Harmonic generation- "11 culture stations"

1960- Maiman: LASER

1961- Franken *et al.*: 2-nd HG by a crystal

1967- New and Ward: 3-rd HG in rares

1987- McPherson *et al.*: Plateau

1993- Budil *et al.*: Suppression of HG in elliptically-polarized fields

1997- Chang *et al.*: $\lambda_{\text{out}} = 2.7$ nm

1998- Schnürer *et al.*: $\lambda_{\text{out}} \leq 2.5$ nm

1999- Constant *et al.*: Conversion efficiency of 4×10^{-5}

1999- Andiel *et al.*: Two-color CC of HHG

2000- Bartels *et al.*: "Teaching lasers to control HHG"

2001- Velotta *et al.*: "HHG in aligned molecules"

Question: Can we control the HHG spectrum (HHGS) such that, e.g.,

$$\mathbf{n} = 1, \cancel{3}, \cancel{5}, \cancel{7}, \cancel{9}, \cancel{11}, \ldots, 101?$$

The first high$-$order harmonic is $\mathbf{n} = 101$!

Answer: Yes, using the DS properties of the Floquet Hamiltonian.

A simpler question: Why atoms emit only odd harmonics?

$$\mathbf{n} = 1, \cancel{2}, 3, \cancel{4}, 5, \cancel{6}, 7, \ldots, 2j-1?$$

Answer: Selection rules (SR's) $\Longrightarrow$ Matrix elements; Symmetry operators

Radiation by an accelerating charge

Larmor formula (classical electrodynamics):

$$P_{class}(t) = \frac{2e^2}{3c^3}\ddot{\mathbf{r}}^2(t), \qquad \ddot{\mathbf{r}}(t) \equiv \frac{d^2\mathbf{r}}{dt^2}$$

Quantum mechanical analog:

$$P_{quan}(t) = \frac{2e^2}{3c^3}\left|\frac{d^2}{dt^2}\left\langle\psi(\mathbf{r},t)\left|\hat{\mathbf{r}}\right|\psi(\mathbf{r},t)\right\rangle\right|^2$$

Time-dependent Schrödinger equation:

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t) = \left[\frac{\hat{\mathbf{p}}^2}{2m} + \hat{V}(\mathbf{r}) + e\mathbf{r}\cdot\mathbf{E}(t)\right]\psi(\mathbf{r},t)$$

Frequency domain (harmonic generation):

$$I_{quan}(\Omega) \propto \left|\frac{1}{\tau}\int_0^\tau dt e^{-i\Omega t}\frac{d^2}{dt^2}\left\langle\psi(\mathbf{r},t)\left|\hat{\mathbf{r}}\right|\psi(\mathbf{r},t)\right\rangle\right|^2$$

τ is the duration of the laser pulse.

Atoms in monochromatic, LP-light

$$\hat{\mathcal{H}}_f(\mathbf{r},t) = \frac{\hat{\mathbf{p}}^2}{2m} + V(r) + eE_0 x \cos(\omega t) - i\hbar\frac{\partial}{\partial t}$$

$$\left[\hat{\mathcal{H}}_f, \hat{P}_2\right] = 0, \qquad \hat{P}_2 = \left(x \to -x,\ t \to t + \frac{T}{2}\right)$$

$$\boxed{\hat{P}_2 \Phi_{\varepsilon,p} = e^{i\pi p}\Phi_{\varepsilon,p}, \quad p = 0, 1.}$$

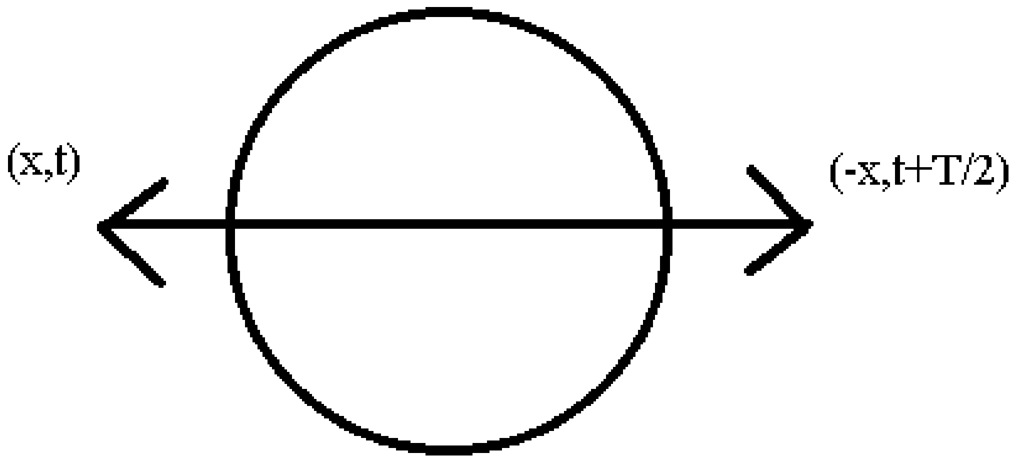
Assume that the atomic–field state is described by a Floquet state:

The **n**-th harmonic intensity (matrix element):

$$\begin{aligned} I_\varepsilon^{(\mathbf{n})} &\propto \left| \frac{1}{T}\int_0^T e^{-i\mathbf{n}\omega t}\frac{d^2}{dt^2}\langle \Phi_{\varepsilon,p}|x|\Phi_{\varepsilon,p}\rangle \right|^2 \\ &= (\mathbf{n}\omega)^4 \left| \frac{1}{T}\int_0^T e^{-i\mathbf{n}\omega t}\langle \Phi_{\varepsilon,p}|x|\Phi_{\varepsilon,p}\rangle \right|^2 \\ &\equiv (\mathbf{n}\omega)^4 \left| \langle\langle \Phi_{\varepsilon,p}|x e^{-i\mathbf{n}\omega t}|\Phi_{\varepsilon,p}\rangle\rangle \right|^2 \end{aligned}$$

$$\langle\langle \ldots \rangle\rangle = \frac{1}{T}\int_0^T dt \int_{-\infty}^{\infty} d\mathbf{r}$$

Sambe's and Howland's extended Hilbert space.


(x,t)
(-x,t+T/2)

DS analysis:

$$\langle\langle \Phi_{\varepsilon,p} | x e^{-i\mathbf{n}\omega t} | \Phi_{\varepsilon,p} \rangle\rangle =$$

$$= \langle\langle \Phi_{\varepsilon,p} | \left(\widehat{P}_2^{-1} \widehat{P}_2 \right) x e^{-i\mathbf{n}\omega t} \left(\widehat{P}_2^{-1} \widehat{P}_2 \right) | \Phi_{\varepsilon,p} \rangle\rangle =$$

$$= \langle\langle \widehat{P}_2 \Phi_{\varepsilon,p} | \widehat{P}_2 x e^{-i\mathbf{n}\omega t} \widehat{P}_2^{-1} | \widehat{P}_2 \Phi_{\varepsilon,p} \rangle\rangle =$$

$$= \langle\langle \Phi_{\varepsilon,p} | \widehat{P}_2 x e^{-i\mathbf{n}\omega t} \widehat{P}_2^{-1} | \Phi_{\varepsilon,p} \rangle\rangle \neq 0 \Longrightarrow$$

$$\Longrightarrow \widehat{P}_2 x e^{-i\mathbf{n}\omega t} \widehat{P}_2^{-1} = x e^{-i\mathbf{n}\omega t}$$

$$x e^{-i\mathbf{n}\omega t} \overset{?}{=} (-x) e^{-i\mathbf{n}\omega (t+T/2)} \overset{?}{=} (-1)^{\mathbf{n}+1} x e^{-i\mathbf{n}\omega t} \Longrightarrow$$

n is odd

All harmonics are linearly polarized as the incident field is.

Atoms in monochromatic, CP-light

$$\widehat{\mathcal{H}}_f(\mathbf{r},t) = \frac{\widehat{\mathbf{p}}^2}{2m} + V(r) + eE_0\rho\cos(\varphi - \omega t) - i\hbar\frac{\partial}{\partial t}$$

$$\left[\widehat{\mathcal{H}}_f, \widehat{P}_\infty\right] = 0,\ \widehat{P}_\infty = (\varphi \to \varphi + \delta\varphi,\ t \to t + \delta t)\,,\ \delta t = \frac{\delta\varphi}{\omega}$$

$$\boxed{\widehat{P}_\infty \Phi_{\mathbf{r},p}(\varphi,t) = e^{+ip\delta\varphi}\Phi_{\varepsilon,p}(\mathbf{r},t), \quad p \in \mathcal{Z}.}$$

Assume that the atomic–field state is described by a Floquet state:

The **n**-th harmonic intensity:

$$I^{(\mathbf{n})}_{\varepsilon,\pm} \propto \mathbf{n}^4 \left| \langle\langle \Phi_{\varepsilon,p} \left| \rho e^{\pm i\varphi} e^{-i\mathbf{n}\omega t} \right| \Phi_{\varepsilon,p} \rangle\rangle \right|^2$$

(D)SALC:

$$\{x, y\} \implies \left\{x \pm iy = \rho e^{\pm i\varphi}\right\}$$

DS Analysis:

$$\widehat{P}_\infty e^{\pm i\varphi} e^{-i\mathbf{n}\omega t} \widehat{P}_\infty^{-1} = e^{\pm i\varphi} e^{-i\mathbf{n}\omega t} \implies$$

