Electronic properties of graphene antidot lattices

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- Introduction to graphene antidot lattices

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- Tight-binding band structure, appearance of midgap states, comparison with other approaches

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- Electron-phonon coupling, mass renormalization as a signature of polaronic behavior

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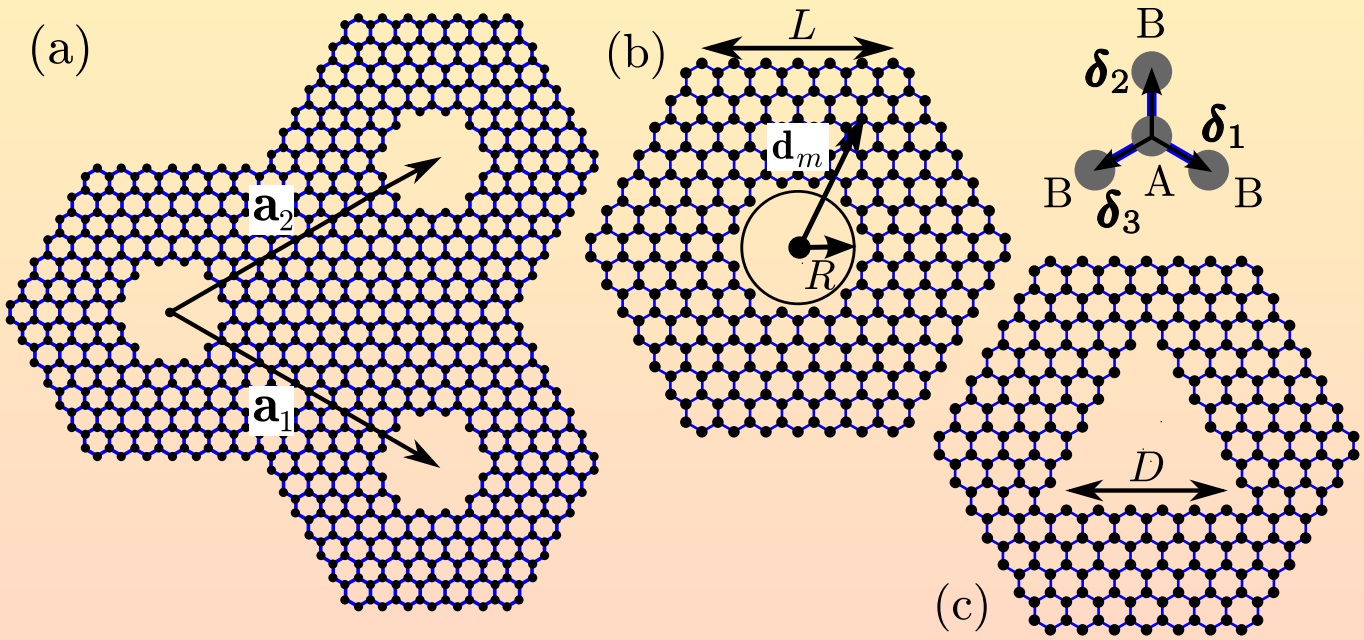
- Conclusions and Outlook

Graphene antidot lattices

T. G. Pedersen *et al.*, PRL **100**, 136804 (2008)



triangular superlattices with circular antidots $\{L, R\}$,
or triangular antidots $\{L, D\}$

Research on graphene antidot lattices: an overview

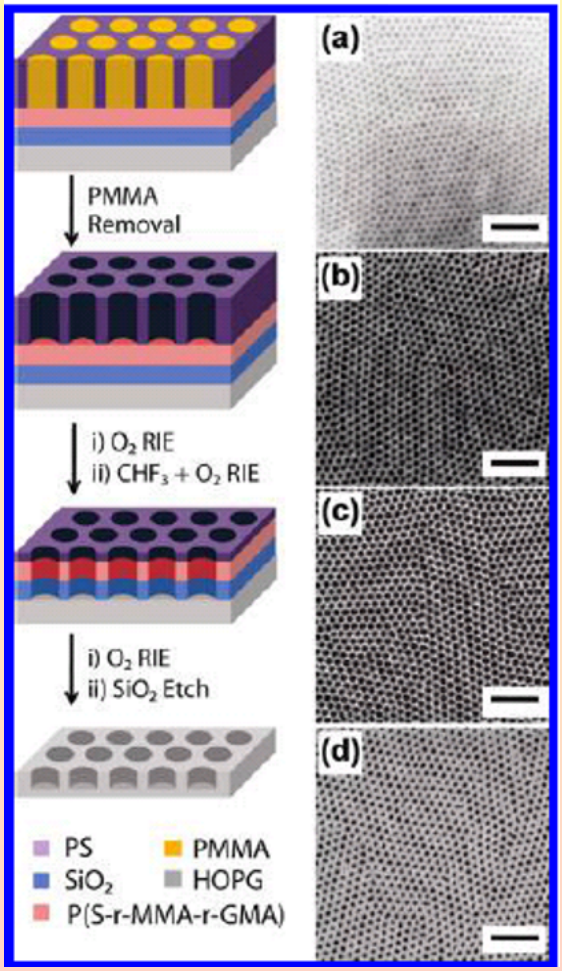
Theory:

- T. G. Pedersen *et al.*, PRL **100**, 136804 (2008);
- T. G. Pedersen *et al.*, PRB **77**, 245431 (2008);
- M. Vanević, VMS, and M. Kindermann, PRB **80**, 045410 (2009);
- J. A. Fürst *et al.*, PRB **80**, 115117 (2009);
- J. A. Fürst *et al.*, New J. Phys. **11**, 095020 (2009);
- L. Rosales *et al.*, PRB **80**, 073402 (2009);
- X. H. Zheng *et al.*, PRB **80**, 075413 (2009);
- W. Liu *et al.*, PRB **80**, 233405 (2009);
- R. Petersen and T. G. Pedersen, PRB **80**, 113404 (2009);
- N. Vukmirović, VMS, and M. Vanević, PRB **81**, 041408 (R) (2010);
- VMS, N. Vukmirović, and C. Bruder, PRB **82**, 165410 (2010).

