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Collapsed, Chiral, Single-Walled Carbon Nanotubes (SWCNTs) as Quasi-1D Electronic Systems

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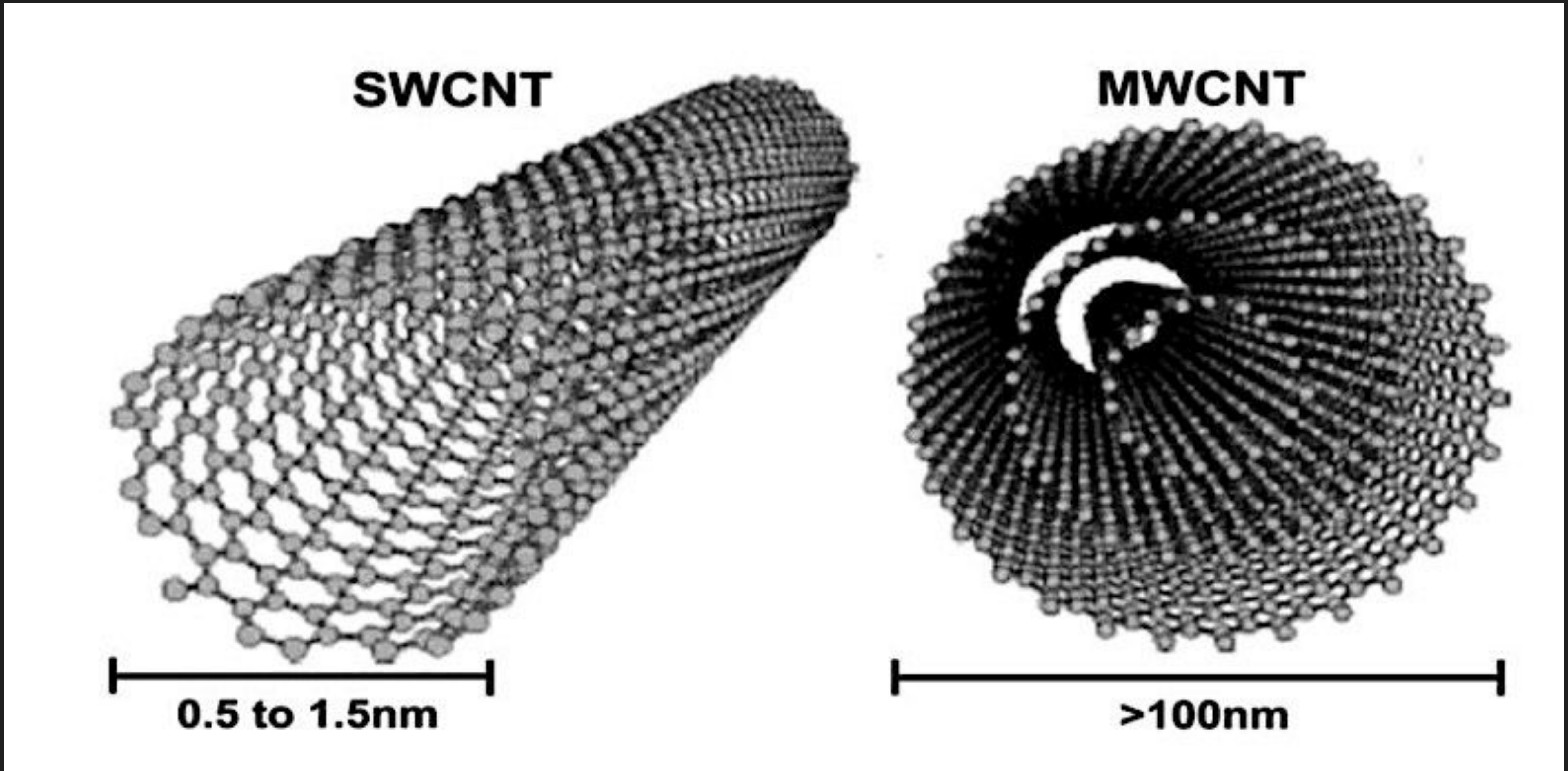
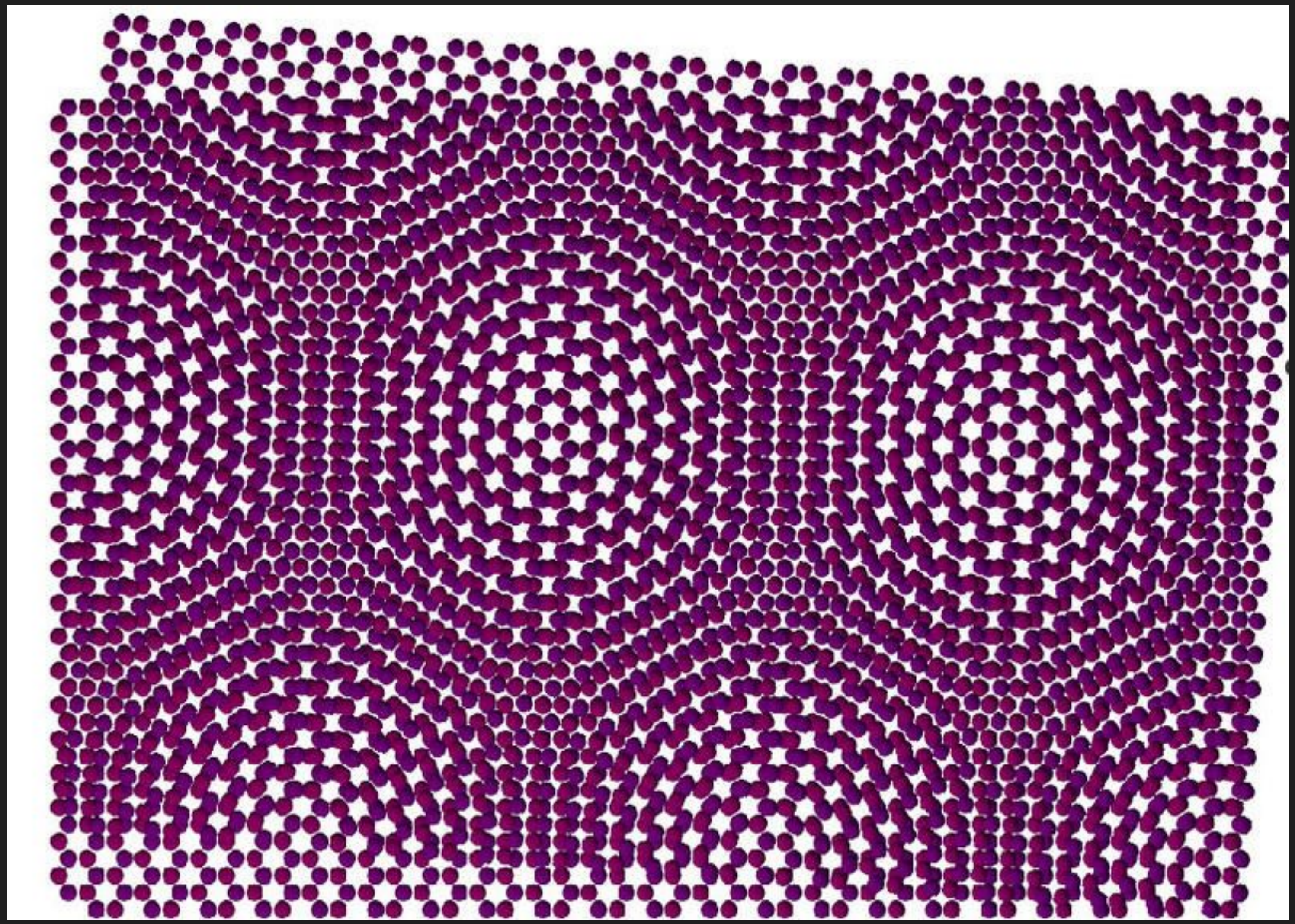
Context and motivation

- SWCNTs are essentially a rolled up sheet of monolayer graphene.
- Unique structure and electronic properties.



Ribeiro, B., Botelho, E., Costa, M. L., & Bandeira, C. F. (2017). Carbon nanotube buckypaper reinforced polymer composites: A review. Polímeros: Ciência e Tecnologia, 27(Ahead), e2017042.

Twisted Bilayer Graphene (TBG).



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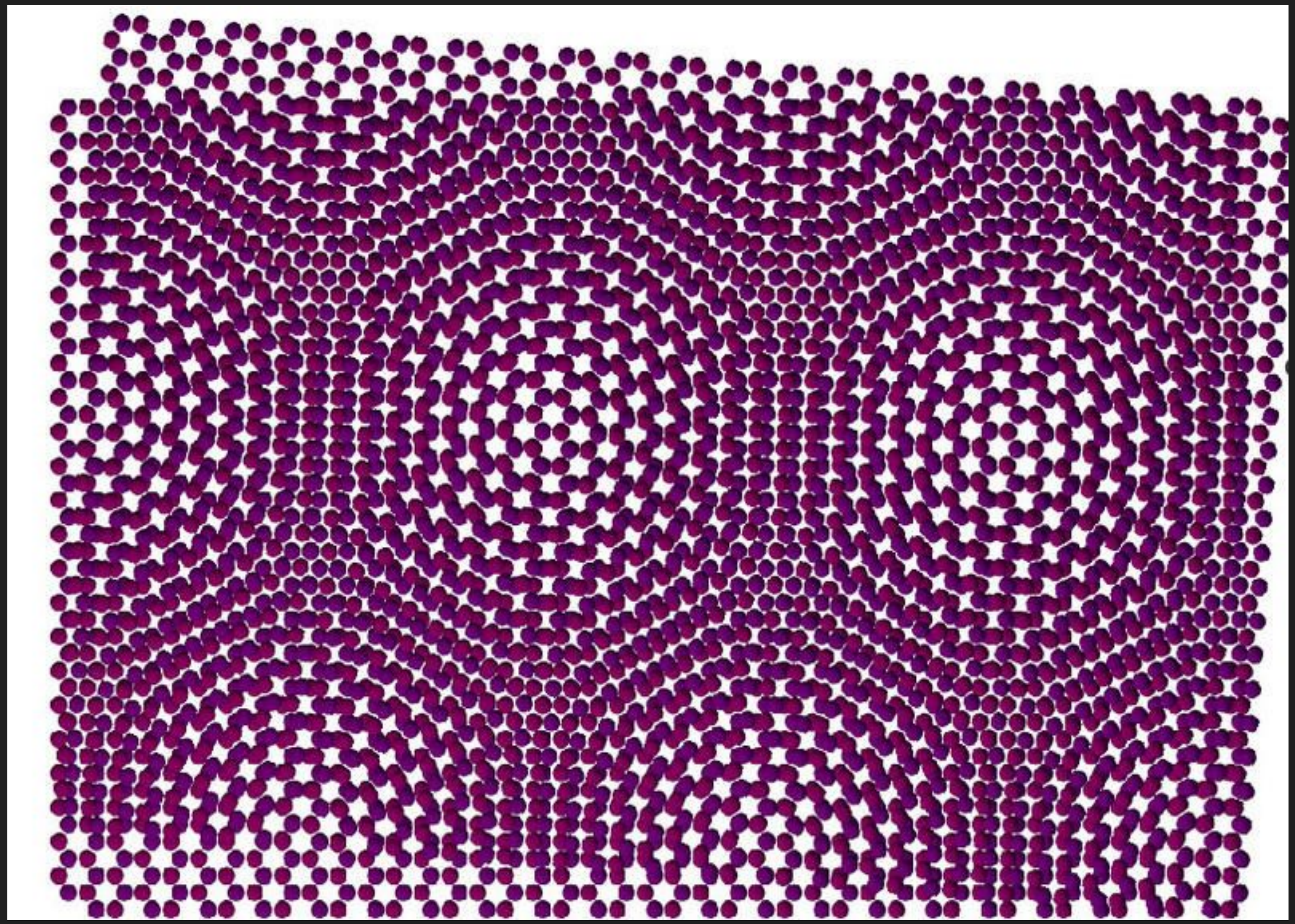