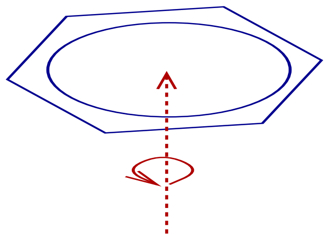
$$\implies \quad e^{-i\delta\varphi(\mathbf{n}\mp 1)} = 1 \quad \implies$$

n=1 only

(no high-order harmonics)

SR's for the HHGS by systems possessing the N-th order DS $\hat{P}_N$

The principle



A simple model

$$\hat{\mathcal{H}}_f(\mathbf{r},t) = \frac{\hat{\mathbf{p}}^2}{2m} + V_N(\varphi) + eE_0\rho\cos(\varphi - \omega t) - i\hbar\frac{\partial}{\partial t}$$

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

Assume that the molecular–field state is described by a Floquet state:

The **n**-th harmonic intensity:

$$I^{(n)}_{\varepsilon,\pm} \propto \mathbf{n}^4 \left| \langle\langle \Phi_\varepsilon \left| \rho e^{\pm i\varphi} e^{-i\mathbf{n}\omega t} \right| \Phi_\varepsilon \rangle\rangle \right|^2$$

$$\hat{P}_N e^{\pm i\varphi} e^{-i\mathbf{n}\omega t} \hat{P}_N^{-1} = e^{\pm i\varphi} e^{-i\mathbf{n}\omega t} \implies e^{-i\frac{2\pi}{N}(\mathbf{n}\mp 1)} = 1$$

$$\boxed{\mathbf{n} = 1, N\pm 1, 2N\pm 1, 3N\pm 1, \ldots}$$

"+"–harmonics are polarized as the incident field,

"-"–harmonics are polarized in the opposite direction.

Validity in the many-electron case

The Floquet Hamiltonian:

$$\widehat{\mathcal{H}}_{f,M}(\bar{\mathbf{r}},t) = \sum_{i=1}^{M} \widehat{H}(\mathbf{r}_i, t) + \sum_{i<j=1}^{M} \frac{e^2}{\left|\mathbf{r}_i - \mathbf{r}_j\right|} - i\hbar\frac{\partial}{\partial t},$$

$\bar{\mathbf{r}} \equiv (\mathbf{r}_1, \ldots, \mathbf{r}_M)$.

The permutation and DS groups:

M-electron permutation group: $\mathcal{S}_M$;

$$\widehat{P}_{N,M} = \left(\bar{\varphi} \to \bar{\varphi} + \frac{2\pi}{N}, t \to t + \frac{T}{N}\right),$$

$\bar{\varphi} \equiv (\varphi_1, \ldots, \varphi_M)$.

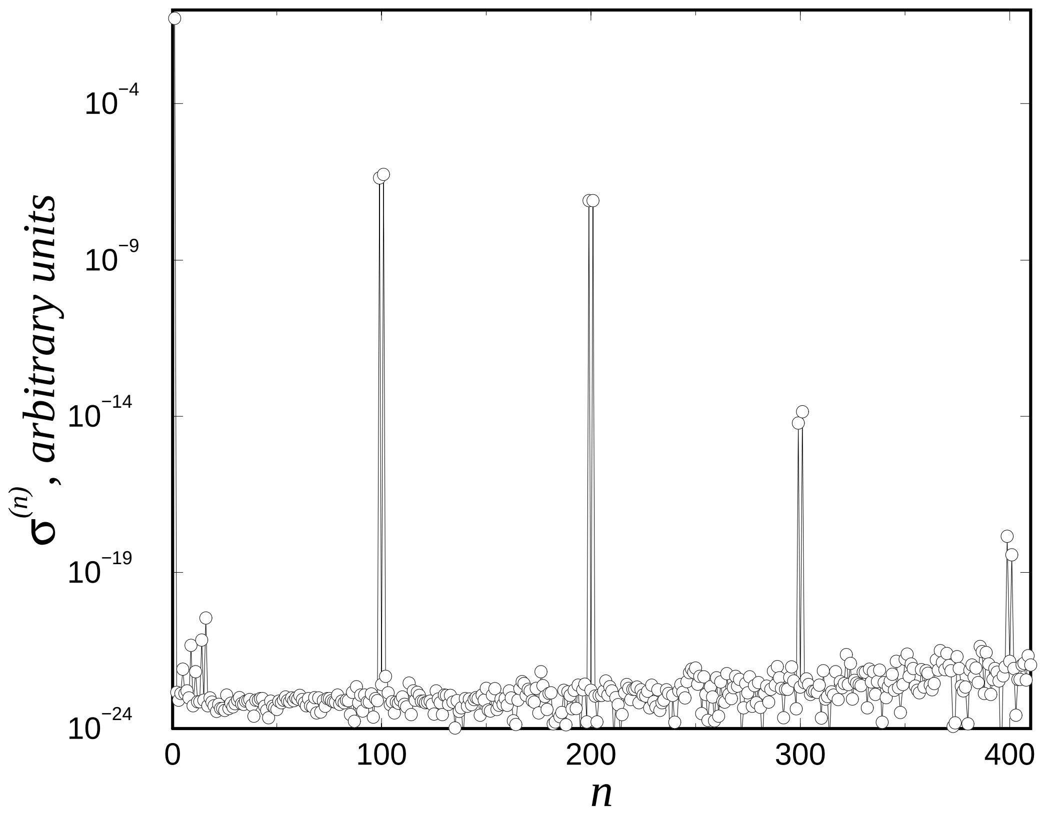
The $\mathbf{n}$-th harmonic intensity:

$$I^{(\mathbf{n})}_{\varepsilon,\pm,M} \propto \mathbf{n}^4 \left| \left\langle\!\!\left\langle \Phi_{\varepsilon,p,M} \left| \left(\sum_{j=1}^{M} \rho_j e^{\pm i\varphi_j} \right) e^{-i\mathbf{n}\omega t} \right| \Phi_{\varepsilon,p,M} \right\rangle\!\!\right\rangle \right|^2.$$

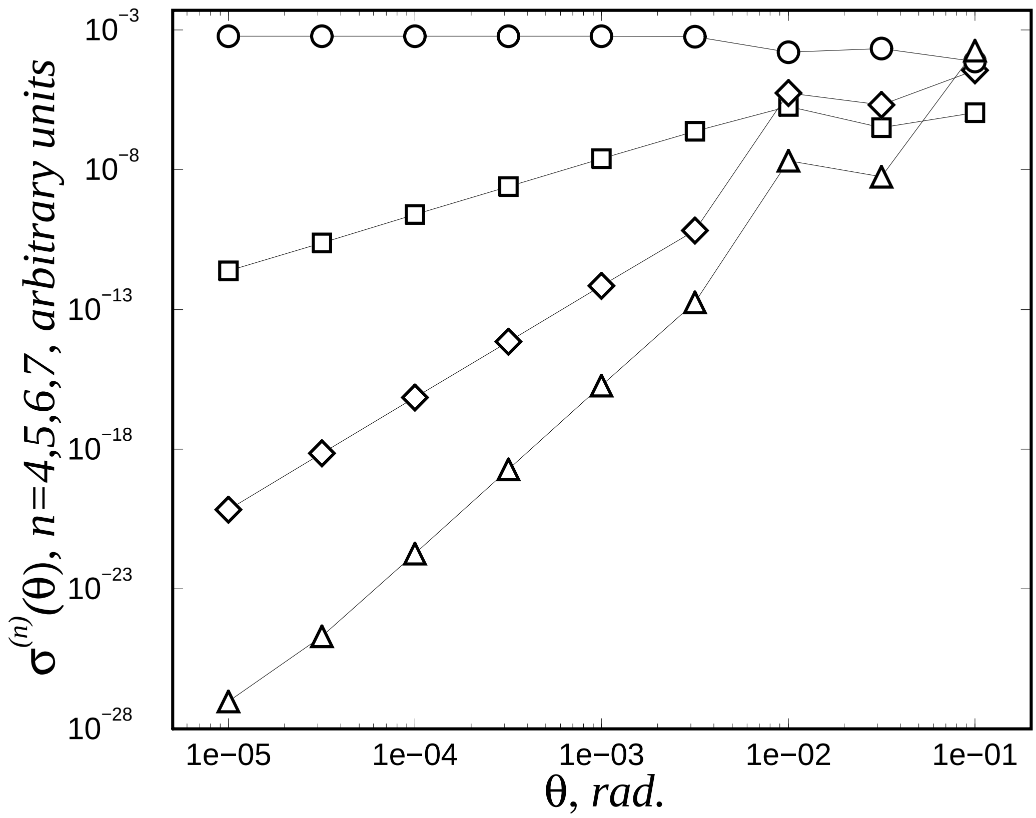
$\Longrightarrow$

Identical SR's.