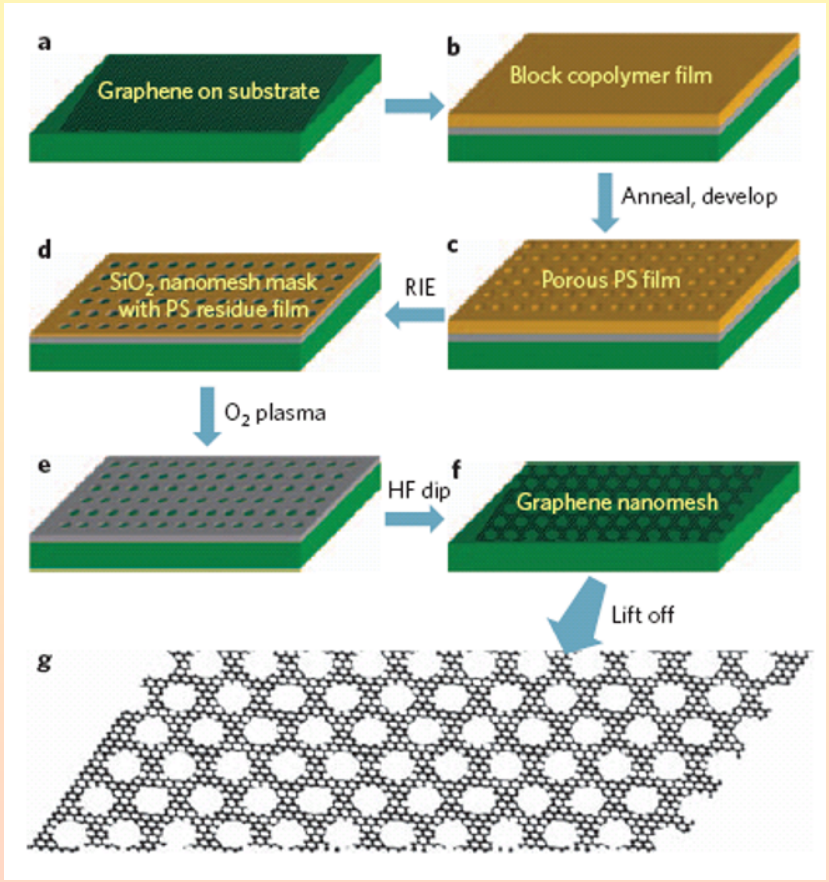
Experiment:

- T. Shen *et al.*, APL **93**, 122102 (2008);
- J. Eroms and D. Weiss, New J. Phys. **11**, 095021 (2009);
- J. Bai *et al.*, Nature Nanotech. **5**, 190 (2010);
- M. Kim *et al.*, Nano Lett. **10**, 1125 (2010).

Realization of graphene antidot lattice (March 2010)



M. Kim *et al.*,
Nano Lett. **10**, 1125 (2010)



J. Bai *et al.*,
Nature Nanotech. **5**, 190 (2010)

Relevant numbers

triangular antidot lattice period: $La\sqrt{3}$ $\quad (\, a \approx 2.46$ Å $)$

UCLA lattices: period $27 - 39$ nm

Wisconsin lattices: period 36.4 nm

our lattices: $\{L, R = 5\}$ family $(\, 9 \leq L \leq 19\,)$

$\{L, R = 7\}$ family $(\, 12 \leq L \leq 20\,)$

$\Rightarrow$ period ≤ 8.2 nm

$N_{\mathrm{at}} \sim 300 - 1600$ C atoms per unit cell

Electronic structure of antidot lattices (I)

DFT band-structure calculation not conceivable!

Tight-binding model (n.n. hopping integral $t \approx 2.8$ eV):

$$\hat{H}_{\mathrm{e}} = -\frac{t}{2} \sum_{\mathbf{R},m,\delta} \left(\hat{a}^{\dagger}_{\mathbf{R}+\mathbf{d}_m+\delta} \hat{a}_{\mathbf{R}+\mathbf{d}_m} + \mathbf{h.c.} \right)$$

Good comparison with DFT results for lattices with small unit cells!
[J. A. Fürst *et al.*, NJP **11**, 095020 (2009)]

Artifact of the n.n. tight-binding model on a **bipartite lattice**:
exact **particle-hole symmetry**

Electronic structure of antidot lattices (II)

$$\hat{H}_{\mathrm{e}}\psi_{n\mathbf{k}}(\mathbf{r}) = \varepsilon_{n\mathbf{k}}\psi_{n\mathbf{k}}(\mathbf{r}) \quad , \quad \psi_{n\mathbf{k}}(\mathbf{r}) = \sum_m C_m^{n,\mathbf{k}}\phi_{n\mathbf{k}}(\mathbf{r})$$