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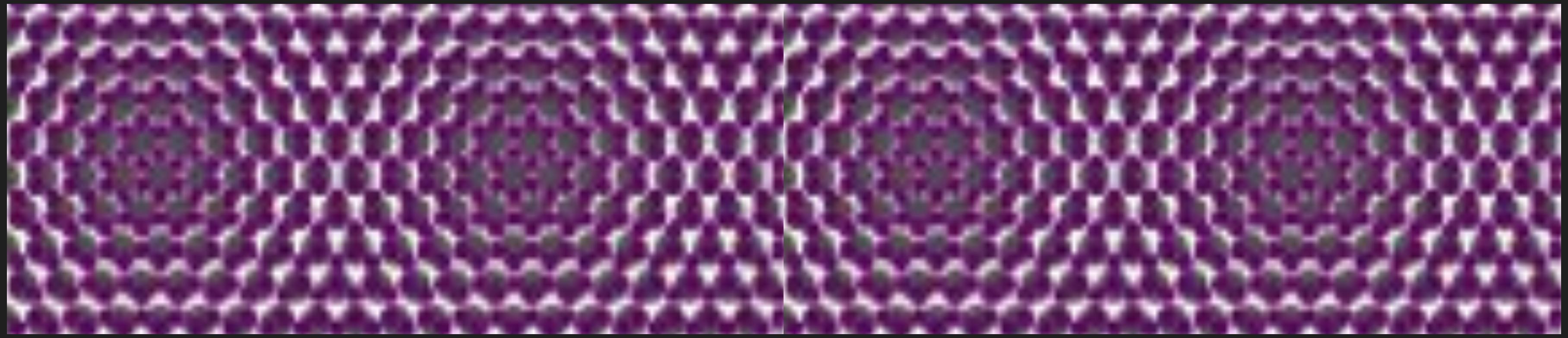
Collapsed SWCNT.



Wikipedia contributors. (n.d.). Moiré pattern. In Wikipedia, The Free Encyclopedia. Retrieved [05/11/24].

- Very highly ordered; no hetero-strain permitted. ***Reductions in moiré structural disorder, distinct from typical TBG!***

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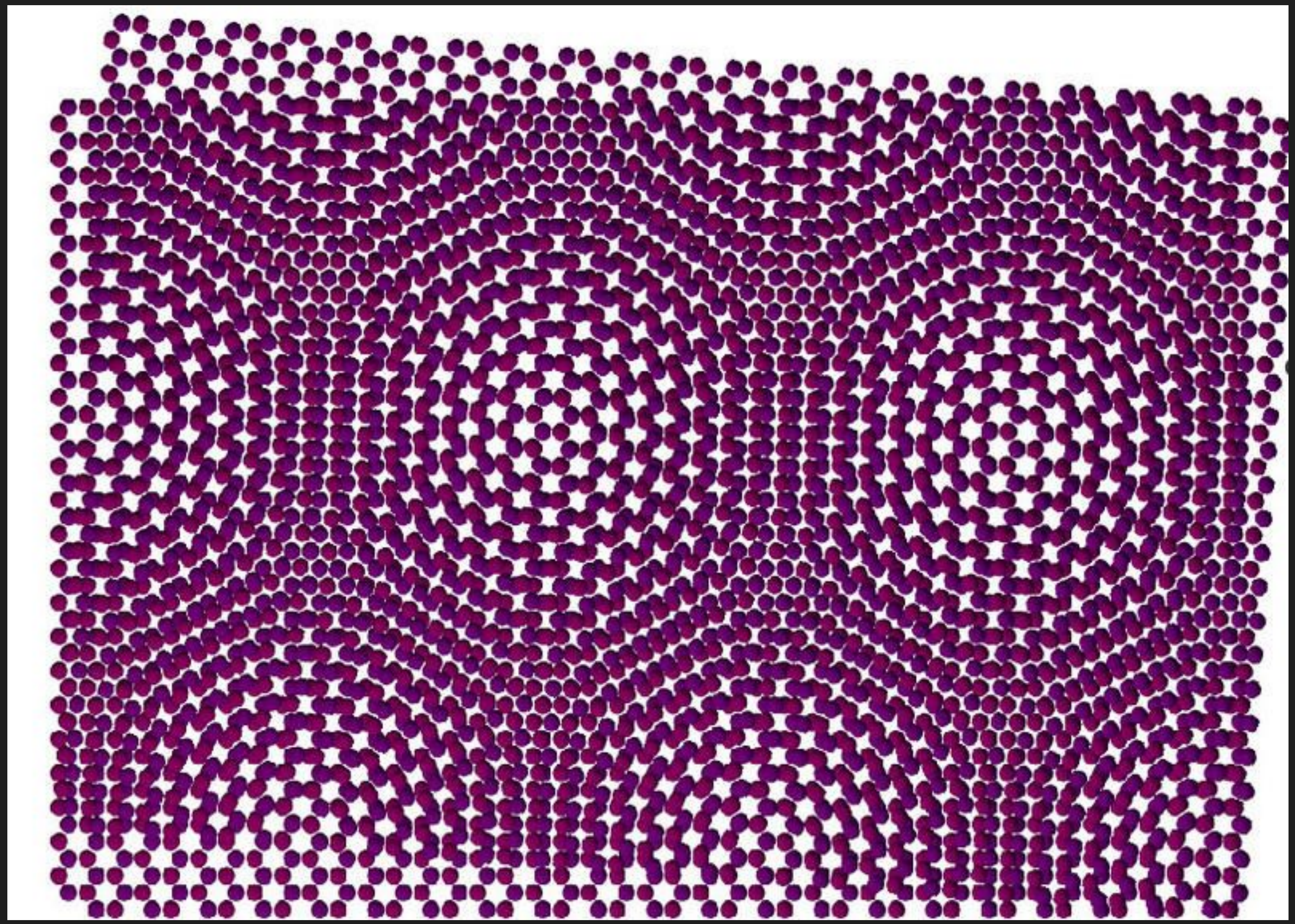

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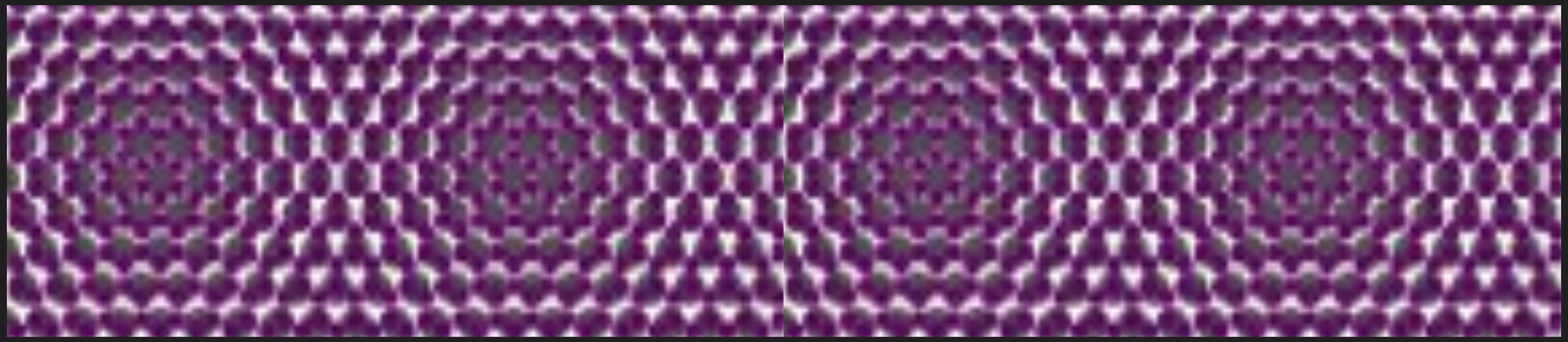

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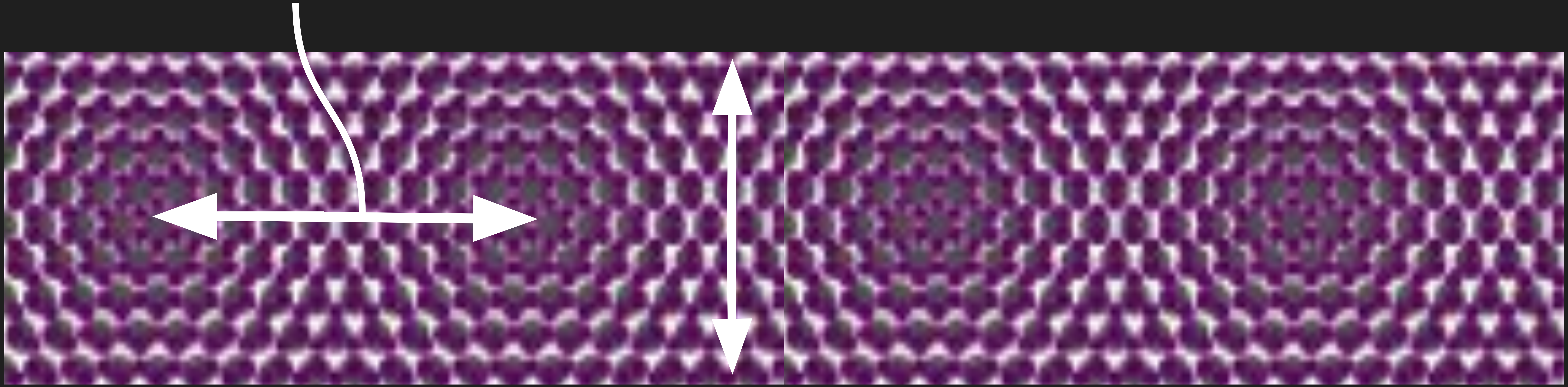
- Very highly ordered; no hetero-strain permitted. ***Reductions in moiré structural disorder, distinct from typical TBG!***

- Potential quasi-one-dimensionality tests the universality and dimensional dependence of moiré physics: do correlated phases persist, disappear, or transform in 1D?