A simple numerical model



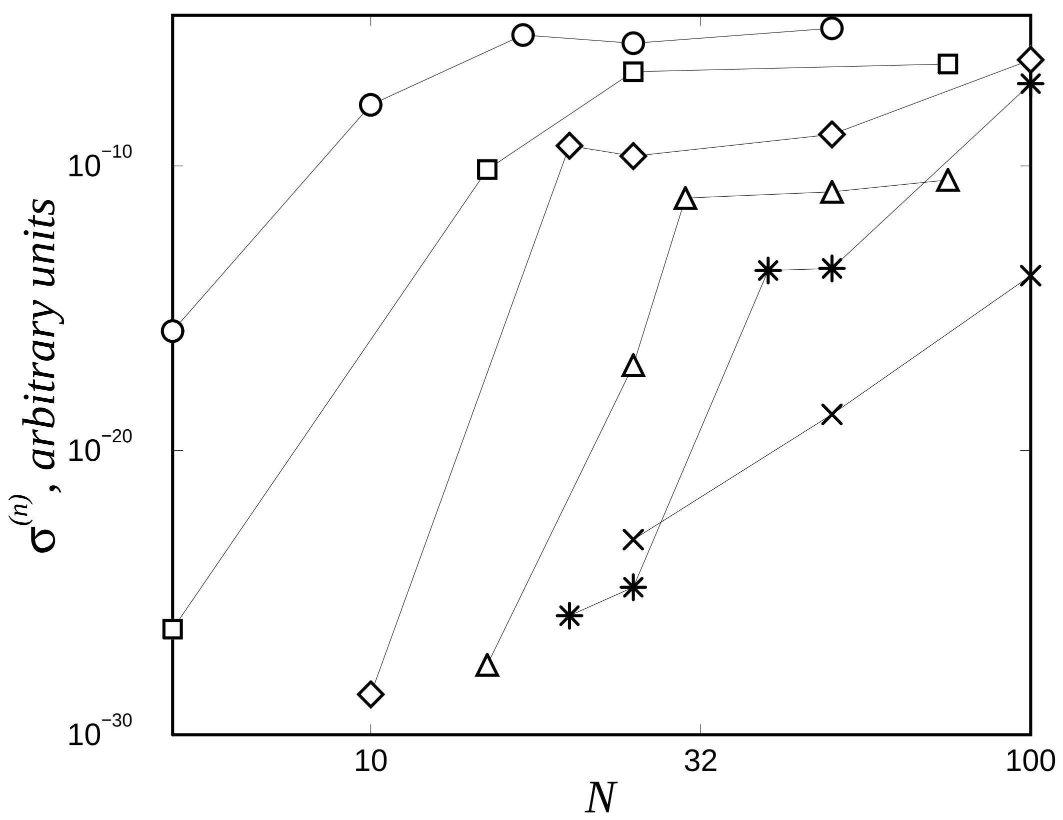
HHGS for a model of $N = 100$ atoms placed equidistantly on a circle and exposed to monochromatic circularly-polarized field of the intensity $1.8 \times 10^{13}\ \frac{W}{cm^2}$ and frequency $\omega = 0.02$ a.u. ($\lambda \approx 2.22\mu m$). The first high-order harmonic has the frequency of $99\omega \approx 2$ a.u..



Stability plot for a cluster with $N = 7$ atoms of the probability to generate the symmetry-allowed 6-th harmonic and the 4-th, 5-th and 7-th (symmetry-forbidden) harmonics. Note that $\sigma^{(6)}$ (circles) is more dominant by several orders of magnitude than that of the symmetry-forbidden harmonics for $\theta < 0.1$ rad.; $\sigma^{(4)}$ (squares) varies initially as θ^2; $\sigma^{(5)}$ (diamonds) varies initially as θ^4; and $\sigma^{(7)}$ (triangles) varies initially as θ^6.

$$I_s^{\text{ellipticity}}\,[N \pm (2s+1)] \;\propto\; \theta^{2s} I_0^{\text{ellipticity}}\,(N \pm 1)\,,$$

$$I_s^{\text{tilt}}\,[N \pm (2s+1)] \;\propto\; \theta^{4s} I_0^{\text{tilt}}\,(N \pm 1)\,,$$



The probability to get the $\mathbf{n}$-th harmonic as function of the number N of "atoms" which are placed equidistantly on a circle (a log-log plot). Circles - the 51-st harmonic (the 50-th for $N = 17$); squares - the 76-th harmonic; diamonds - the 101-st harmonic; triangles - the 151-st harmonic; stars - the 201-st harmonic; $\times$'s - the 301-st harmonic. As N increases the higher harmonics are amplified.

The results presented above show that the DS-based SR's enable one to design a system (a target and a time-dependent electric field) which filters out the selected high-order harmonics only and even amplifies them.

Note however...

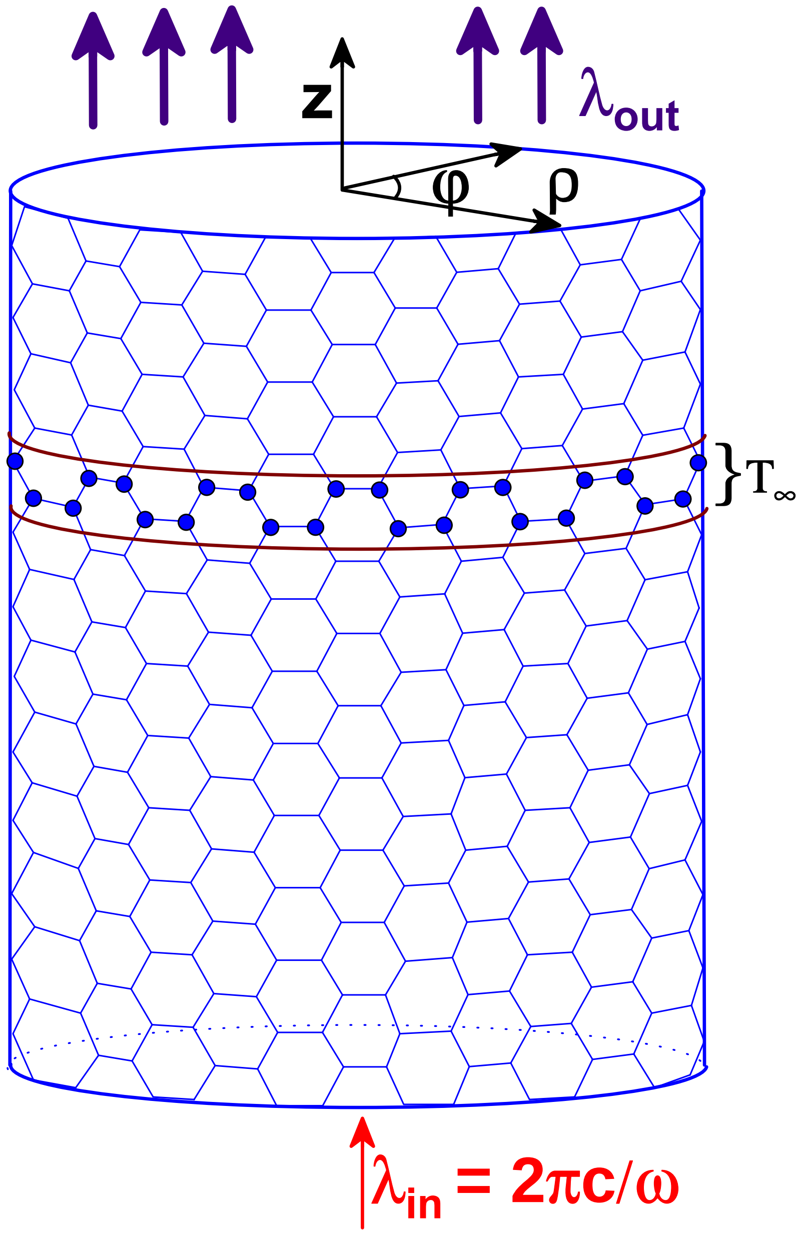
- If one wishes to design a system the lowest high-order harmonic of which is, say, the 100-th, it is required to find a molecular (or, an artificial) system which possesses the 101-fold rotational axis...

The next step:

- Take spatially extended targets, such as carbon nanotubes (CNT's), in order to exploit the DS resulting from screw-axis symmetry.

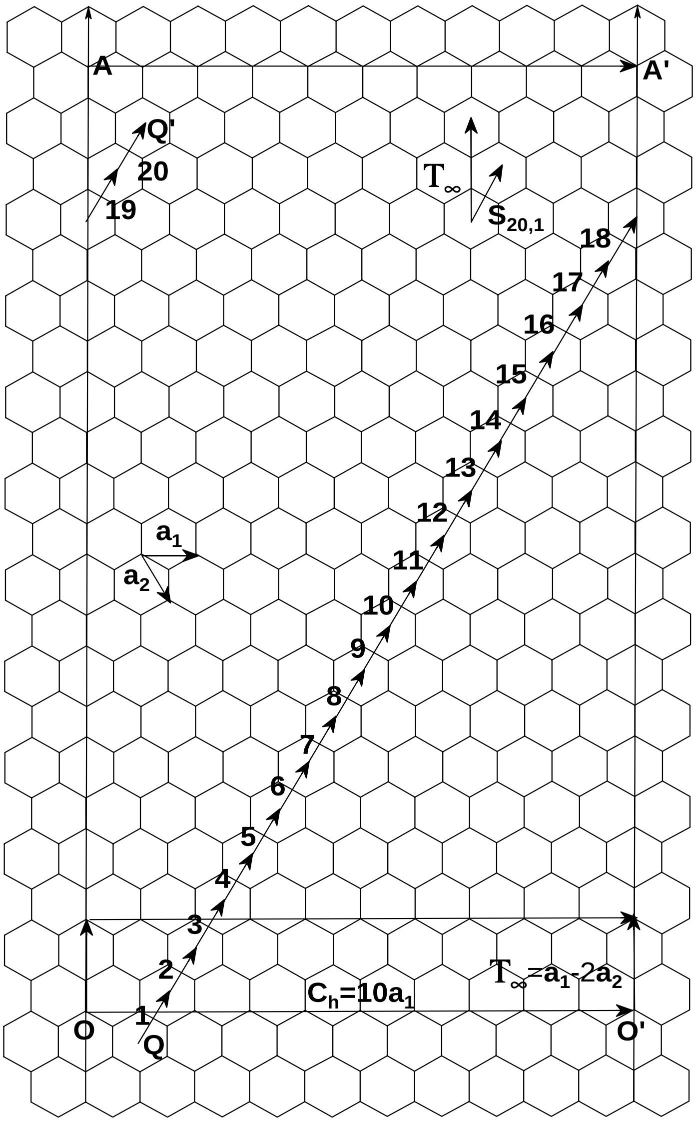
For very high-order harmonics and/or thick targets, the dipole approximation is not valid.
$\Longrightarrow$
Formulate expression for the $\mathbf{n}$-th harmonic intensity *beyond* the dipole approximation.