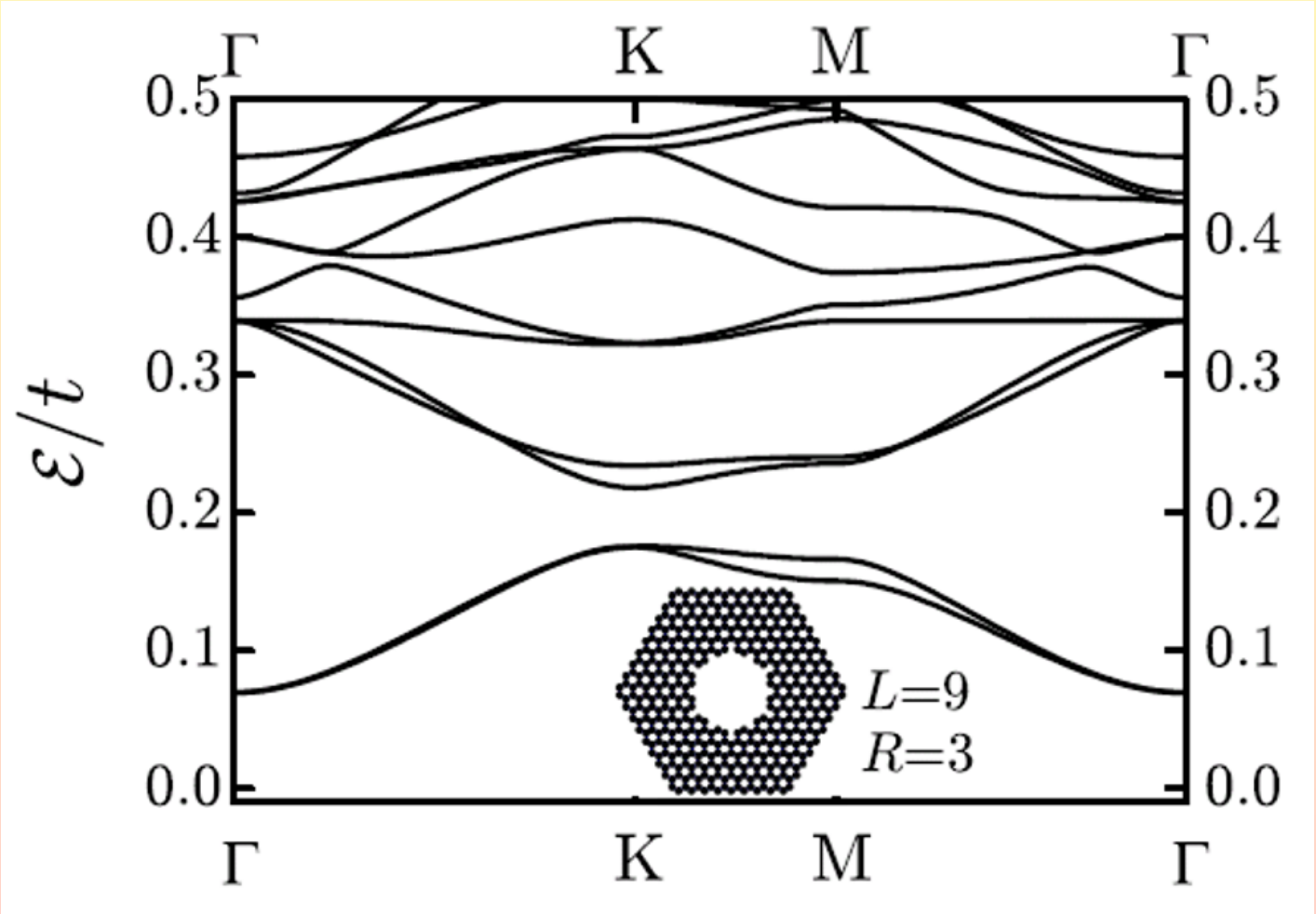
$$\phi_{n\mathbf{k}}(\mathbf{r}) \equiv N^{-1/2}\sum_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}}\varphi(\mathbf{r}-\mathbf{R}-\mathbf{d}_m)$$

$2p_z$ orbital of the C atom at $\mathbf{R}+\mathbf{d}_m$: $\varphi(\mathbf{r}-\mathbf{R}-\mathbf{d}_m)$

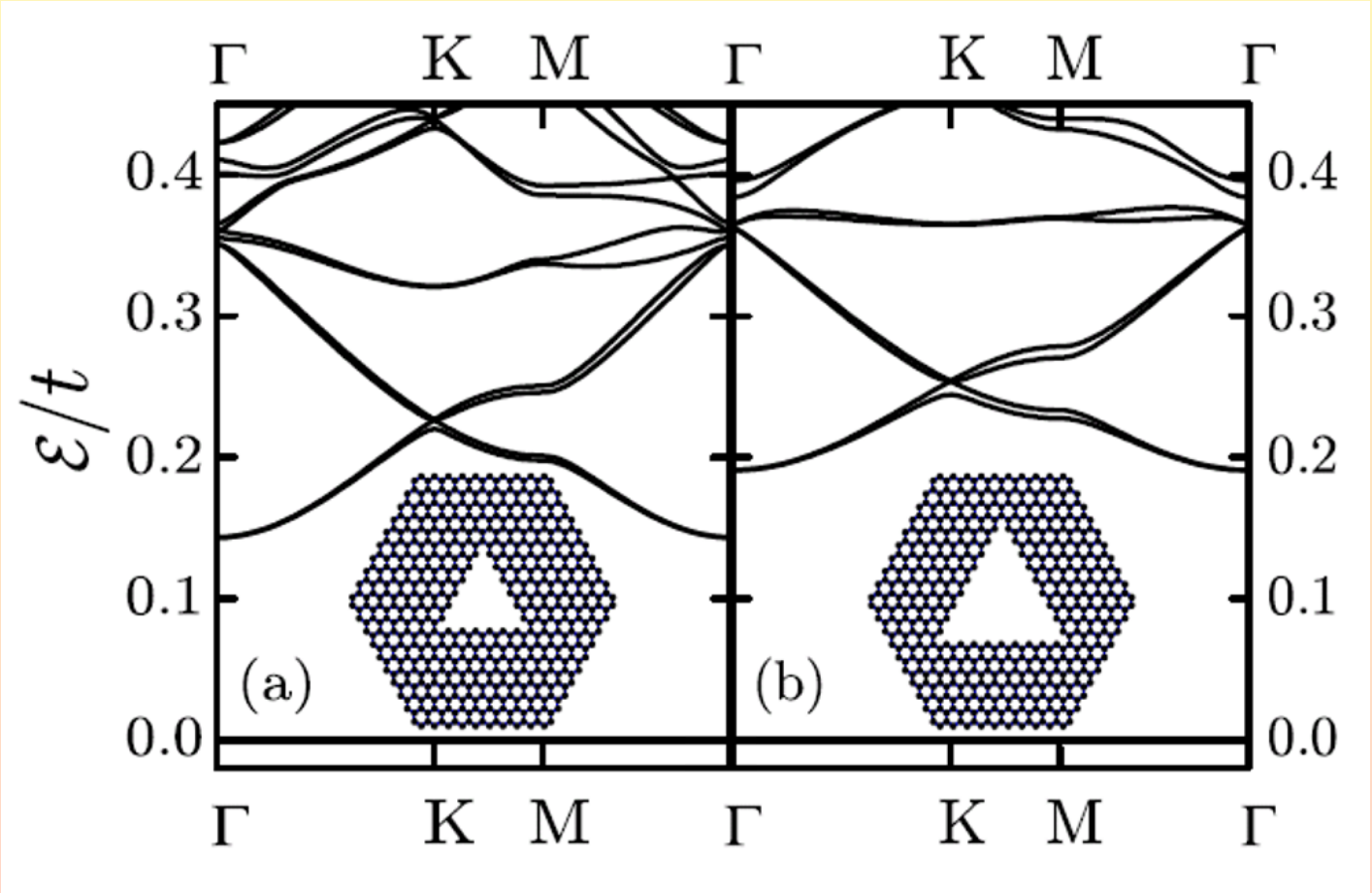
$$\left\langle \varphi(\mathbf{r}-\mathbf{R}'-\mathbf{d}_{m'}) \middle| \varphi(\mathbf{r}-\mathbf{R}-\mathbf{d}_m) \right\rangle = \delta_{\mathbf{R},\mathbf{R}'}\delta_{m,m'}$$

(Overlap of $2p_z$ orbitals on different atoms is negligible)

Band structure: circular-antidot case



Band structure: triangular-antidot case



$\{L = 9, D = 6\}$ $\{L = 9, D = 9\}$

Origin of completely flat bands at $\varepsilon = 0$

Bipartite topology $\Rightarrow$ the degeneracy of zero-energy states equals the site imbalance between the two sublattices !