Some necessary formalism

Moiré wavelength of **collapsed** SWCNT:

$$\lambda_{\mathcal{M}}(n,m) = \frac{a}{2\sin\left(\frac{2\arctan\left(\frac{m\sqrt{3}}{2n+m}\right)}{2}\right)}$$



- All relevant parameters are fundamentally dependent on the chiral indices: $n, m \in \mathbb{Z}^+$

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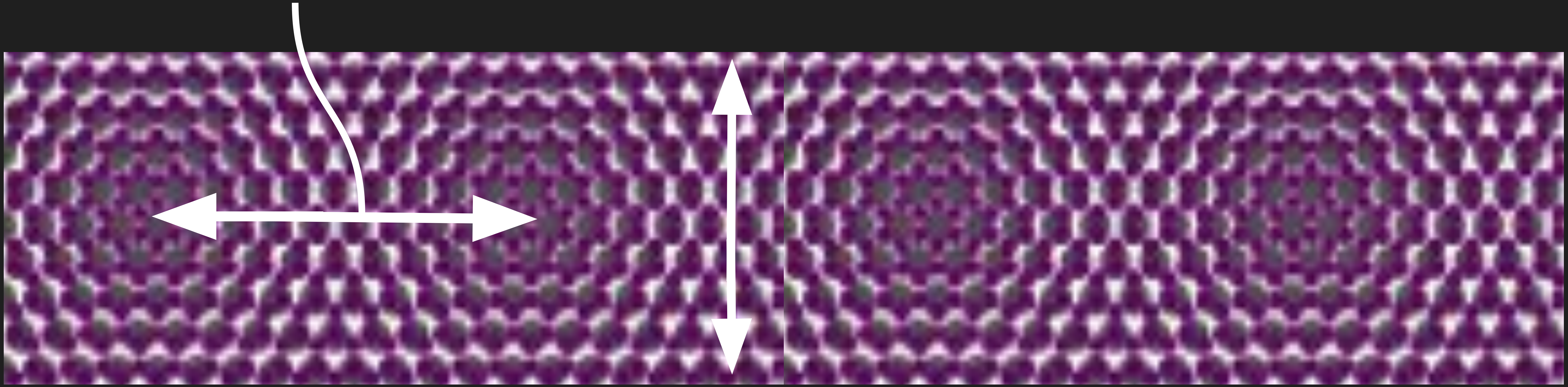
Diameter of **uncollapsed** SWCNT:

$$d(n,m) = \frac{a}{\pi}\sqrt{n^2 + nm + m^2}$$



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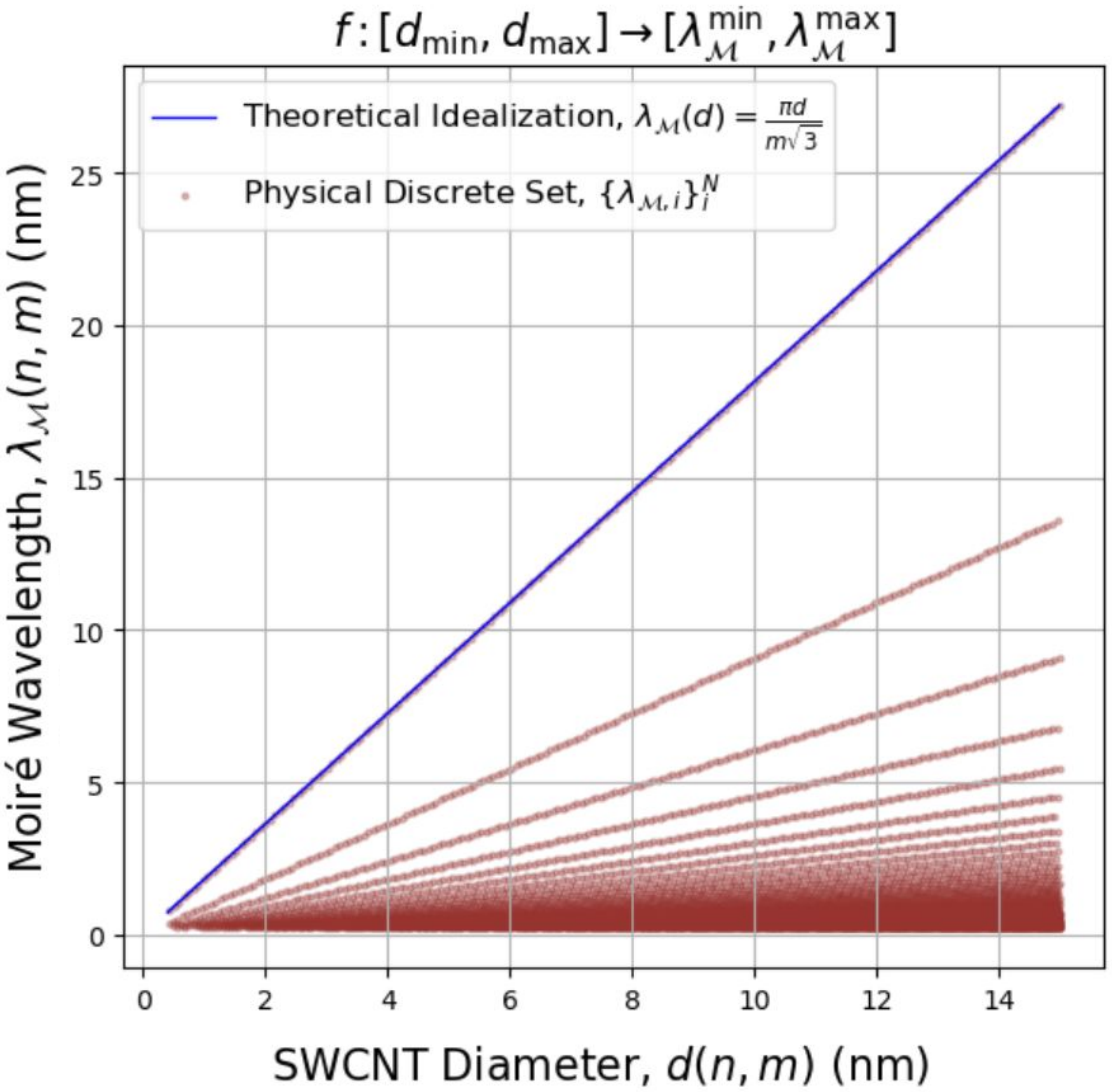
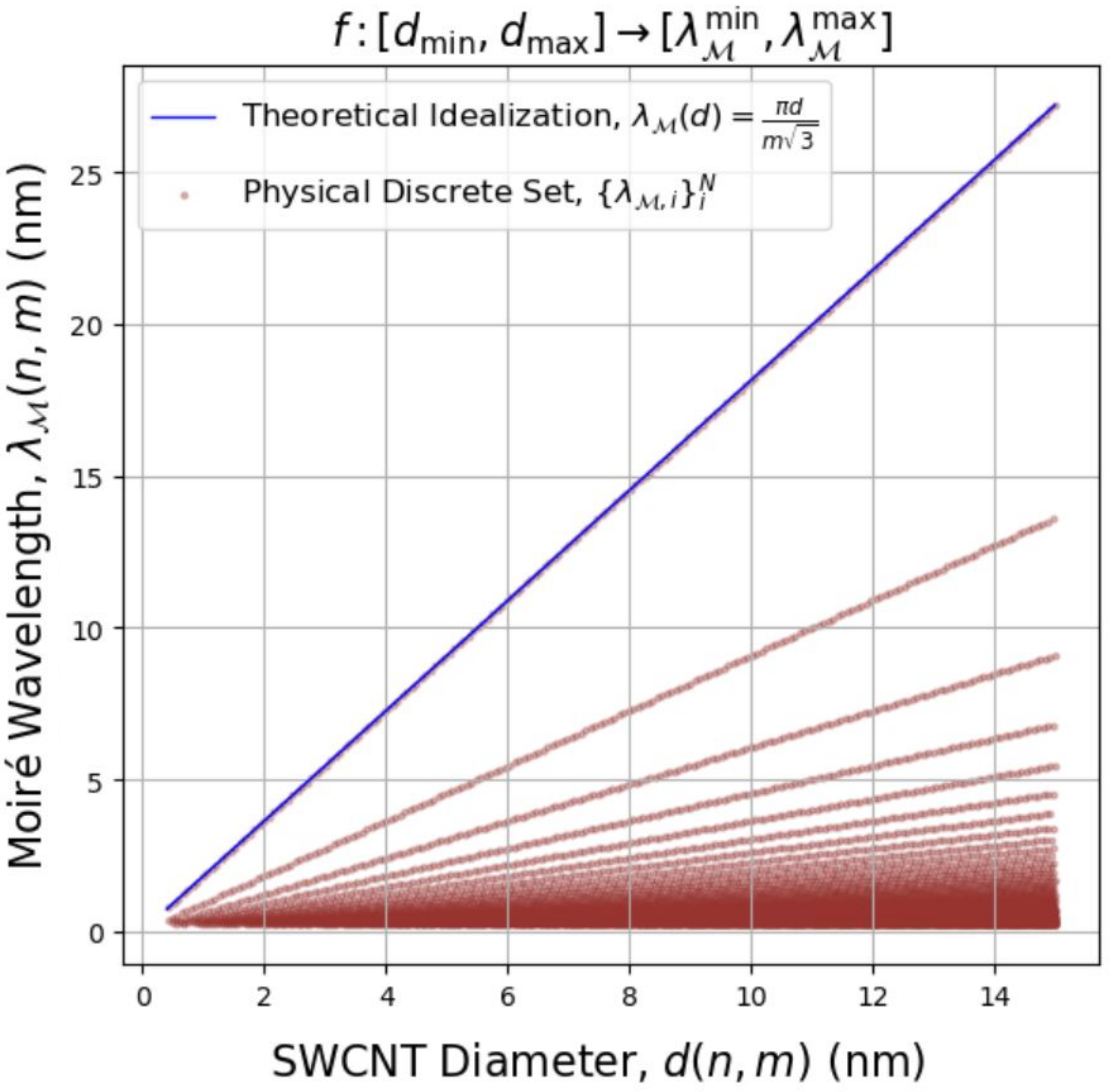
Needles of 1D in a wavelength Haystack



So many options… where does the quasi-one-dimensionality lie?



