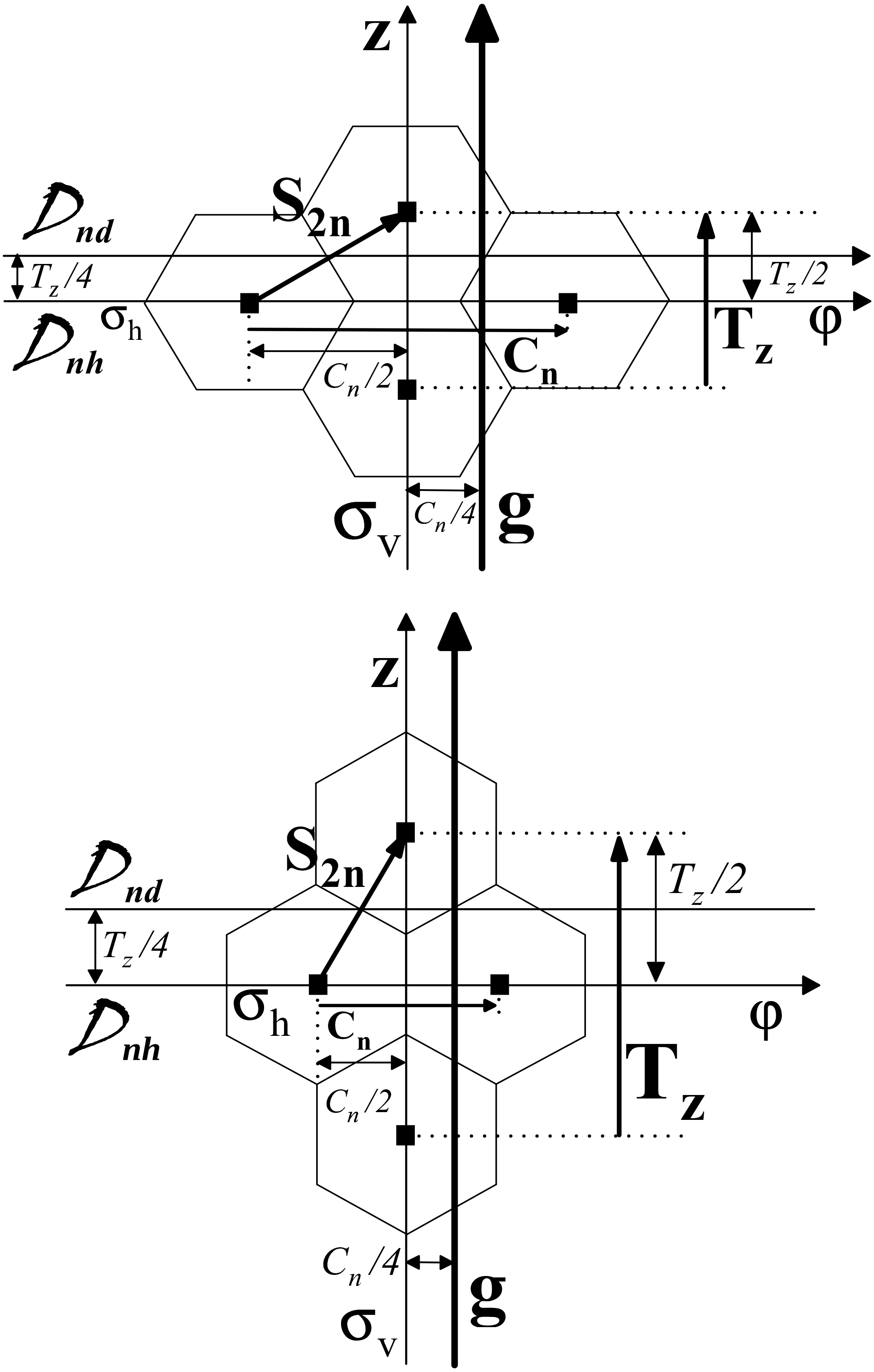
Symmetry of achiral CNT's



NON-Symmorfic



Symmorfic

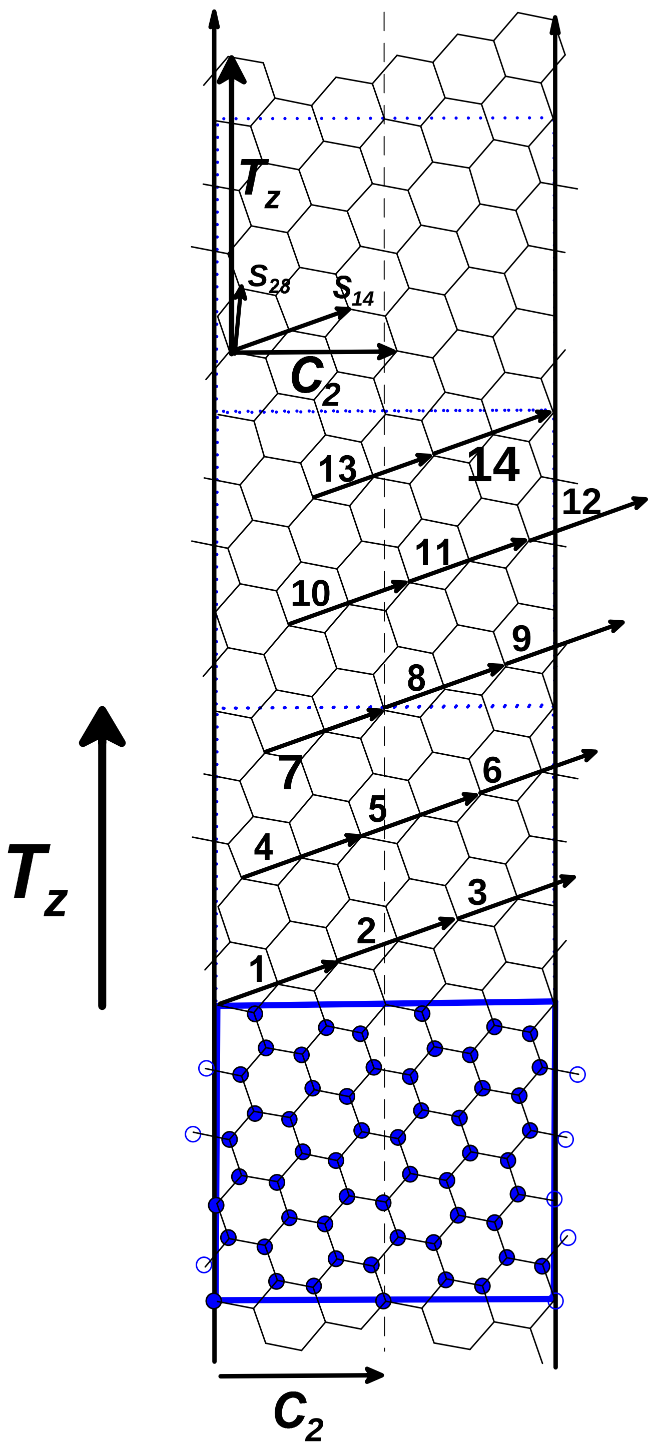


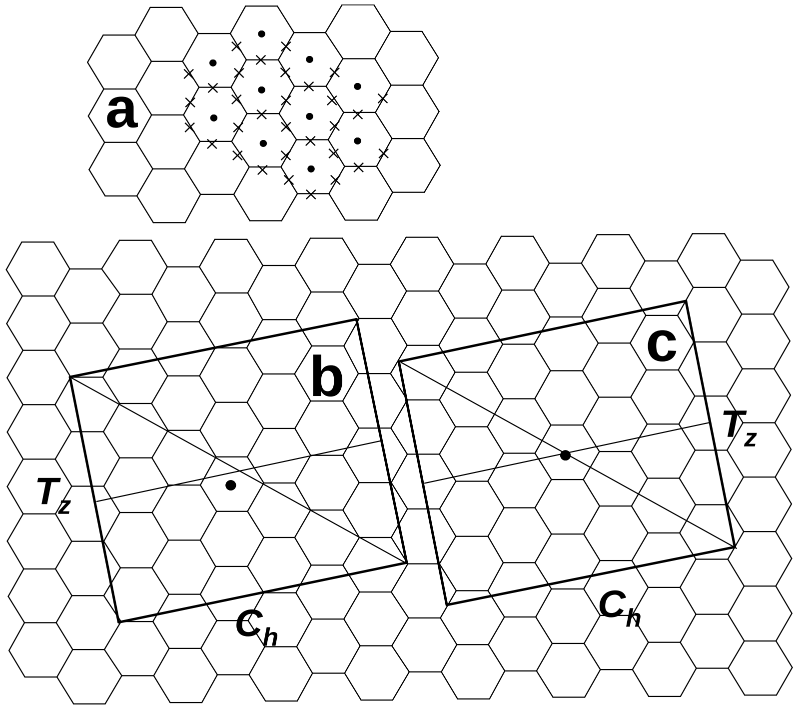
Top: armchair segment; bottom: zigzag segment

$$\mathcal{G}^{a}[n] = \mathcal{L}_{T_z}\times\left[\mathcal{D}_{nh}|_{z=0} \oplus \left(\mathcal{D}_{nd}|_{z=\frac{T_z}{4}} \ominus \mathcal{C}_{nv}\right) \oplus \mathcal{C}_{nv}\times \mathbf{S}_{2n}\right]$$