[M. Inui, S. A. Trugman, and E. Abrahams, PRB $\mathbf{49}$, 3190 (1994)]

Periodic systems: $\varepsilon = 0$ *flat bands* $\Longleftrightarrow$ site imbalance *per unit cell*

Graphene antidot lattices:

circular case $\quad N_A - N_B = 0 \quad \Longrightarrow \quad$ no flat bands

triangular case $\quad N_A - N_B = D \quad \Longrightarrow \quad D$ flat bands

Further implications of bipartite structure

The theorem of Inui et al. holds even when the hopping integral is random and/or not real!

Implication for the case with an **external magnetic field**

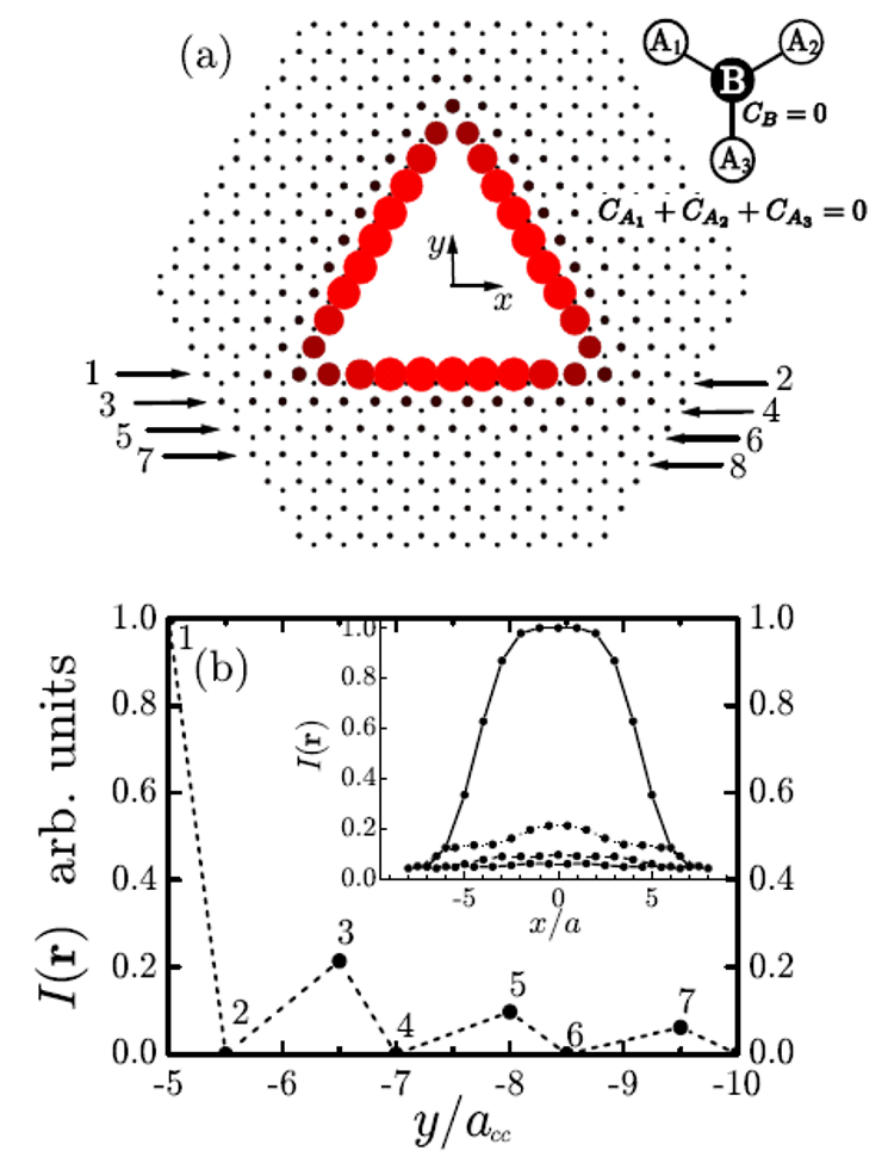
$$t \longrightarrow t_{ij} = t \, \exp\left(-\frac{ie}{\hbar c}\int_{\mathbf{r}_i}^{\mathbf{r}_j} \mathbf{A}\cdot \mathbf{dr}\right)$$

$\varepsilon = 0$ **flat bands remain flat!**

Lieb's theorem: ground state of the Hubbard model on an imbalanced bipartite lattice has total spin