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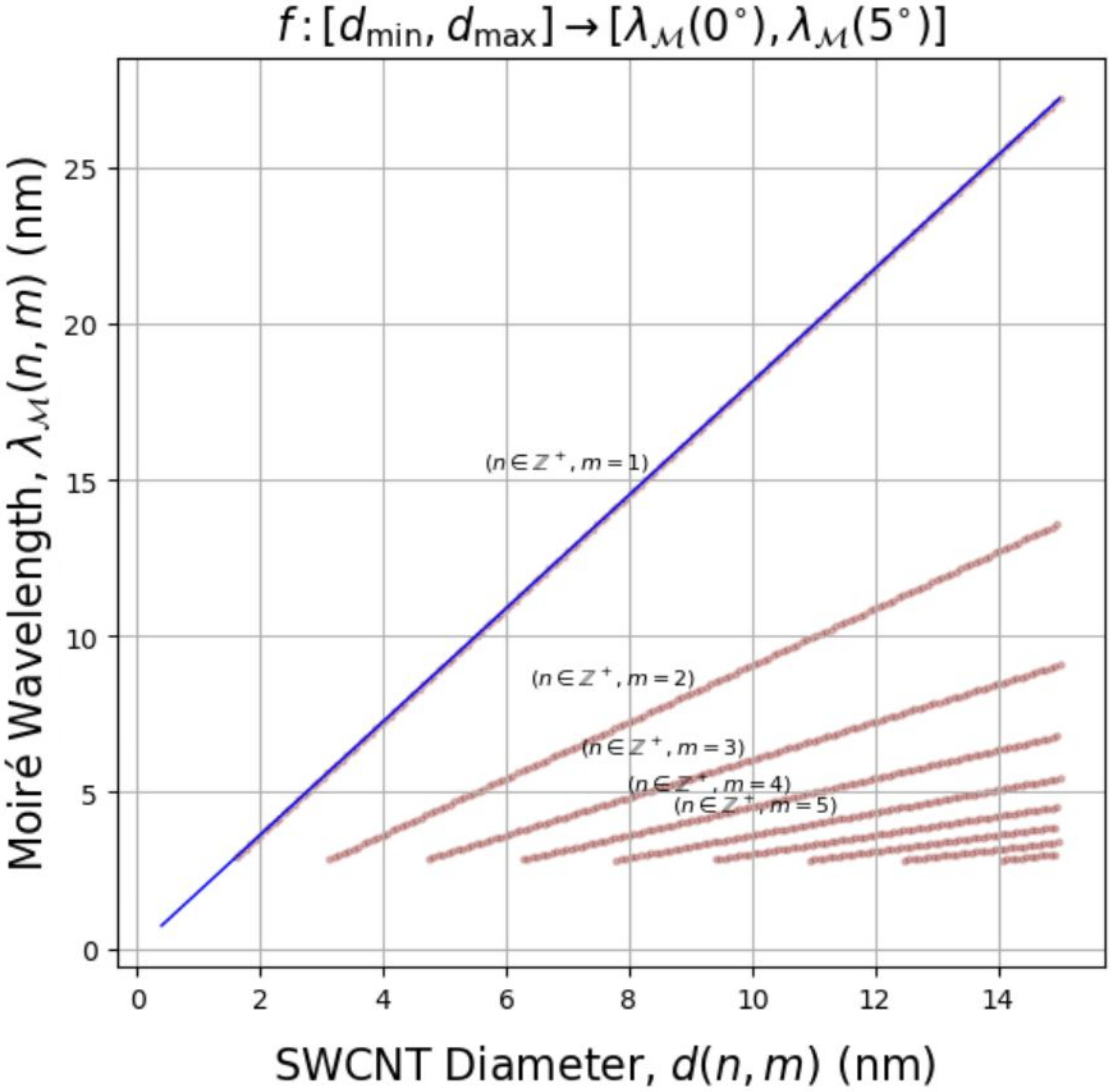
A given branch is defined by a family of chiral indices (n, m) where $\forall(\lambda_{\mathcal{M}}, d) \in \text{branch},\ m = c \in \mathbb{Z}^+$ and n runs over non-negative integer values, $n \in \mathbb{Z}^+$ (this maps out the curve).



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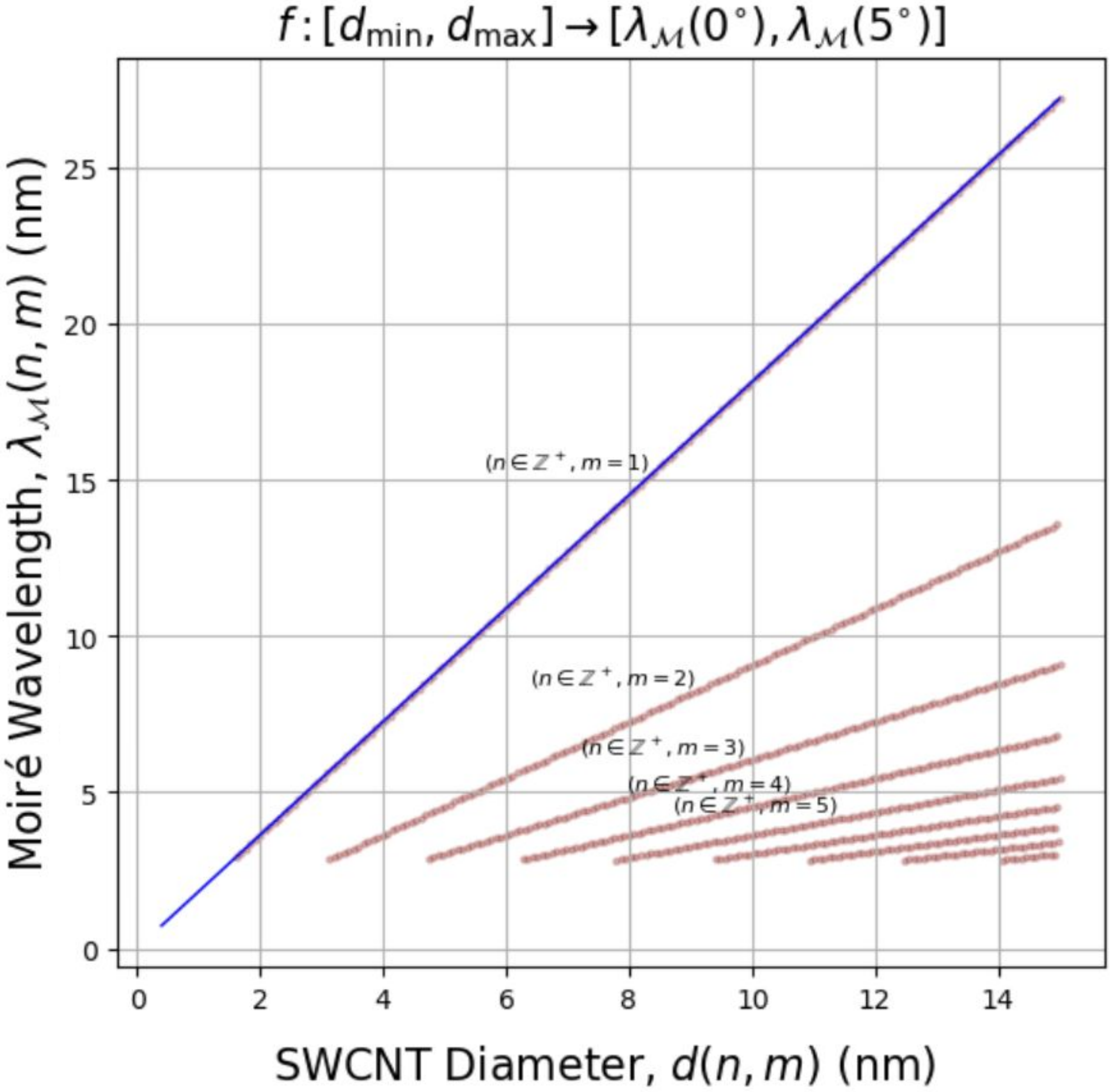

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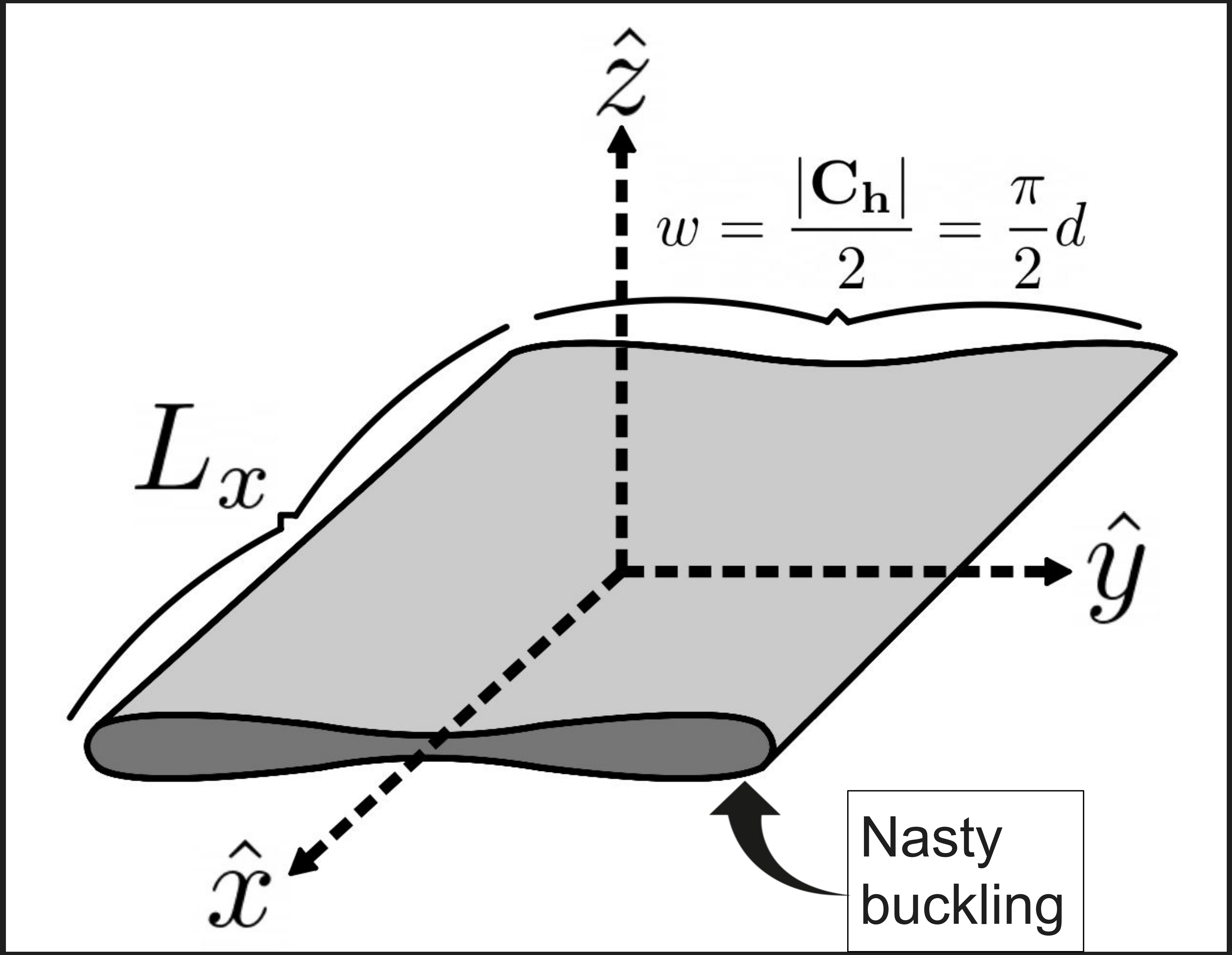
$$\implies \lambda_{\mathcal{M}}(d) = \frac{\pi d}{m\sqrt{3}}$$