HHG by CNT's



(10,0)-zigzag CNT

diameter $= 0.793\,nm$



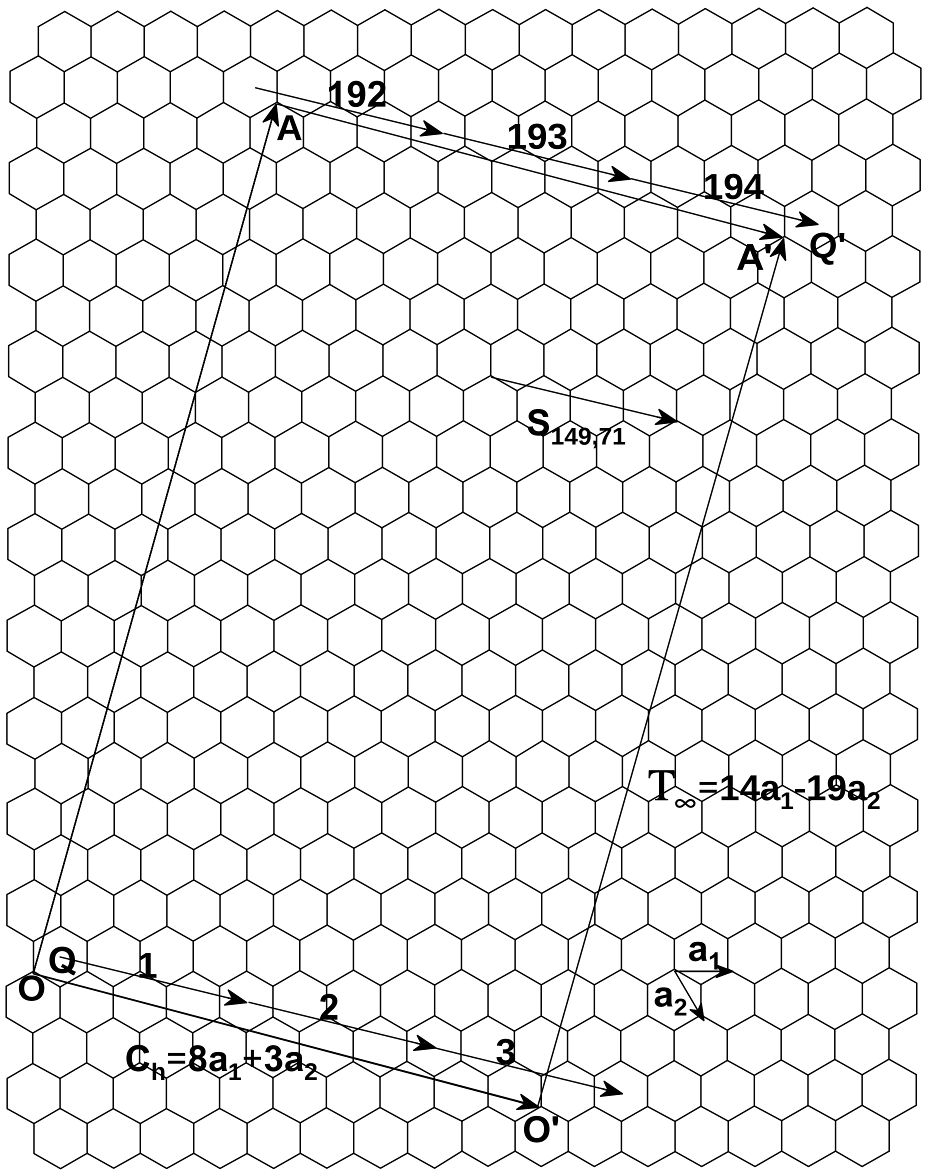
Screw axis:

$$S_{20} = \left(\varphi \to \varphi + \frac{2\pi}{20}, z \to z + \frac{T_z}{2}\right)$$

Order is 20

(8,3)-chiral CNT

diameter $= 0.781\, nm$



Screw axis:

$$S_{194} = \left(\varphi \to \varphi + \frac{71 \cdot 2\pi}{194},\, z \to z + \frac{T_z}{194}\right)$$

Order is 194

The n-th harmonic intensity

Classical electrodynamics:

$$I^{(\mathbf{n},\mathbf{z})} \propto \mathbf{n}^2 \left| \left(\mathbf{z} \times \mathbf{A}^{(\mathbf{n},\mathbf{z})} \right) \times \mathbf{z} \right|^2$$

$$\mathbf{A}^{(\mathbf{n},\mathbf{z})} \propto \int_0^\infty dt \int_{-\infty}^\infty d\mathbf{r}\, \mathbf{J}(\mathbf{r},t) e^{-i\mathbf{n}(\omega t - k_0 z)},$$

QM current density, $\mathbf{J}(\mathbf{r},t)$

$$\mathbf{A}_\varepsilon^{(\mathbf{n},\mathbf{z})} \propto \left\langle\!\!\left\langle \Phi_\varepsilon \left| \widehat{\mathbf{A}}^{(\mathbf{n},\mathbf{z})} \right| \Phi_\varepsilon \right\rangle\!\!\right\rangle + \left\langle\!\!\left\langle \Phi_\varepsilon^* \left| \left(\widehat{\mathbf{A}}^{(\mathbf{n},\mathbf{z})} \right)^{\dagger^*} \right| \Phi_\varepsilon^* \right\rangle\!\!\right\rangle$$

$$\widehat{\mathbf{A}}^{(\mathbf{n},\mathbf{z})} = \hat{\mathbf{v}} e^{-in(\omega t - k_0 z)}, \quad \left(\widehat{\mathbf{A}}^{(\mathbf{n},\mathbf{z})} \right)^{\dagger^*} = \widehat{\mathbf{v}^*} e^{-i\mathbf{n}(\omega t - k_0 z)},$$

$$\hat{\mathbf{v}} = \frac{1}{m} \left[\hat{\mathbf{p}} - \frac{e}{c} \widehat{\mathbf{A}}(\mathbf{r},t) \right]$$

(D)SALC:

$$\hat{A}_\pm^{(\mathbf{n},\mathbf{z})} \equiv \hat{A}_x^{(\mathbf{n},\mathbf{z})} \pm i \hat{A}_y^{(\mathbf{n},\mathbf{z})}, \quad \left(\hat{A}_\pm^{(\mathbf{n},\mathbf{z})} \right)^{\dagger^*} \equiv \left(\hat{A}_x^{(\mathbf{n},\mathbf{z})} \right)^{\dagger^*} \pm i \left(\hat{A}_y^{(\mathbf{n},\mathbf{z})} \right)^{\dagger^*}$$

$$\hat{A}_\pm^{(\mathbf{n},\mathbf{z})} = \left[\frac{\hbar}{i} e^{\pm i\varphi} \left(\frac{\partial}{\partial \rho} \pm \frac{i}{\rho} \frac{\partial}{\partial \varphi} \right) - \frac{e E_0}{\omega} e^{\pm i\alpha(\omega t - k_0 z)} \right] e^{-in(\omega t - k_0 z)}$$

$$\left(\hat{A}_\pm^{(\mathbf{n},\mathbf{z})} \right)^{\dagger^*} = \left[-\frac{\hbar}{i} e^{\pm i\varphi} \left(\frac{\partial}{\partial \rho} \pm \frac{i}{\rho} \frac{\partial}{\partial \varphi} \right) - \frac{e E_0}{\omega} e^{\pm i\alpha(\omega t - k_0 z)} \right] e^{-i\mathbf{n}(\omega t - k_0 z)}$$

DS analysis $\Longrightarrow$ SR's

$$\widehat{P}_{\infty}\left\{\widehat{A}_{\pm}^{(\mathbf{n},\mathbf{z})};\left(\widehat{A}_{\pm}^{(\mathbf{n},\mathbf{z})}\right)^{\dagger^*}\right\}\widehat{P}_{\infty}^{-1}\stackrel{?}{=}\left\{\widehat{A}_{\pm}^{(\mathbf{n},\mathbf{z})};\left(\widehat{A}_{\pm}^{(\mathbf{n},\mathbf{z})}\right)^{\dagger^*}\right\}\Longrightarrow$$

$$\mathbf{n}=1,2,3,4,\ldots$$

$$\widehat{P}_{N}\left\{\widehat{A}_{\pm}^{(\mathbf{n},\mathbf{z})};\left(\widehat{A}_{\pm}^{(\mathbf{n},\mathbf{z})}\right)^{\dagger^*}\right\}\widehat{P}_{N}^{-1}\stackrel{?}{=}\left\{\widehat{A}_{\pm}^{(\mathbf{n},\mathbf{z})};\left(\widehat{A}_{\pm}^{(\mathbf{n},\mathbf{z})}\right)^{\dagger^*}\right\}\Longrightarrow$$

$$\mathbf{n}=1,N\pm1,2N\pm1,3N\pm1,\ldots$$

"+"–harmonics are polarized as the incident field,
"-"–harmonics are polarized in the opposite direction.