$$S_{\mathrm{tot}} = \frac{1}{2}\,|N_A - N_B|$$

Zero-energy midgap states in real space



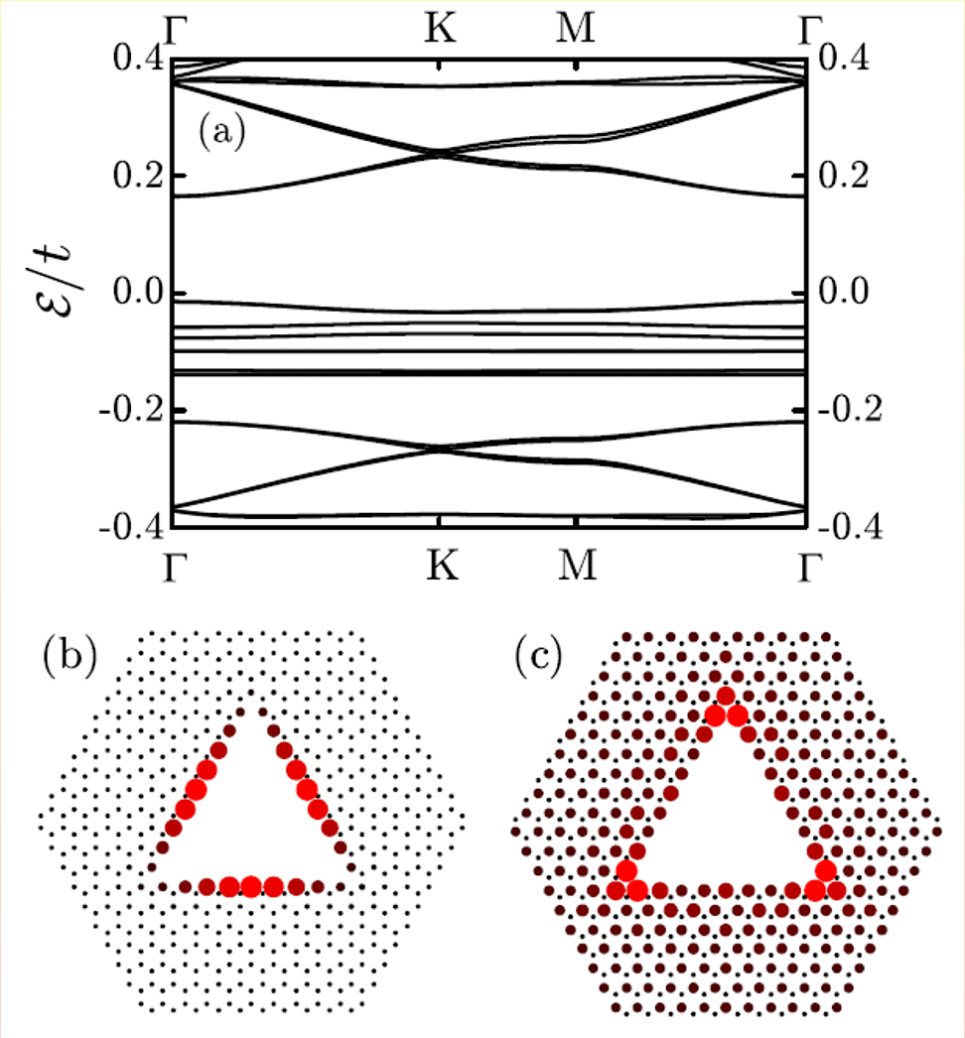
tunnelling current

$$\mathcal{I}(\mathbf{r}) \propto \int_{\varepsilon_F}^{\varepsilon_F + eV} d\varepsilon \; \rho(\mathbf{r}, \varepsilon)$$

$\rho(\mathbf{r}, \varepsilon)$ – local electronic DOS

the $\varepsilon = 0$ midgap states
are **pseudospin polarized**
and **exponentially localized**!

Influence of an on-site potential along the edges



$$\{L = 9, D = 6\}$$

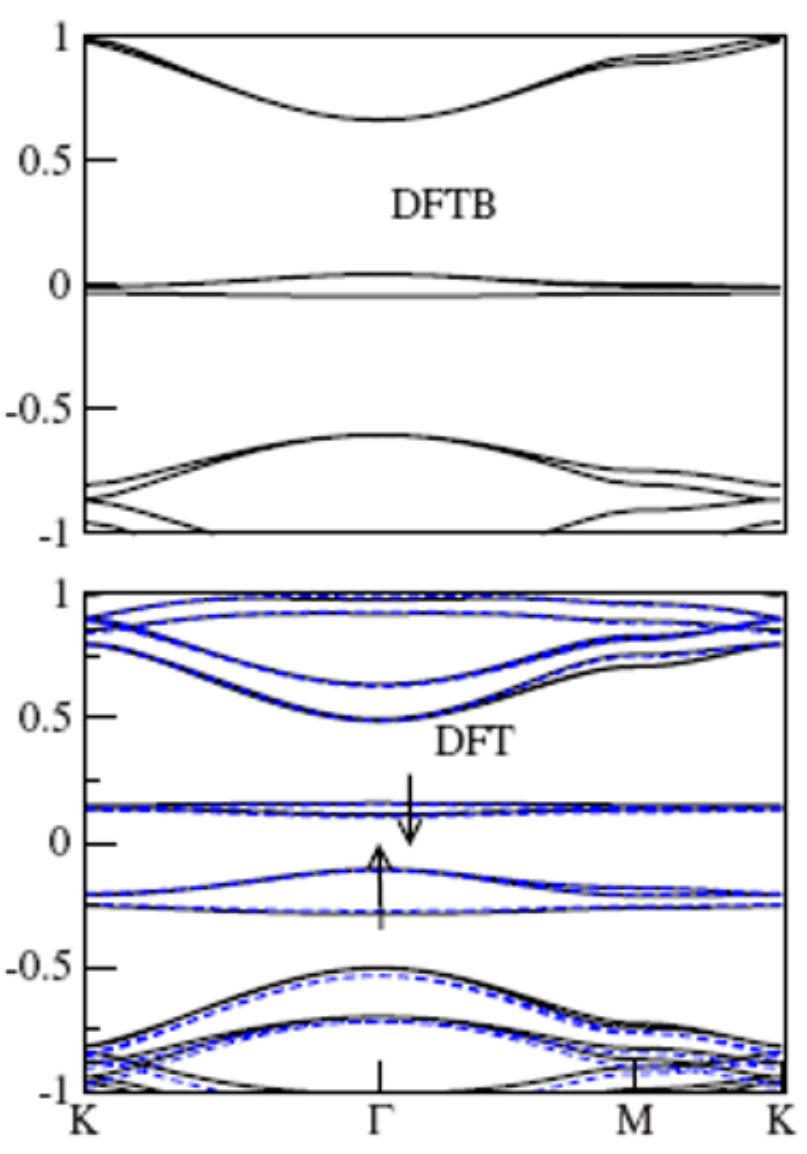
On-site potential mimics hydrogen passivation within a TB model !