Necessary condition for quasi-one-dimensionality

$$\underbrace{w = \frac{|\mathbf{C_h}|}{2} = \frac{\pi}{2} d}_{\text{Width of collapsed SWCNT (ignoring edge buckling)}}$$



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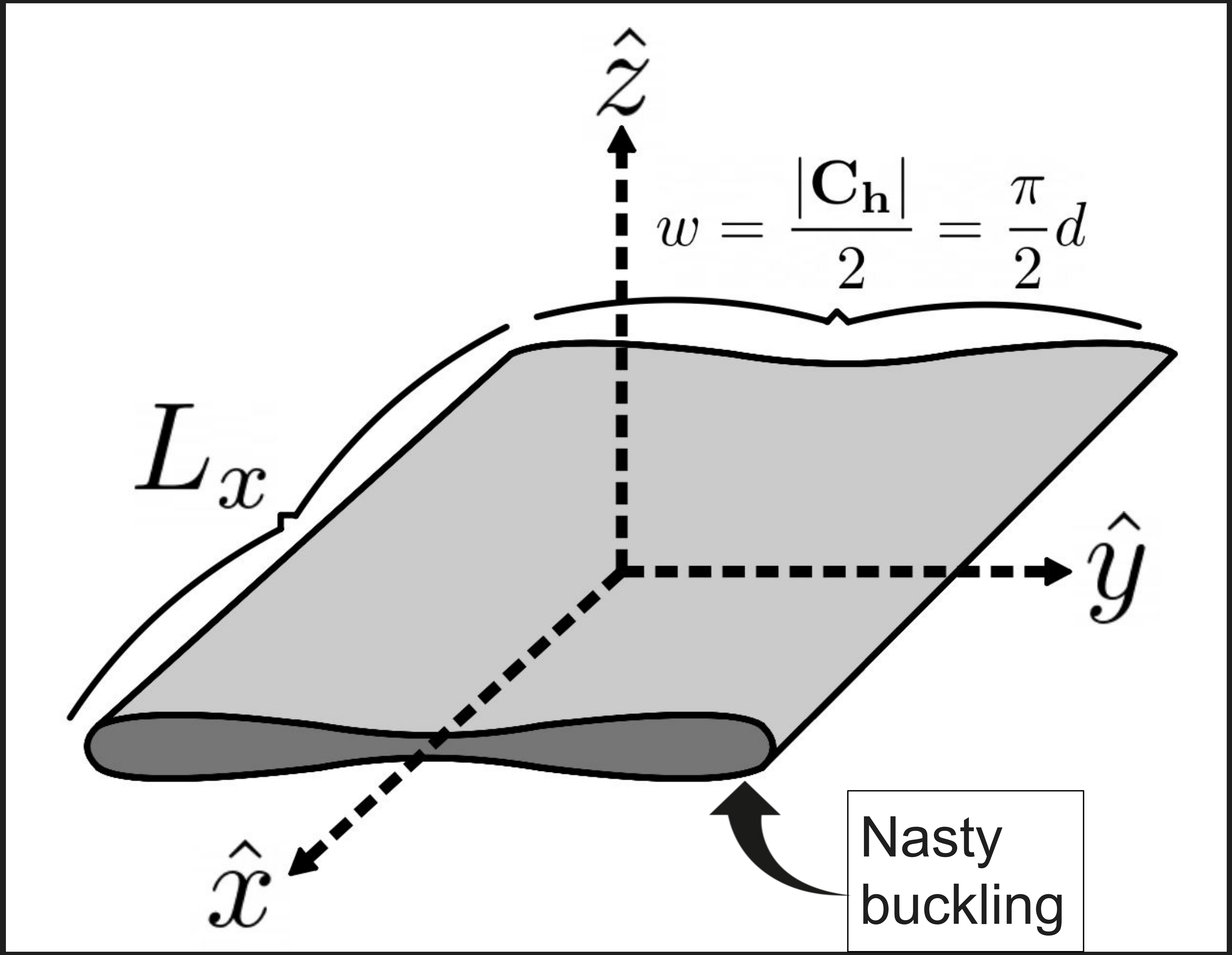

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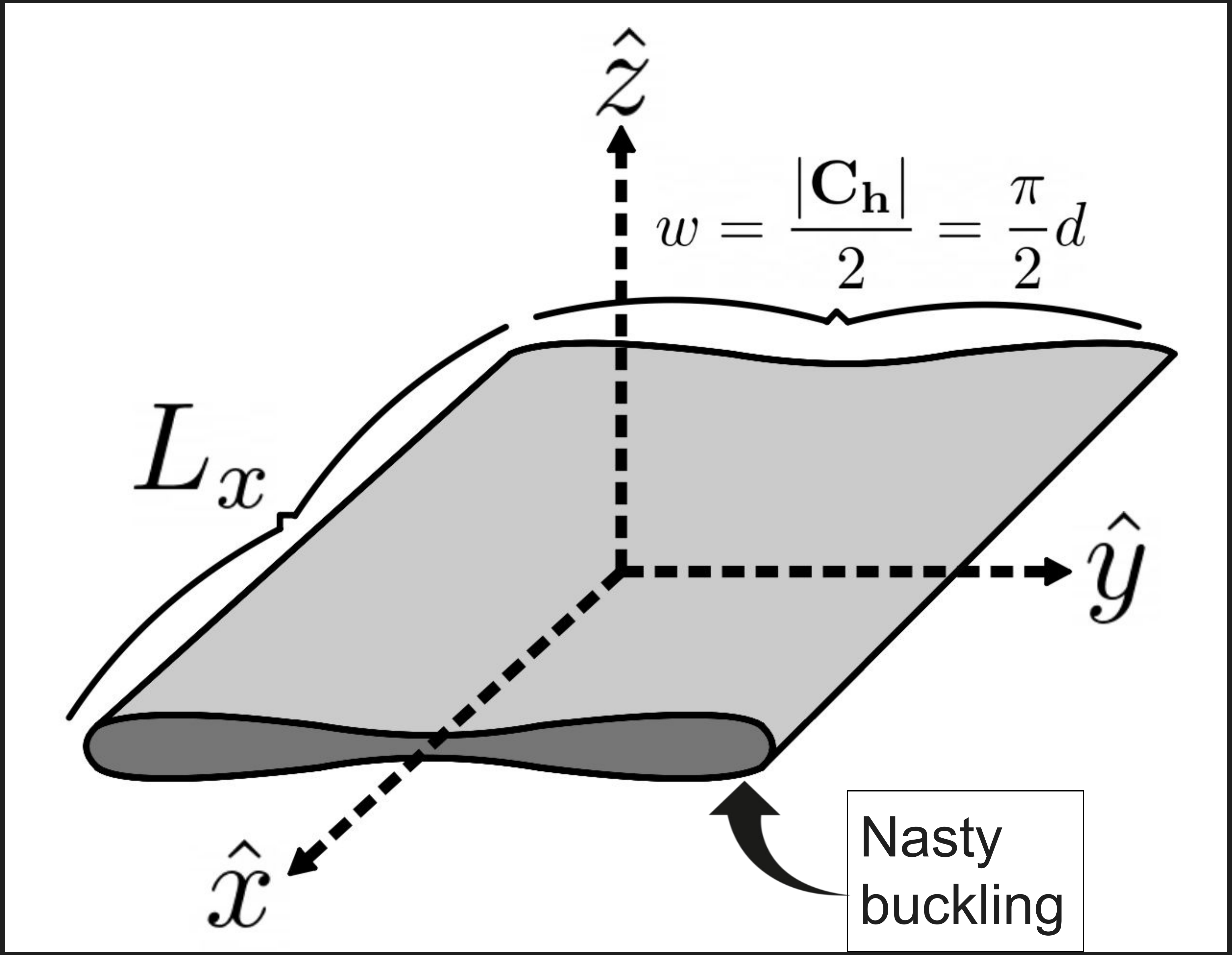

- When is this approximation true? When $m = 1$.