$(10,0)$-zigzag $\rightarrow \mathbf{n}=1,19,21,39,41\ldots$

e.g. $\lambda=200nm\Longrightarrow$ 10.5 nm...

$(8,3)$-chiral $\rightarrow \mathbf{n}=1,193,195,387,389\ldots$

$\lambda=200nm\Longrightarrow\approx$1 nm...

Estimation of the HHG intensities: A tight-binding-based model

A. Hückel approximation:

Take carbon $2p_z$ orbitals,

$$H_{i,j} = \begin{cases} \alpha & i = j \\ \beta & i = j \pm 1 \text{ (neighbours)} \\ 0 & \text{(otherwise)} \end{cases}$$

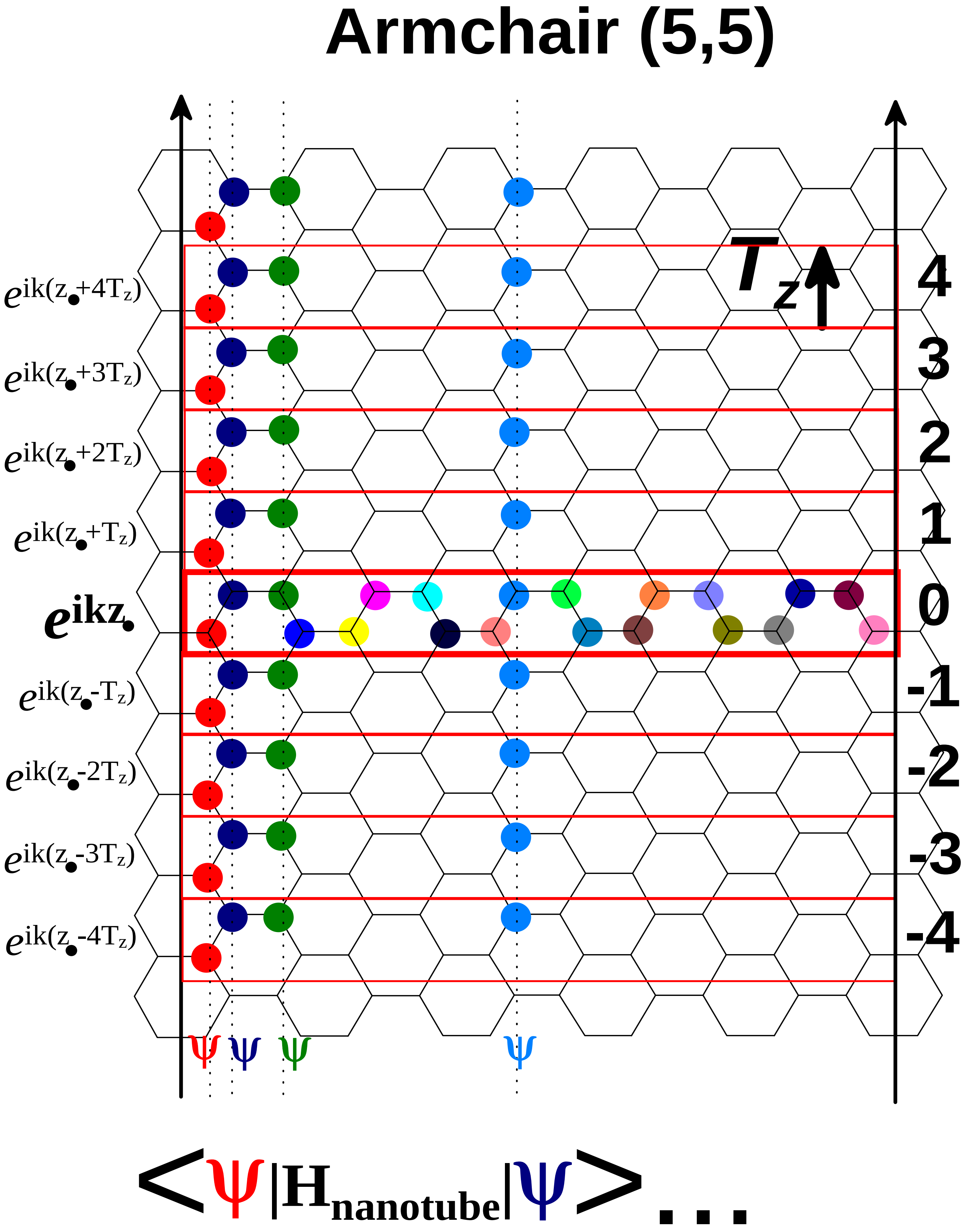
$\beta \approx 2.7$eV [J. W. G. Wildöer, L. C. Venema, A. G. Rinzler, R. E. Smalley and C. Dekker, *Nature* **391**, 59-62 (1998)].

B. Bloch wave-functions:

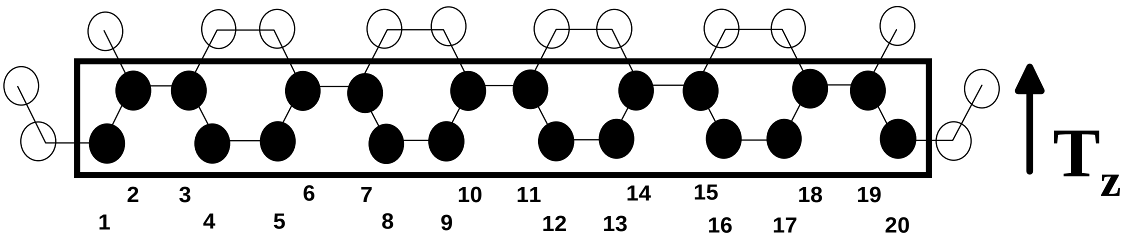
Periodicity along the z-axis $\Longrightarrow$

$$\psi_k(z) \equiv e^{ikz}\phi(z)\,, \quad \phi(z) = \phi(z + T_z)$$

How to combine them for nanotubes?



Armchair (5,5)
T_z
4
3
2
1
0
-1
-2
-3
-4
$e^{ik(z_\bullet+4T_z)}$
$e^{ik(z_\bullet+3T_z)}$
$e^{ik(z_\bullet+2T_z)}$
$e^{ik(z_\bullet+T_z)}$
$e^{ikz_\bullet}$
$e^{ik(z_\bullet-T_z)}$
$e^{ik(z_\bullet-2T_z)}$
$e^{ik(z_\bullet-3T_z)}$
$e^{ik(z_\bullet-4T_z)}$
ψ ψ ψ ψ
$<\psi|H_{nanotube}|\psi>\ldots$



Constructing the basis functions:

The positions of the carbon atoms in the 0-th unit cell

$$\varphi_p^{(0)} = \left\{ \begin{array}{l} \frac{2\pi}{30} + \frac{p-1}{4}\frac{2\pi}{5} : p = 1,5,9,13,17 \\ \frac{4\pi}{30} + \frac{p-2}{4}\frac{2\pi}{5} : p = 2,6,10,14,18 \\ \frac{8\pi}{30} + \frac{p-3}{4}\frac{2\pi}{5} : p = 3,7,11,14,19 \\ \frac{10\pi}{30} + \frac{p-4}{4}\frac{2\pi}{5} : p = 4,8,12,16,20 \end{array} \right\},$$

$$z_p^{(0)} = \left\{ \begin{array}{l} \frac{T_z}{4} : p = 1,4,5,8,9,\ldots,20 \\ \frac{3T_z}{4} : p = 2,3,6,7,10,\ldots,19 \end{array} \right\}.$$

$$\psi_p^k(\rho_0, \varphi, z) = \sum_{q=-\infty}^{\infty} e^{+ikz_{p,q}} \cdot \psi_{2p_z}\Big(\rho_0, \varphi - \varphi_p^{(0)}, z - z_{p,q}\Big)$$

$$z_{p,q} = z_p^{(0)} + qT_\infty, \quad p = 1,2,\ldots,20(2N), \; q \in \mathcal{Z}$$

$$-\frac{\pi}{T_z} \le k < \frac{\pi}{T_z}$$

The matrix elements of the field-free Hamiltonian

$$\mathrm{H}_{\mathrm{CNT}}^{(5,5)}(k) = \beta \begin{pmatrix} 0 & b & 0 & 0 & 0 & \cdots & 0 & 1 \\ b & 0 & 1 & 0 & 0 & \cdots & 0 & 0 \\ 0 & 1 & 0 & b & 0 & \cdots & 0 & 0 \\ 0 & 0 & b & 0 & 1 & \cdots & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & 0 & \cdots & 0 & b \\ 1 & 0 & 0 & 0 & 0 & \cdots & b & 0 \end{pmatrix}$$

$$T_z^{(5,5)} = 0.249\, nm, \quad b = b(k) = 2\cos\left(\frac{kT_z}{2}\right)$$

$$\left\langle \psi_{2p_z} \left| \hat{\mathrm{H}}_{\mathrm{nanotube}}^{(5,5)} \right| \psi_{2p_z} \right\rangle = 0.$$

C. Dipole approximation:

$$T_z \gg \frac{2\pi}{k_0}$$

$$\hat{H}_{\mathrm{interaction}}^{\mathrm{dip.}} = eE_0\rho_0 \cos\left(\omega t - \varphi\right)$$

(length gauge)

$$\left\langle \psi_p^k \left| \frac{1}{2}\left(e^{i\omega t}e^{i\varphi} + e^{-i\omega t}e^{-i\varphi}\right) \right| \psi_{p'}^{k'} \right\rangle = \frac{1}{2}\left(e^{i\omega t}e^{i\varphi_p^{(0)}} + e^{-i\omega t}e^{-i\varphi_p^{(0)}}\right)\delta_{k,k'}\delta_{p,p'}.$$

$\Longrightarrow$ The interaction term is diagonal.