Attractive on-site potential:

$$V = -0.15\,t$$

partially lifts the flat-band degeneracy and "flatness"

Beyond tight-binding model: $D = 3$ example

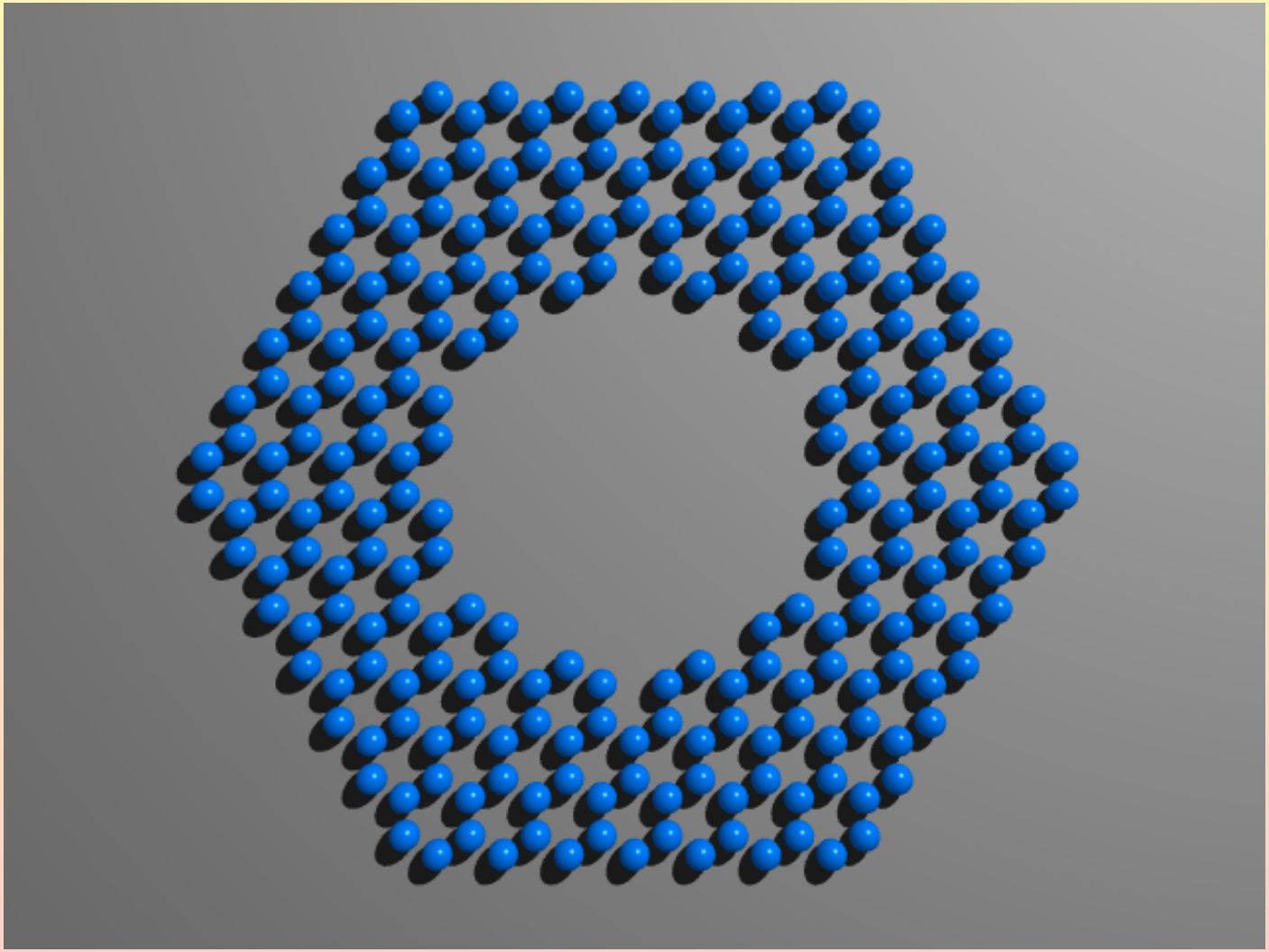


Calculations by J. A. Fürst *et al.*,
PRB **80**, 115117 (2009)

Conclusions:

- one completely flat
  + two low-dispersion bands!
- spin-splitting in the sDFT!
- Lieb's theorem still works!

Electron-phonon coupling in graphene antidot lattices



Motivation

QUESTION: **Worth of studying at all?**

Phonons in graphene are comparatively unimportant:

ARPES data on inelastic carrier lifetime explained
without even invoking phonon-related effects!

[A. Bostwick *et al.*, Nature Phys. **3**, 36 (2007)]

However, graphene antidot lattice is a totally different system:

narrow-band semiconductor

Bandgaps & bandwidths

Graphene antidot lattices are narrow-band systems!

$\{L, 5\}$ family:

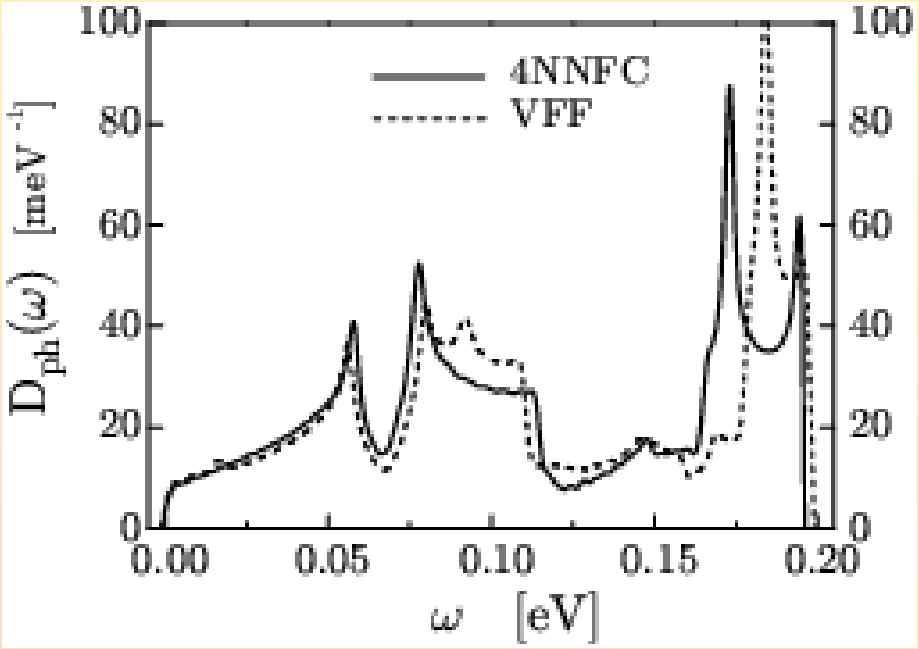
$$W_c = 0.11 - 0.14\,\text{eV}$$
(increases with L)

$$E_g = 0.18 - 0.74\,\text{eV}$$
(decreases with L)