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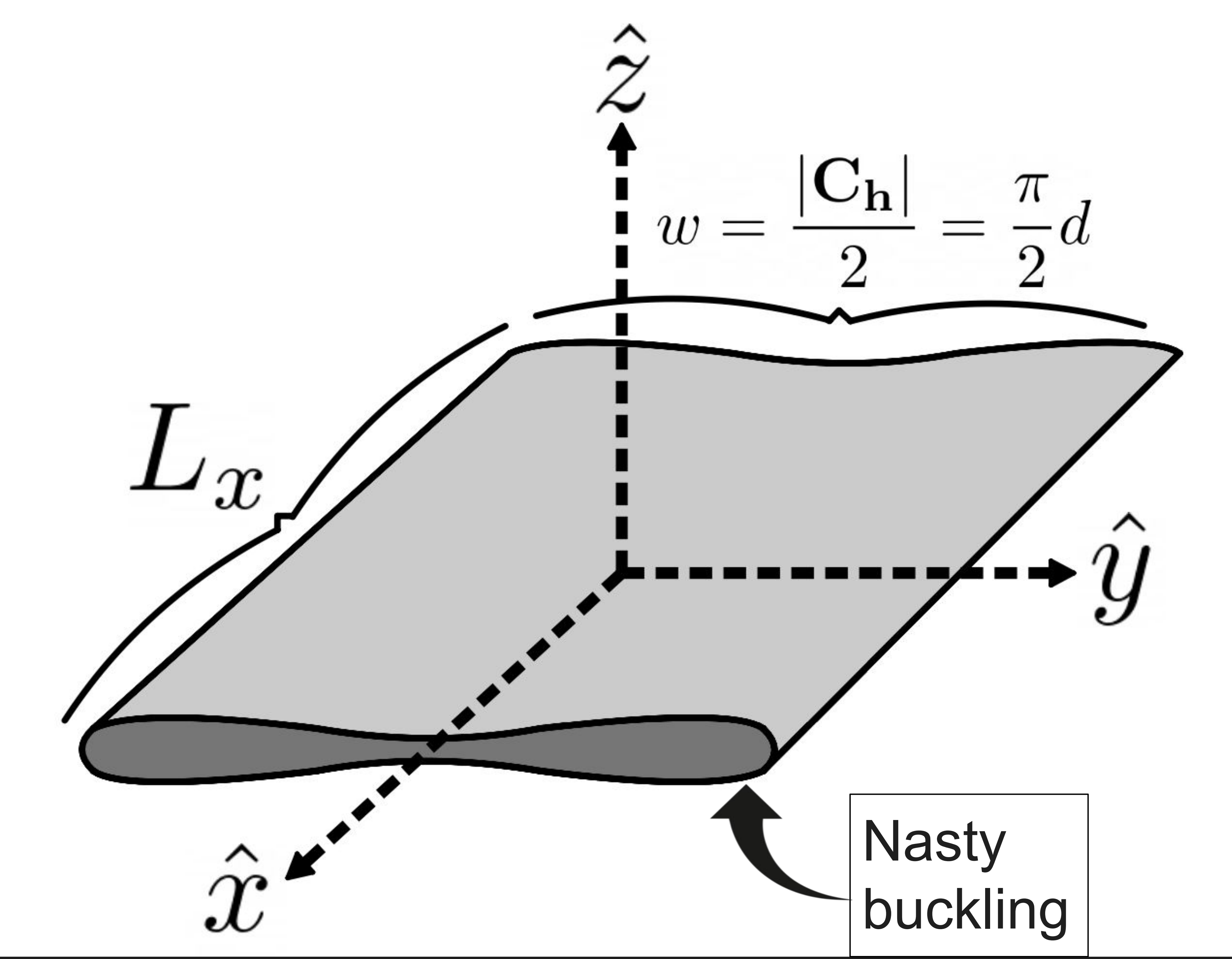
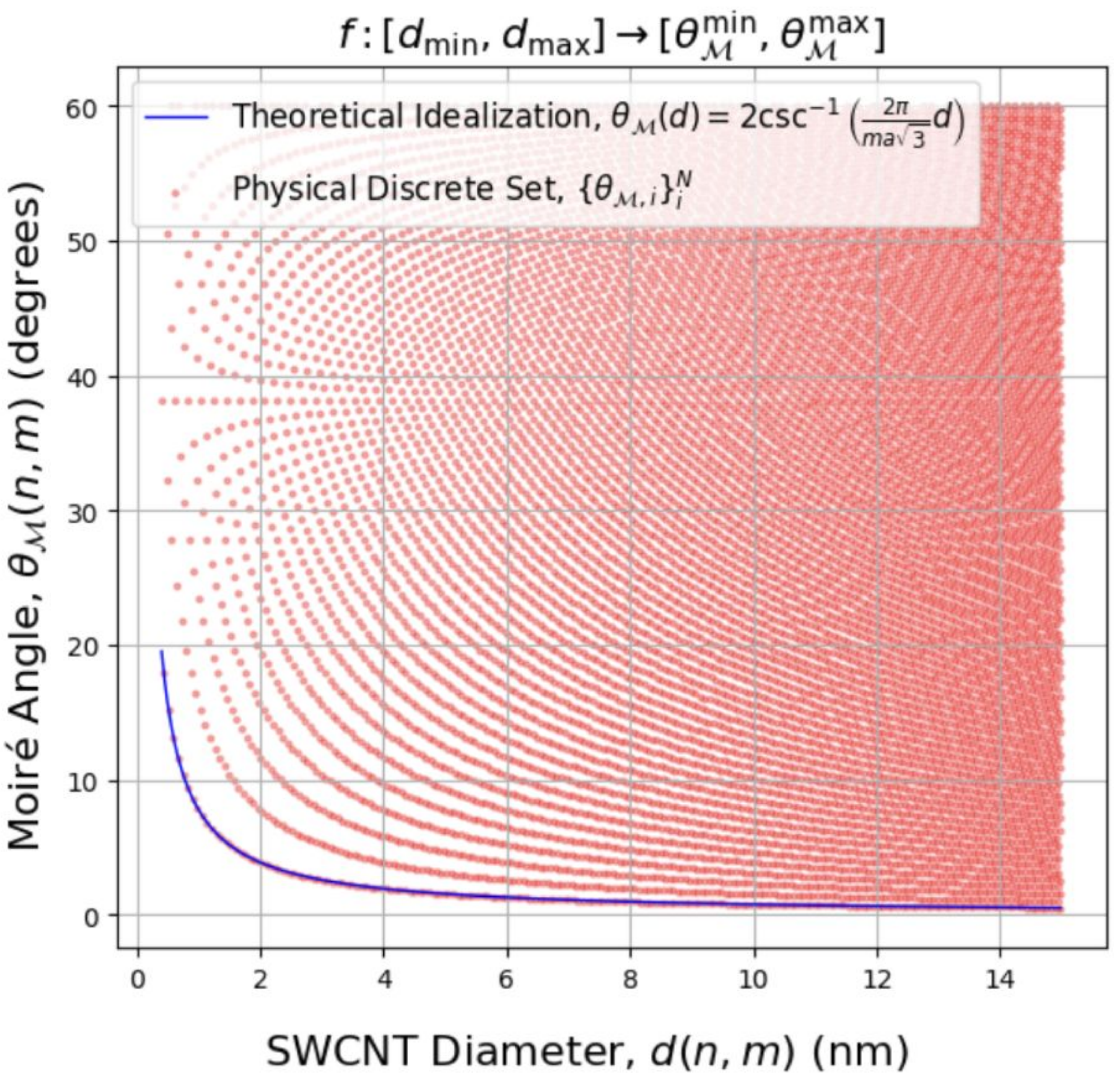
- When is this approximation true? When $m = 1$.

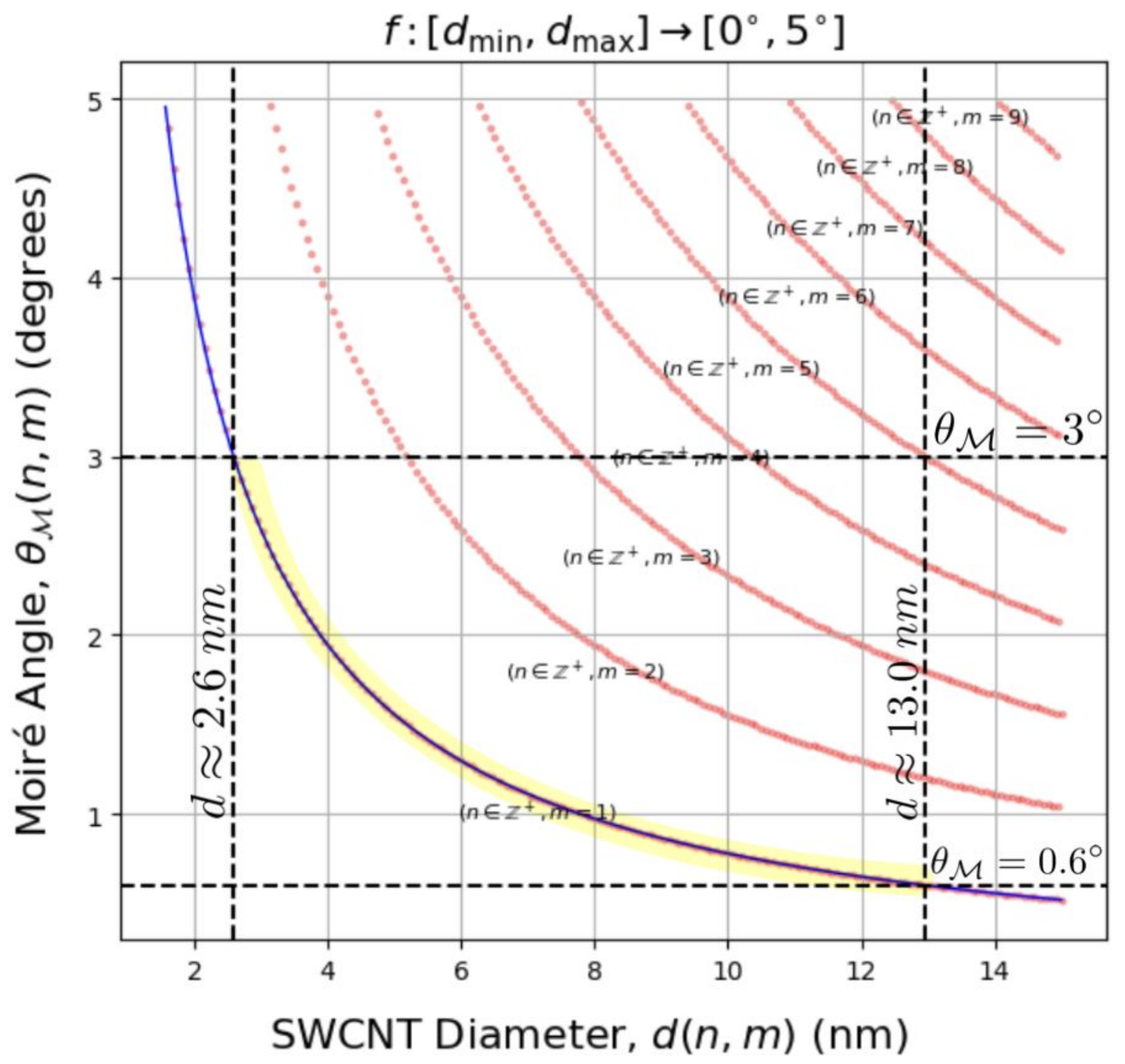
$$\lambda_{\mathcal{M}}\Big|_{m=1} \approx w$$

- We are interested in branches where $m = 1$.