Symmetry of chiral CNT's

Chiral (4,2)





a. A segment of 2D graphite

b. (4,2) CNT conventional unit cell

c. (4,2) CNT translated unit cell

NON-Commutative

$$\mathcal{G}^{c}[N] = \mathcal{L}_{T_z} \times \mathcal{D}_d \times \left[\sum_{j=0}^{\frac{N}{d}-1} \mathbf{S}^{j}_{\frac{N}{d}}\right] = \mathcal{L}_{T_z} \times \mathcal{D}_1 \times \left[\sum_{j=0}^{N-1} \mathbf{S}^{j}_{N}\right]$$

The Floquet Hamiltonian beyond the dipole approximation

$$\hat{\mathcal{H}}_f(\mathbf{r},t) = \frac{\left(\hat{\mathbf{p}} - \frac{e}{c}\hat{\mathbf{A}}(\mathbf{r},t)\right)^2}{2m} + \hat{V}_{\text{nanotube}}(\mathbf{r}) - i\hbar\frac{\partial}{\partial t}$$

$$\hat{\mathcal{H}}_f(\mathbf{r},t)\Phi_\varepsilon(\mathbf{r},t) = \varepsilon\Phi_\varepsilon(\mathbf{r},t)$$

$$\mathbf{A}(\mathbf{r},t) = \frac{cE_0}{\omega}\left[\cos(\omega t - k_0 z), \alpha \sin(\omega t - k_0 z), 0\right]$$

$$\mathbf{k} = (0,0,k_0) \qquad \alpha = \pm 1$$

$$\hat{H}_{\text{interaction}} \propto \cos\left[\alpha\left(\omega t - k_0 z\right) - \varphi\right]$$

Anti-unitary DS operation

$$R_{\varphi z t} = (\varphi \to -\varphi,\ z \to -z,\ t \to -t, *)$$

Basic symmetry operations:

Screw-axis (N-order, R-revolutions, M-translations):

$$S_N = (\varphi \to \varphi + \psi,\ z \to z + \tau),\ \psi = \frac{2\pi R}{N}, \tau = \frac{MT_z}{N}$$

Primitive translation:

$$T_\infty = (z \to z + T_z)$$

Unitary DS operations

$$P_N = \left(\varphi \to \varphi + \psi,\, z \to z + \tau,\, t \to t + \frac{k_0 \tau}{\omega} + \alpha \frac{\psi}{\omega}\right)$$

$$P_\infty = \left(z \to z + T_z,\, t \to t + \frac{k_0 T_z}{\omega}\right)$$

Properties of the unitary DS's:

$$\hat{P}_\infty^{-M} \cdot \hat{P}_N^N = \mathbf{I}.$$

Quantum numbers:

$$\hat{P}_\infty \Phi_{\varepsilon,q,p}(\mathbf{r},t) = e^{+iqT_z} \Phi_{\varepsilon,q,p}(\mathbf{r},t), \quad -\frac{\pi}{T_z} < q \leq +\frac{\pi}{T_z},$$

$$\hat{P}_N \Phi_{\varepsilon,q,p}(\mathbf{r},t) = e^{+iqT_z\frac{M}{N}} e^{+i\frac{2\pi R}{N}p} \Phi_{\varepsilon,q,p}(\mathbf{r},t), \quad p = 0, 1, \ldots, N-1.$$

The symmetry of and the non-accidental degeneracies in the QEB's...

Summary of PART I.

(1) Symmetry (space) + Light (time & space) $\Longrightarrow$ Dynamical Symmetry (space–time).

(2) Label QE's and their corresponding Floquet states.

(3) Determine symmetry properties of QEB's.

(4) Analyze non-accidental degeneracies and, especially, the effect of GTRS on QE/QEB spectra.

The next step:

Can DS aid in evaluating 'suitable' matrix elements?...