Solving the Floquet eigenvalue problem

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

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

Approach I.:

Diagonalizing the Floquet Hamiltonian matrix (basis functions):

Spatial:

$$\psi_p^k\left(\rho_0,\varphi,z\right), \quad p = 1,2,\ldots,20(2N), \quad -\frac{\pi}{T_z} \le k < \frac{\pi}{T_z},$$

Temporal:

$$\frac{1}{\sqrt{T}}e^{+il\omega t}, \;\; l \in \mathcal{Z}, \quad (l = -Q, -Q+1, \ldots, 0, \ldots, Q-1, Q).$$

Approach II.:

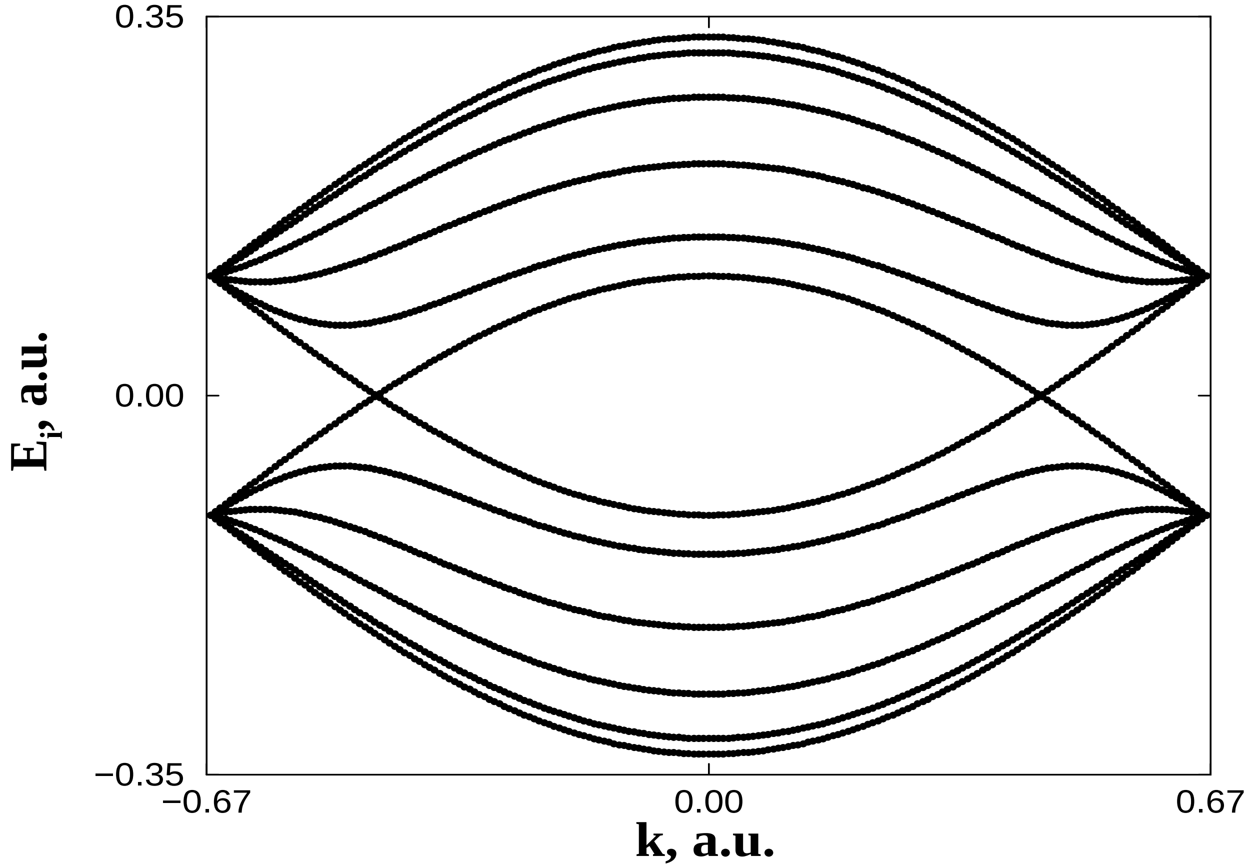
Diagonalizing the time-evolution operator for $t = T$ and propagating in time from 0 to T:

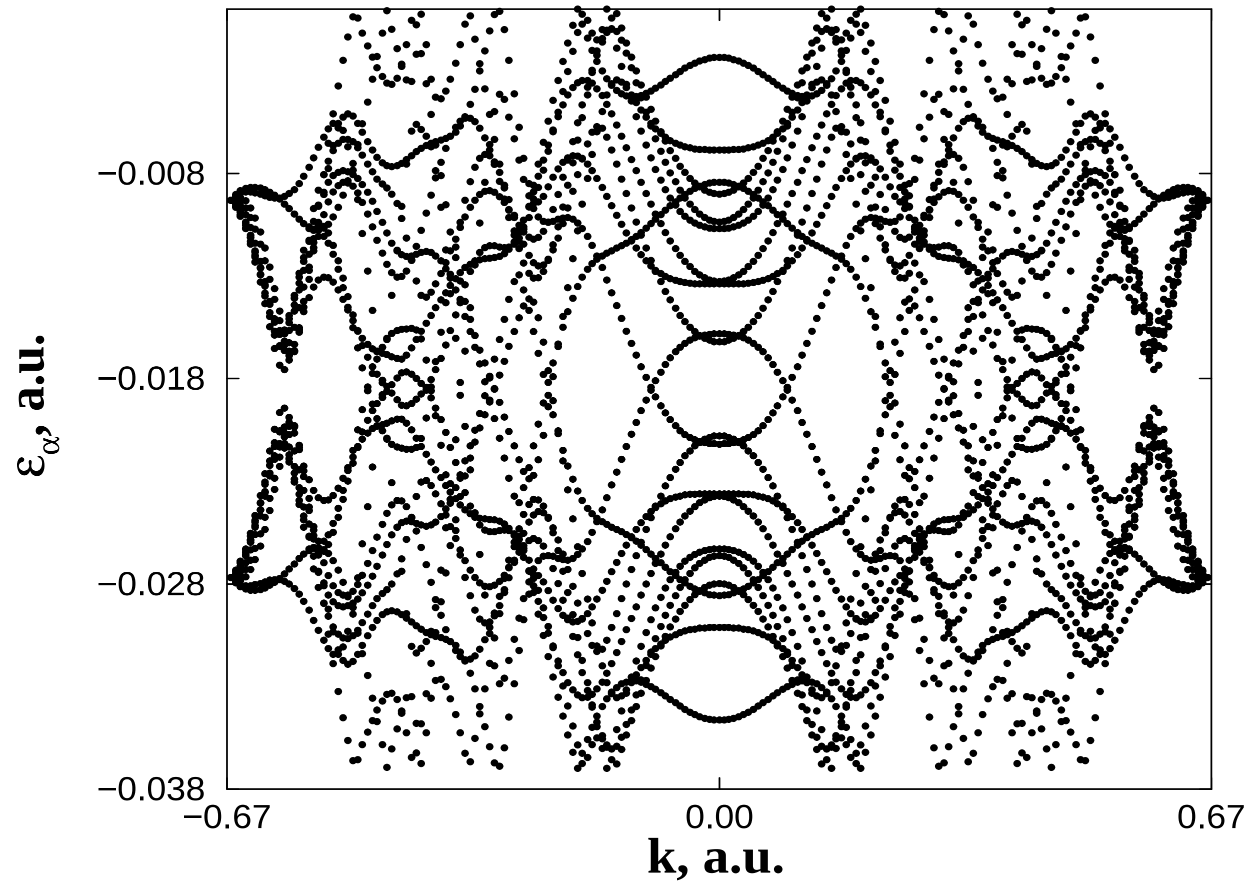
$$\widehat{U}(0 \to t) = \sum_{l=-\infty}^{l=+\infty} e^{+il\omega t}\left[\frac{1}{T}\int_0^T dt' e^{-il\omega t'} e^{-i\widehat{\mathcal{H}}_f(t')\cdot t/\hbar}\right]$$

$$\widehat{U}(0 \to T)\Psi_\varepsilon(0) = e^{-\frac{i}{\hbar}\varepsilon T}\Psi_\varepsilon(0)$$