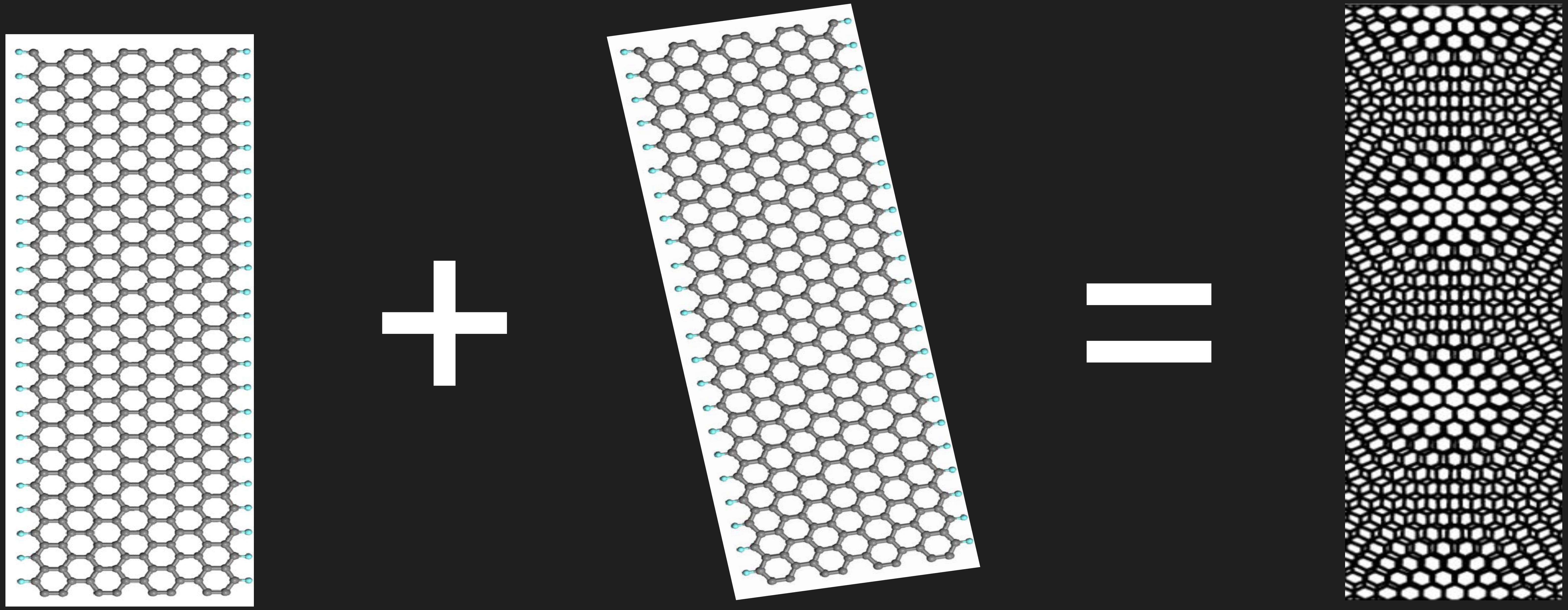


$f:[d_{\min}, d_{\max}] \to [\theta_{\mathcal{M}}^{\min}, \theta_{\mathcal{M}}^{\max}]$
Theoretical Idealization, $\theta_{\mathcal{M}}(d) = 2\csc^{-1}\left(\frac{2\pi}{ma\sqrt{3}}d\right)$
Physical Discrete Set, $\{\theta_{\mathcal{M},i}\}_i^N$
Moiré Angle, $\theta_{\mathcal{M}}(n,m)$ (degrees)
SWCNT Diameter, $d(n,m)$ (nm)
0
10
20
30
40
50
60
0
2
4
6
8
10
12
14



- We have isolated the region of potential quasi-1D geometry, greatly reducing our search area.
- Experimentalists, this is for you.

Band structure of collapsed SWCNT?
Let's start simple.



Quasi-1D Monolayer Graphene
(Nano-ribbon)

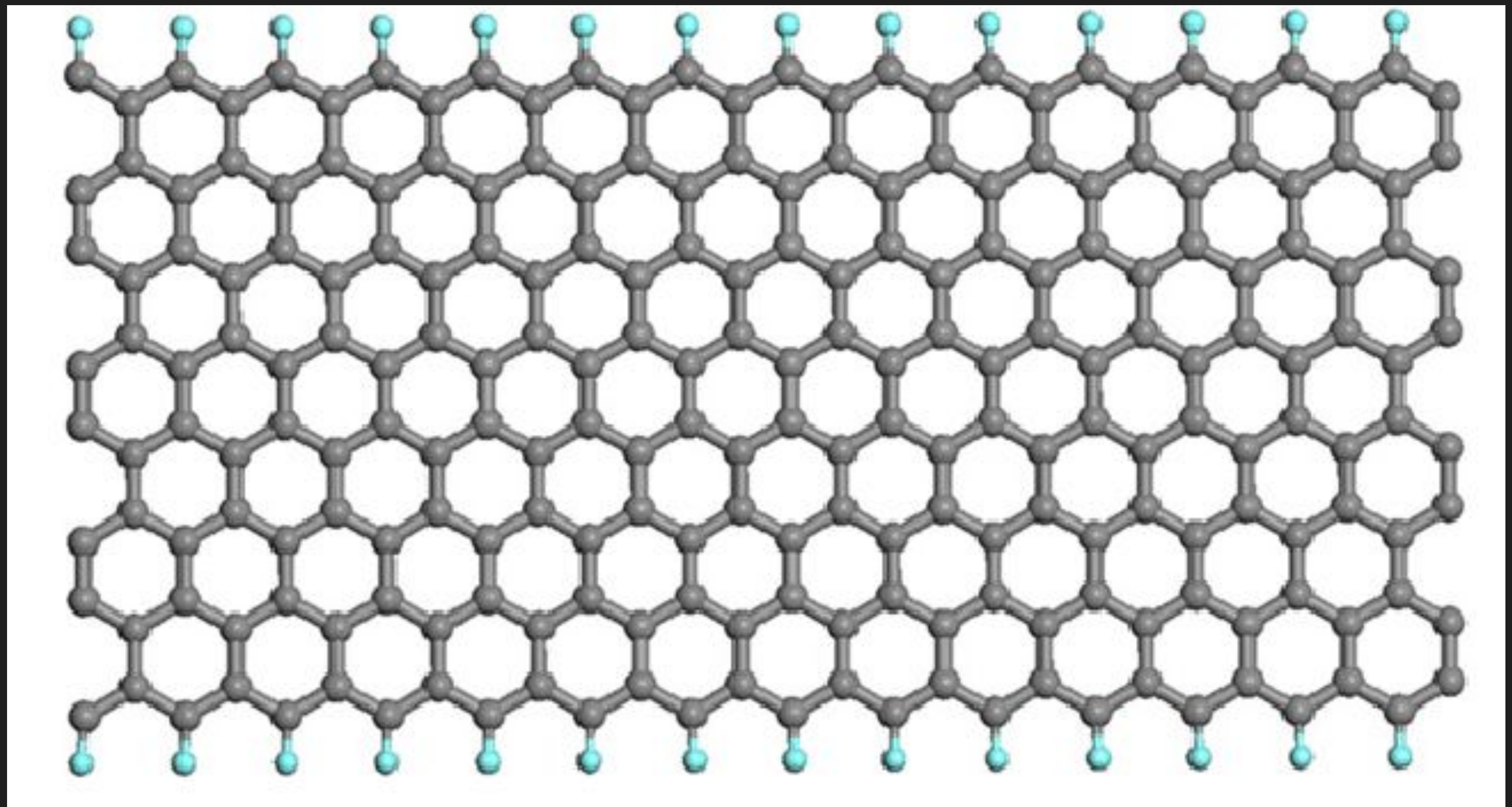
Quasi-1D
collapsed SWCNT

Start from established model of 2D monolayer graphene

- Low-energy Dirac Hamiltonian for electrons near the K-point in 2D monolayer graphene (real-space):

$$\hat{\mathcal{H}}_{2\mathrm{D}}(x, y) = -i\hbar v_F(\sigma_x \partial_x + \sigma_y \partial_y)$$

Terrones, H., Lv, R., Terrones, M., & Dresselhaus, M. S. (2012). The role of defects and doping in 2D graphene sheets and 1D nanoribbons. Reports on Progress in Physics, 75(6), 062501.



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- Assume real-space separable wavefunction of the form:

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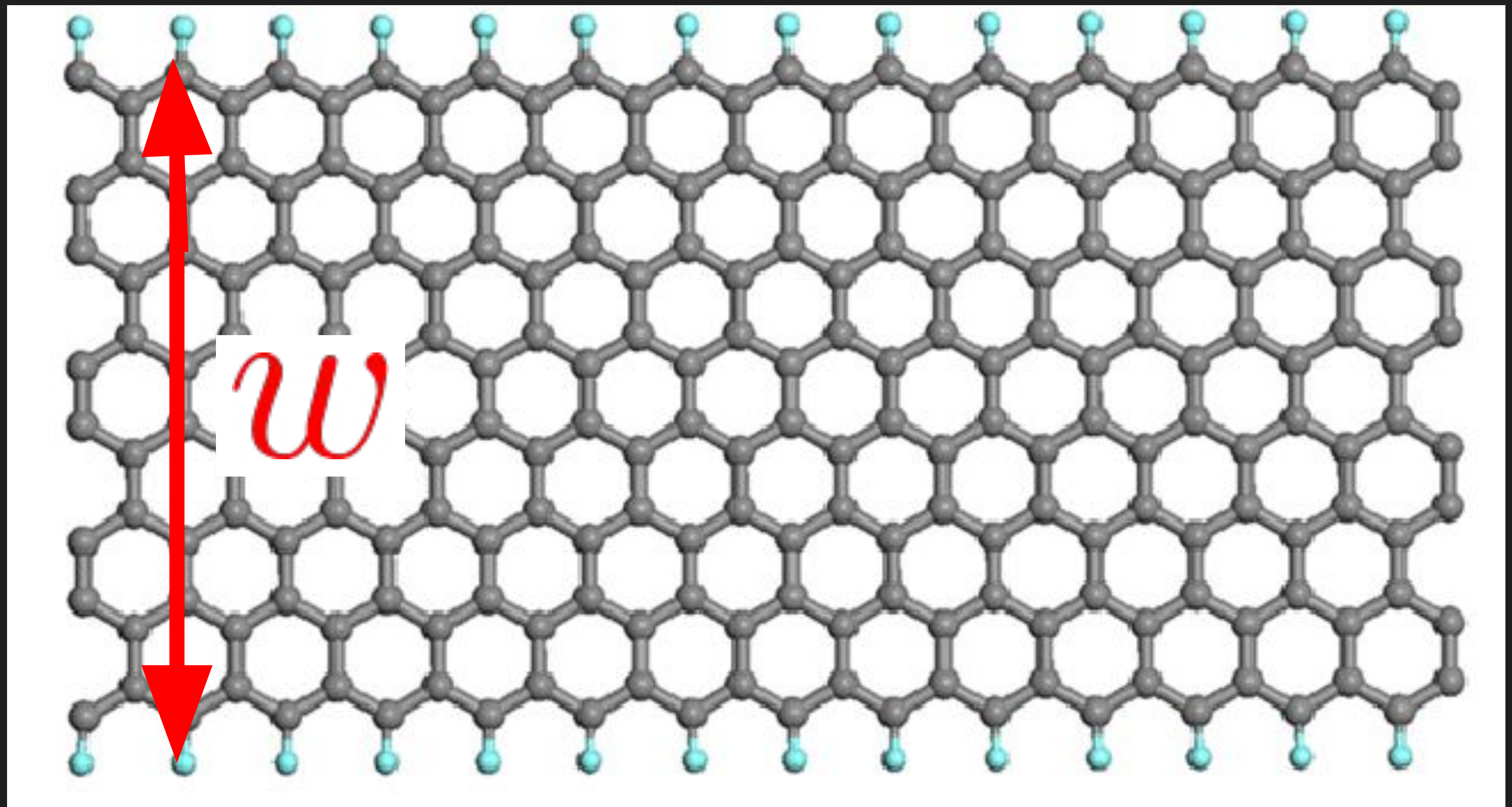

$$\Psi(x, y) = \sum_{n,k} c_{n,k} \chi_k(x) \phi_n(y)$$

- Hard-wall boundary conditions:

$$\Psi(x, 0) = \Psi(x, w) = 0$$

- Dirac equation reads: $\hat{\mathcal{H}}_{\mathrm{2D}}\Psi(x,y) = E\Psi(x,y)$

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- **Mode Projection:** Multiply by $\phi^\dagger_m(y)$ and integrate dy over $y \in [0,w]$, yielding a quasi-1D Dirac equation (projected along the length of the nanoribbon).