Scaling law:

$$E_g \propto \frac{R}{L^2} = \frac{(R/L)}{L}$$

Phonons in graphene antidot lattices



$$D_{\mathrm{ph}}(\omega) \equiv N^{-1} \sum_{\mathbf{q},\lambda} \delta[\omega - \omega_{\lambda}(\mathbf{q})]$$

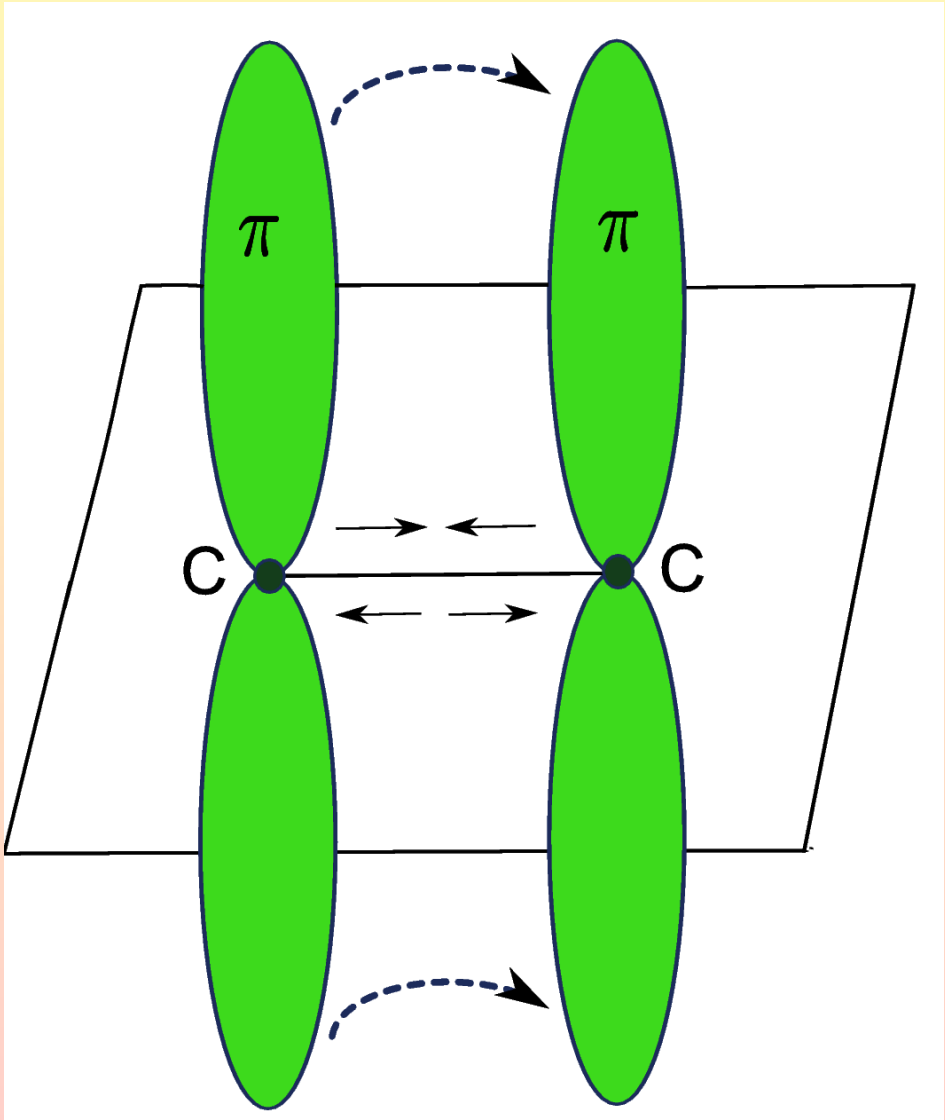
Two methods:

- **valence force field** (**VFF**)
  V. Perebeinos & J. Tersoff,
  PRB **79**, 241409(R) (2009)

- **4$^{\mathrm{th}}$ nn force constant** (**4NNFC**)
  J. Zimmermann *et al.*,
  PRB **78**, 045410 (2008)

Model parameters extracted from graphene data!

Peierls-type electron-phonon coupling



π-electron hopping integral is dynamically bondlength-dependent!

$$t \rightarrow t(\Delta u_{\mathrm{cc}}) = t + \alpha \Delta u_{\mathrm{cc}}$$

also known as:

- SSH coupling
- BLF coupling

Peierls-type electron-phonon coupling

phonon modulation of π-electron hopping integrals

$$t \longrightarrow t + \alpha \sum_{\lambda} \left[\hat{\mathbf{u}}_{\lambda,\mathbf{R}+\mathbf{d}_m+\delta} - \hat{\mathbf{u}}_{\lambda,\mathbf{R}+\mathbf{d}_m} \right] \cdot \bar{\delta}$$

$$\alpha = 5.27\,\mathrm{eV/\AA}\ ; \qquad \bar{\delta} \equiv \delta / \|\delta\|$$

electron-phonon coupling Hamiltonian in real space:

$$\hat{H}_{\mathrm{ep}} = \frac{\alpha}{2} \sum_{\mathbf{R},m,\delta,\lambda} \left(\hat{a}^{\dagger}_{\mathbf{R}+\mathbf{d}_m+\delta} \hat{a}_{\mathbf{R}+\mathbf{d}_m} + \mathbf{h.c.} \right) \left[\hat{\mathbf{u}}_{\lambda,\mathbf{R}+\mathbf{d}_m+\delta} - \hat{\mathbf{u}}_{\lambda,\mathbf{R}+\mathbf{d}_m} \right] \cdot \bar{\delta}$$

$$\hat{\mathbf{u}}_{\lambda,\mathbf{R}+\mathbf{d}_m} \equiv \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} \frac{e^{i\mathbf{q}\cdot\mathbf{R}} (\hat{b}^{\dagger}_{-\mathbf{q},\lambda} + \hat{b}_{\mathbf{q},\lambda})}{\sqrt{2M\omega_{\lambda}(\mathbf{q})}} \mathbf{v}^{\lambda}_{m}(\mathbf{q})$$