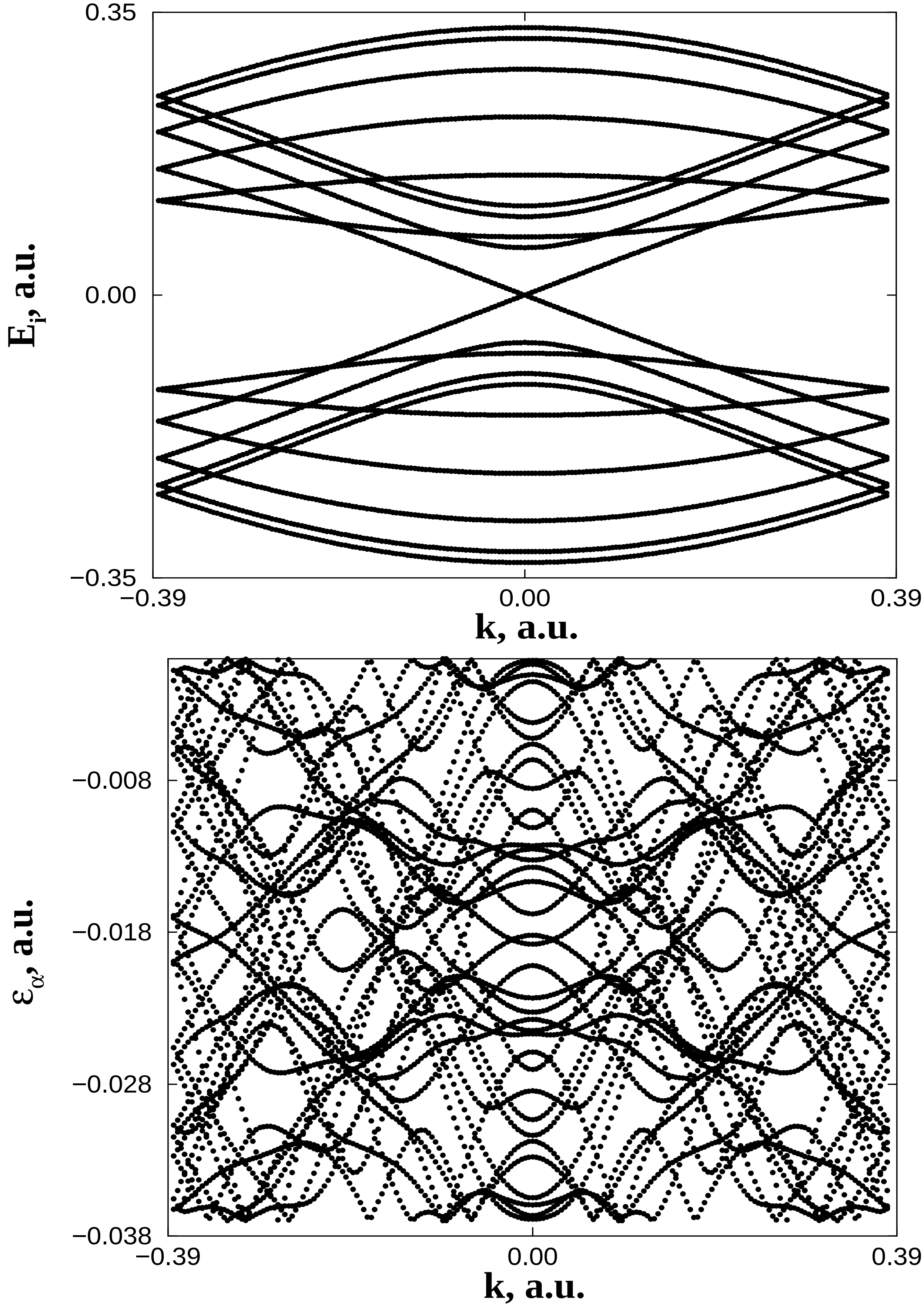
$$\widehat{U}(0 \to t)\Psi_\varepsilon(0) = \Psi_\varepsilon(t), \quad 0 \le t \le T.$$

EB's and QEB's of the (5, 5)-armchair CNT

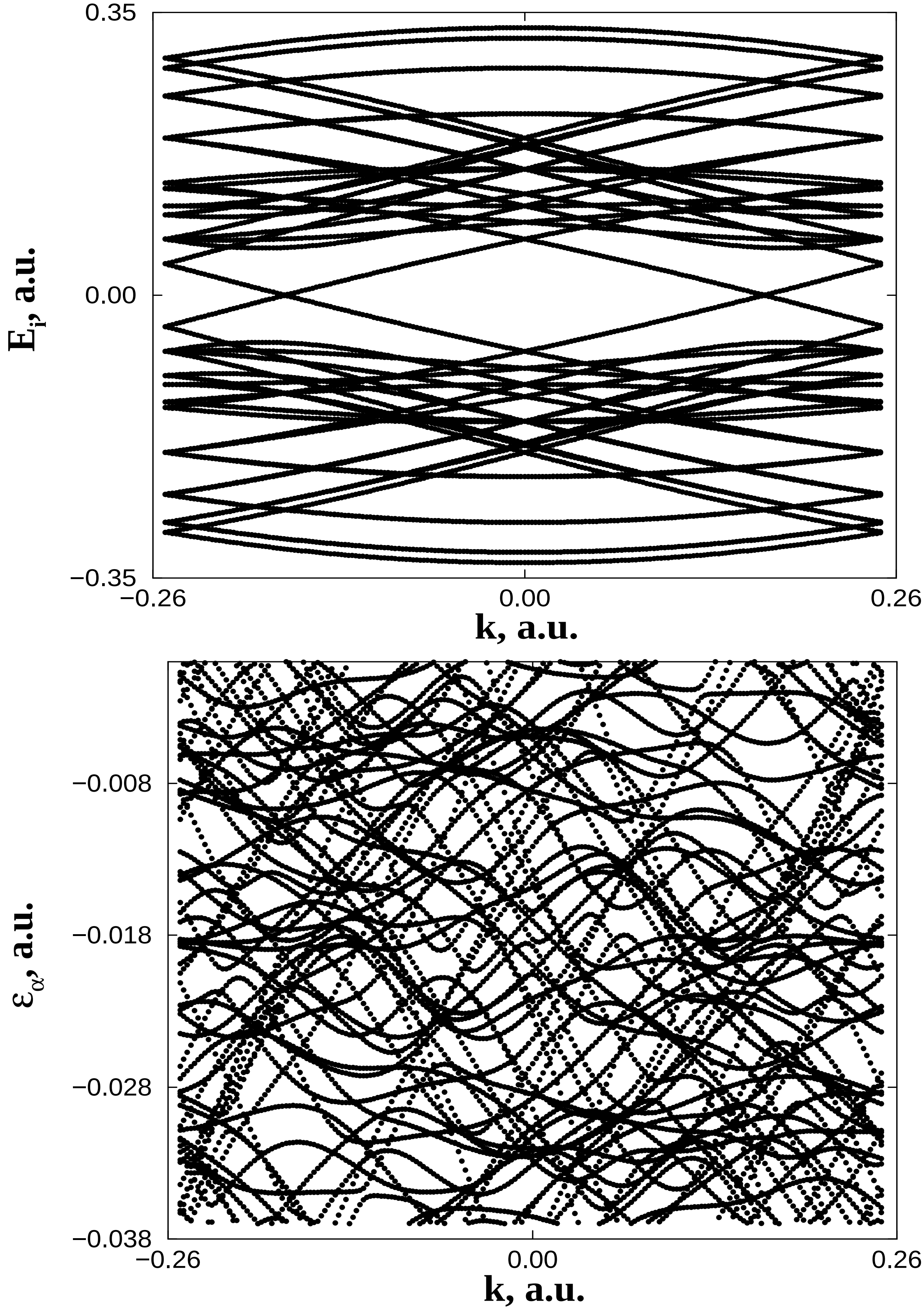




EB's and QEB's of the (9,0)-zigzag CNT



EB's and QEB's of the (8, 2)-chiral CNT



HHG from electrons up to $\mathcal{E}_{\text{Fermi}}$

The FB states:

$$\Phi_\alpha^k(t) = \sum_{p=1}^{20(2N)} C_{\alpha,p}^k(t)\psi_p^k; \ \varepsilon_\alpha(k), \quad \alpha = 1, 2, \ldots, 20(2N).$$

single-electron contribution:

$$d_{\pm,k}^{(\alpha)}(\mathbf{n}) = \left\langle\!\left\langle \Phi_\alpha^k(t) \left| e\rho_0 e^{\pm i\varphi} e^{-i\mathbf{n}\omega t} \right| \Phi_\alpha^k(t) \right\rangle\!\right\rangle$$

multi-electron summation:

$$d_\pm(\mathbf{n}) = \frac{T_z}{2\pi N} \sum_{\alpha=1}^{N} \int_{k=-\frac{\pi}{T_z}}^{\frac{\pi}{T_z}} dk \ d_{\pm,k}^{(\alpha)}(\mathbf{n})$$

$(5,5)-$armchair : $\quad \mathbf{n} = 1, 9, 11, 19, 21, \ldots$

$(9,0)-$zigzag : $\quad \mathbf{n} = 1, 17, 19, 35, 37, \ldots$

$(8,2)-$chiral : $\quad \mathbf{n} = 1, 27, 29, \ldots,$

Main results

$E_0 = 0.05$ a.u. ($\approx 9 \times 10^{13}$ W/cm^2),
$\omega = 0.037$ a.u. ($\lambda \approx 1.2 \mu m$).