$$\sum_{n,k} c_{n,k}\chi_k(x) \underbrace{\int_0^w dy\, \phi^\dagger_m(y)\hat{\mathcal{H}}_{2D}\phi_n(y)}_{\langle\phi_m|\hat{\mathcal{H}}_{2D}|\phi_n\rangle = [\hat{\mathcal{H}}_{1D}]_{n,m}(x)} = E\sum_k c_{m,k}\chi_k(x)$$

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- Solving yields the spectrum of $[\hat{\mathcal{H}}_{1D}]_{n,m}(x)$, conveniently given by

$$\boxed{E_{k,n} = \pm\hbar v_F\sqrt{k^2 + \frac{n^2\pi^2}{w^2}}, \quad n \in \mathbb{Z}^+}$$

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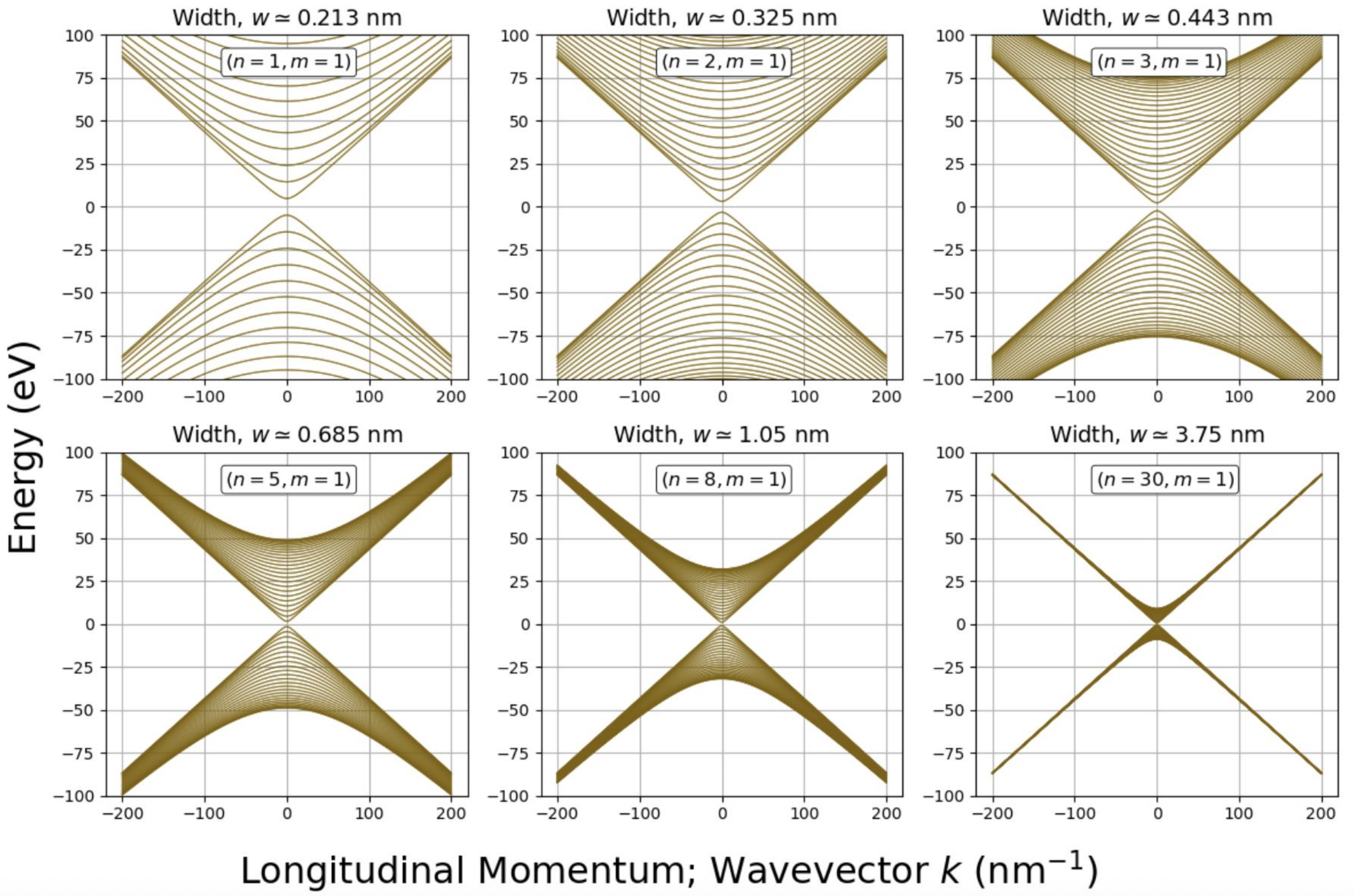
As $w \to \infty$, spectrum for monolayer graphene is recovered, $E(k) = \pm\hbar v_F|k|$.



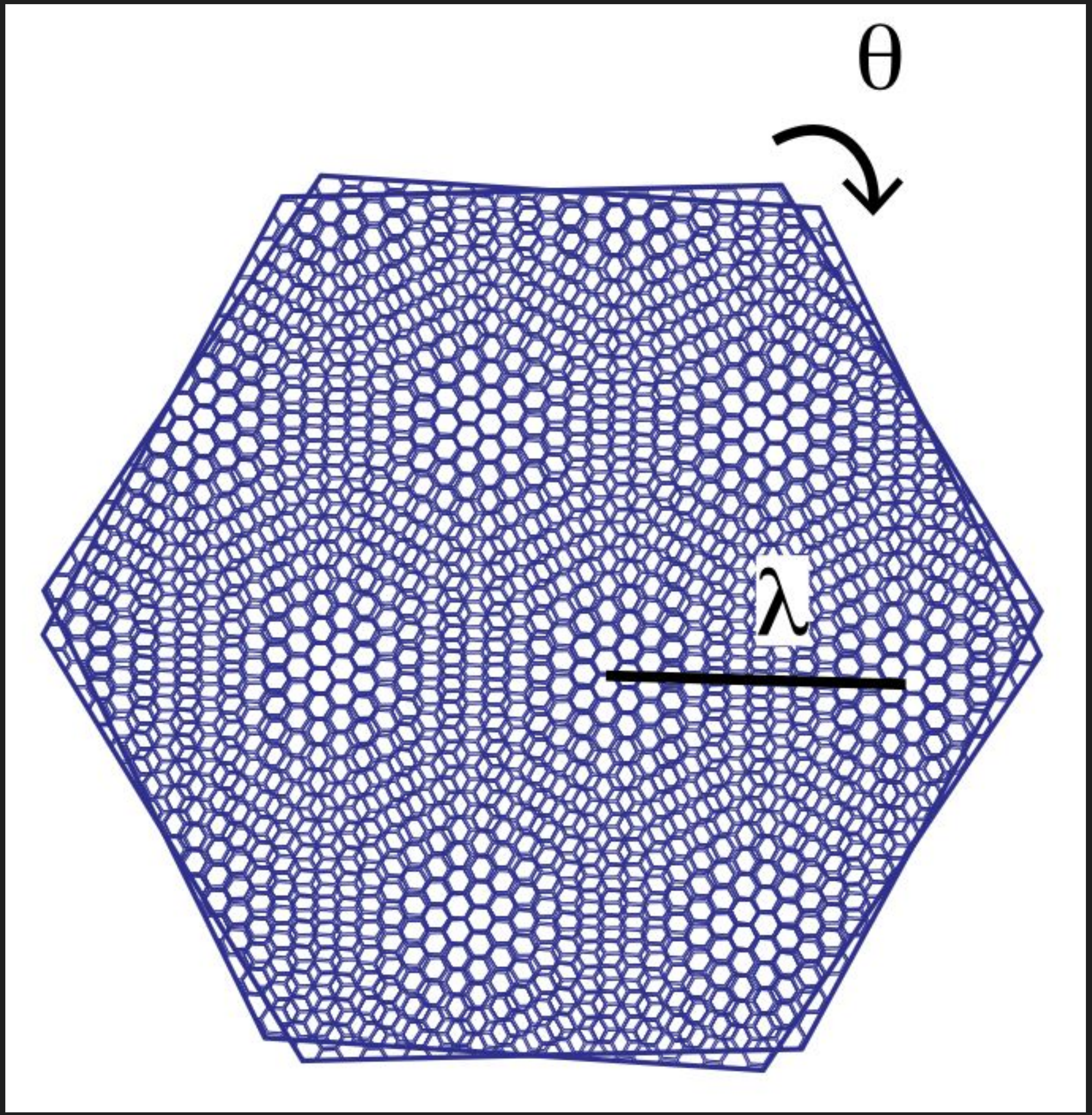
Width, $w \simeq 0.213$ nm
$(n=1, m=1)$
Width, $w \simeq 0.325$ nm
$(n=2, m=1)$
Width, $w \simeq 0.443$ nm
$(n=3, m=1)$
Width, $w \simeq 0.685$ nm
$(n=5, m=1)$
Width, $w \simeq 1.05$ nm
$(n=8, m=1)$
Width, $w \simeq 3.75$ nm
$(n=30, m=1)$
Energy (eV)
Longitudinal Momentum; Wavevector k (nm^{-1})

Applying slicing (projection) to quasi-1D SWCNT

- Use **Bistritzer–MacDonald (BM) model** where $-i\hbar v_F(\boldsymbol{\sigma}_{\theta_\mathcal{M}} \cdot \nabla)$ and $-i\hbar v_F(\boldsymbol{\sigma} \cdot \nabla)$ are the distinct Dirac Hamiltonians for two nanoribbons, stacked atop one another to form the collapsed SWCNT,

$$\hat{\mathcal{H}}_{\text{bilayer}}(\mathbf{r}) = \begin{pmatrix} -i\hbar v_F(\boldsymbol{\sigma} \cdot \nabla) & \hat{\mathcal{T}}(\mathbf{r}) \\ \hat{\mathcal{T}}^\dagger(\mathbf{r}) & -i\hbar v_F(\boldsymbol{\sigma}_{\theta_\mathcal{M}} \cdot \nabla) \end{pmatrix}$$

- Here's the novel part: project $\hat{\mathcal{H}}_{\text{bilayer}}(\mathbf{r})$ along the longitudinal axis of the collapsed SWCNT (akin to previous projection).



Thank you

Summary of findings & Continued interest

- **Finding #1:** Discovered necessary constraint on chiral indices (n, m) to yield quasi-1D behaviour in the collapsed SWCNT.

- **Finding #2:** Successfully utilized slicing (projection) to yield quasi-1D Hamiltonian, reproducing known band structures and dispersion relations for quasi-1D nano-ribbon.

- **Hope:** Having validated slicing, utilize it to produce bands and dispersion relations for quasi-1D collapsed SWCNT (essentially two quasi-1D nano-ribbons stacked together).