The intensities:

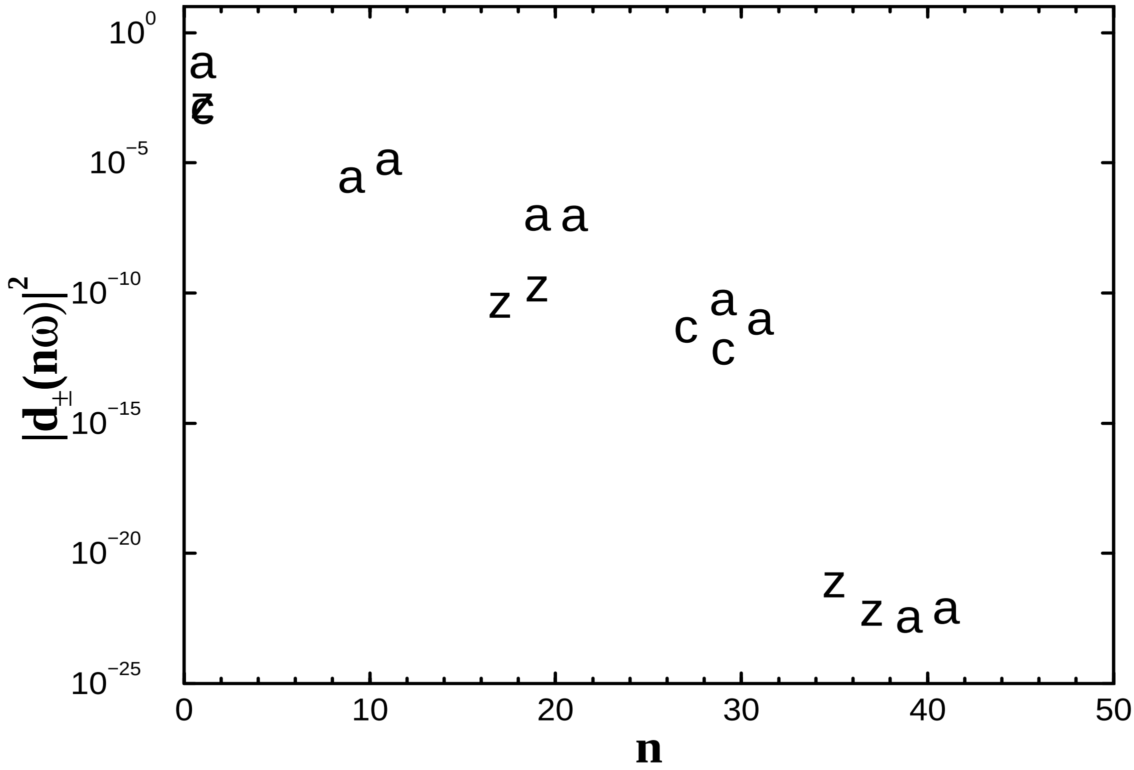
$$(5,5)-\text{armchair}: \quad |d_\pm(\mathbf{n})|^2 \approx 10^{-5} - 10^{-7}\text{a.u.}*$$
$$(9,0)-\text{zigzag}: \quad |d_\pm(\mathbf{n})|^2 \approx 10^{-10}\text{a.u.}$$
$$(8,2)-\text{chiral}: \quad |d_\pm(\mathbf{n})|^2 \approx 10^{-11} - 10^{-12}\text{a.u.}$$

* J. L. Krause, K. J. Schafer and K. C. Kulander, Phys. Rev. A **45**, 4998 (1992).

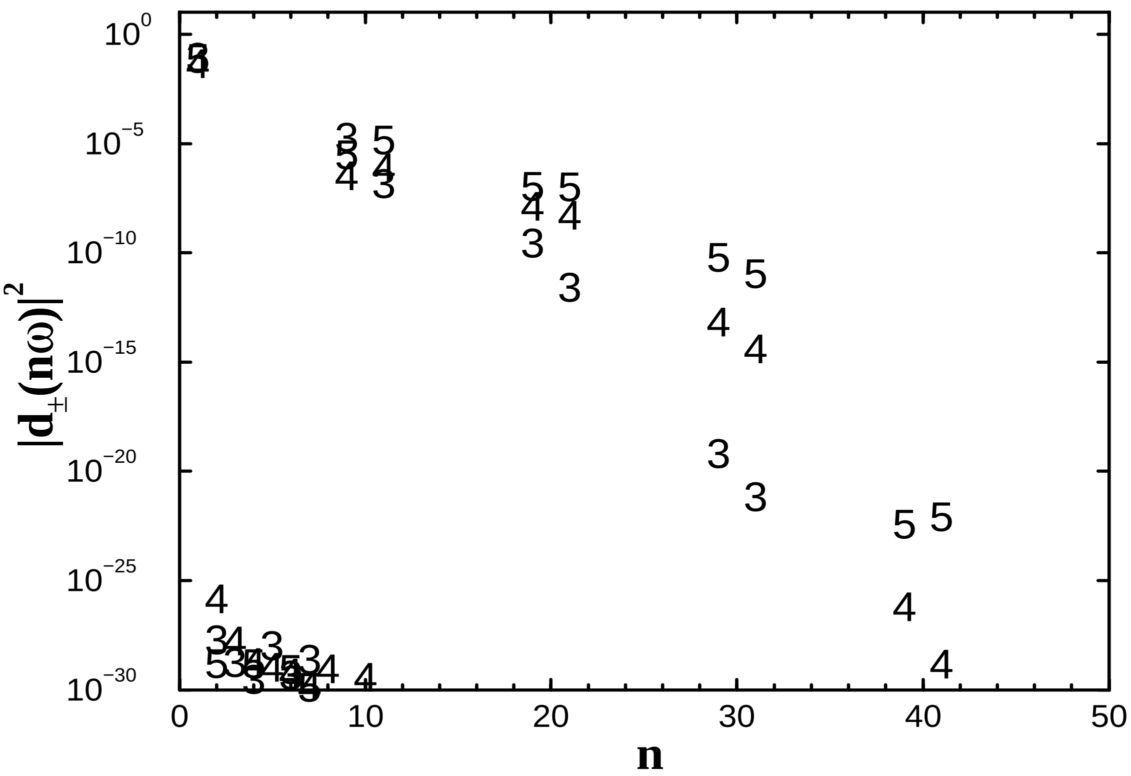
The cutoffs:

$$(5,5)-\text{armchair}: \quad \rho_0 \approx 6.5\,\text{a.u.}$$
$$(9,0)-\text{zigzag}: \quad \rho_0 \approx 6.7\,\text{a.u.}$$
$$(8,2)-\text{chiral}: \quad \rho_0 \approx 6.9\,\text{a.u.}$$

$$\mathbf{n}_{\text{cutoff}} = \frac{2eE_0\rho_0}{\hbar\omega} \approx 20$$



HHGS of the (5, 5)-armchair ("**a**"), the (9, 0)-zigzag ("**z**") and the (8, 2)-chiral ("**c**") CNT's. $E_0 = 0.05$ a.u. ($\approx 9 \times 10^{13}$ W/cm^2). $\omega = 0.037$ a.u. ($\lambda \approx 1.2 \mu m$).



HHGS of the $(5, 5)$-armchair CNT at the field strengths of $E_0 = 0.03$ a.u. ($\approx 3 \times 10^{13}$ W/cm^2), "**3**", $E_0 = 0.04$ a.u. ($\approx 6 \times 10^{13}$ W/cm^2), "**4**", $E_0 = 0.05$ a.u. ($\approx 9 \times 10^{13}$ W/cm^2), "**5**". $\omega = 0.037$ a.u. ($\lambda \approx 1.2 \mu m$).

Summary and conclusions

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

2. DS operations form groups. Consequently, DS analysis of Floquet states is analogous to symmetry analysis of stationary states. In particular, one can label QE's and their corresponding Floquet states with "good" quantum numbers, determine symmetry properties of QEB's and analyze whether non-accidental degeneracies can be found in the QEB spectra following the existence of unitary and anti-unitary DS's.

3. DS analysis of high-order harmonic generation processes in high-intensity laser fields has been placed on a firm foundation, analogously to that possessed by symmetry analysis in "conventional" spectroscopy.

4. In general, for 3D many-electron time-periodic Hamiltonians which are invariant under $\hat{P}_N$ only the $n = 1, N \pm 1, 2N \pm 1, 3N \pm 1, 4N \pm 1, \ldots$ harmonics are generated.

5. The SR's derived for *any* single-walled CNT's are given by $n = 1, N \pm 1, 2N \pm 1, 3N \pm 1, 4N \pm 1, \ldots$, where N is the number of hexagons in its unit cell. These SR's are invariant with respect to the wavelength of the incident circularly-polarized laser radiation, as well as to its helicity.

The considered (5, 5)-*armchair* CNT gives rise to the HHGS which is both selective and efficient enough to be of interest to experimentalists. The *chiral* CNT's are the first example of realistic physical systems for which all harmonics but very high-order ones are forbidden by symmetry.

The selective HHG by CNT's can be used for the structural analysis of the nanotubes samples. The relative strengths of the harmonics allowed for different CNT's can provide an information about the relative abundances of the various symmetry species in the sample.

We hope that the further development of present day technologies of growing arrays of CNT's parallel to each other and preparation of ropes containing CNT's of a single definite symmetry would allow the experimental verification of the above theory.

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