Electron-phonon coupling in momentum space

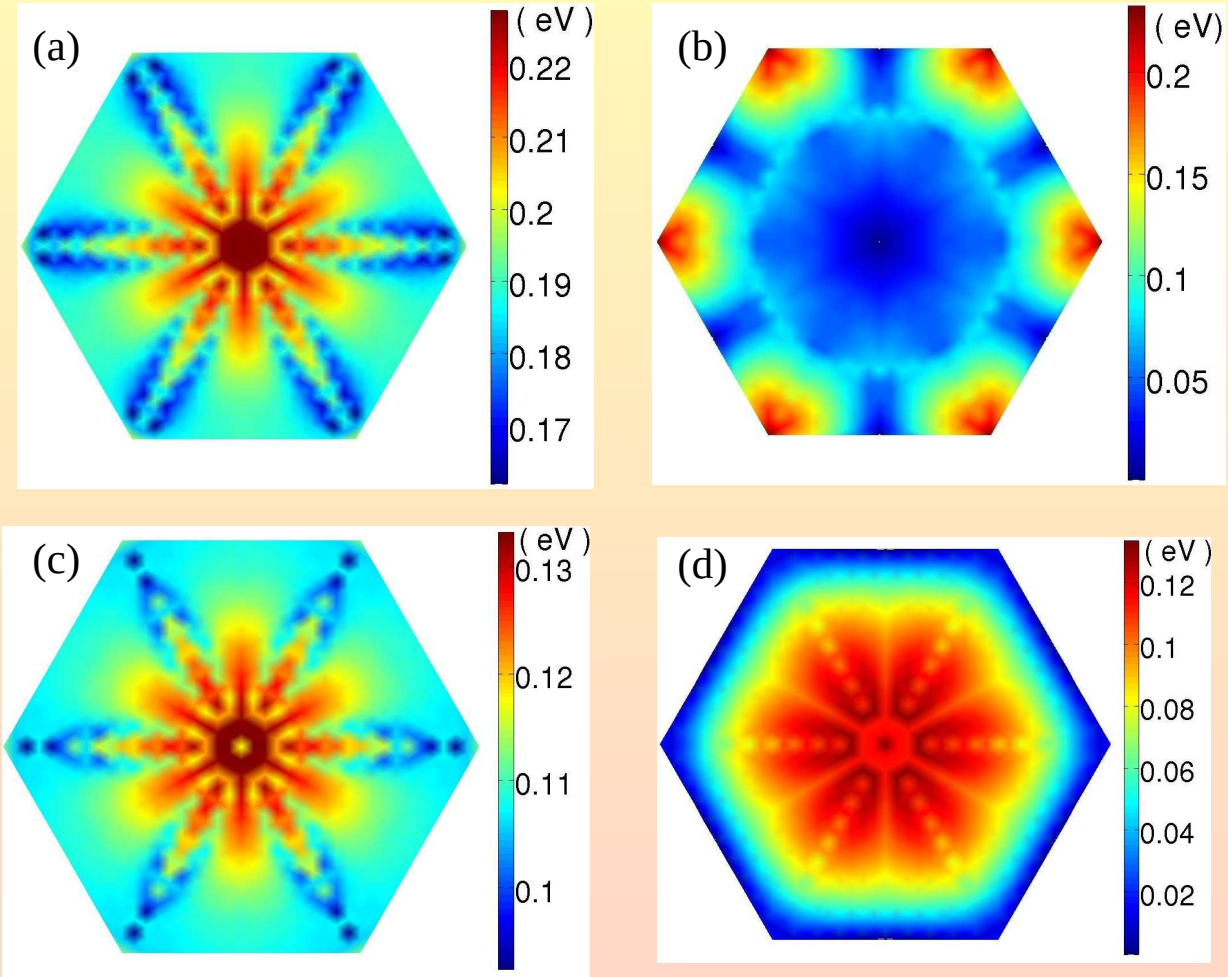
$$\hat{H}_{\rm ep} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k},\mathbf{q},\boldsymbol{\lambda},n} \gamma^{\boldsymbol{\lambda}}_{nn}(\mathbf{k},\mathbf{q})\, \hat{a}^{\dagger}_{n,\mathbf{k}+\mathbf{q}} \hat{a}_{n,\mathbf{k}} (\hat{b}^{\dagger}_{-\mathbf{q},\boldsymbol{\lambda}} + \hat{b}_{\mathbf{q},\boldsymbol{\lambda}})$$

vertex function: $\gamma^{\boldsymbol{\lambda}}_{nn}(\mathbf{k},\mathbf{q}) = V^{\boldsymbol{\lambda}}_{nn}(\mathbf{k},\mathbf{q}) + W^{\boldsymbol{\lambda}}_{nn}(\mathbf{k},\mathbf{q})$

$$V^{\boldsymbol{\lambda}}_{nn}(\mathbf{k},\mathbf{q}) = \frac{\alpha}{\sqrt{8M\omega_{\boldsymbol{\lambda}}(\mathbf{q})}} \sum_{m,\boldsymbol{\delta}} \bar{\boldsymbol{\delta}} \cdot [\mathbf{v}^{\boldsymbol{\lambda}}_{m+\boldsymbol{\delta}}(\mathbf{q}) - \mathbf{v}^{\boldsymbol{\lambda}}_{m}(\mathbf{q})] \times \left[(C^{n,\mathbf{k}+\mathbf{q}}_{m+\boldsymbol{\delta}})^{*} C^{n,\mathbf{k}}_{m} + (C^{n,\mathbf{k}+\mathbf{q}}_{m})^{*} C^{n,\mathbf{k}}_{m+\boldsymbol{\delta}} \right]$$

$$\hat{H}^{(\rm c)}_{\rm ep} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k},\mathbf{q},\boldsymbol{\lambda}} \gamma^{\boldsymbol{\lambda}}_{\rm cc}(\mathbf{k},\mathbf{q})\, \hat{a}^{\dagger}_{\rm c,\mathbf{k}+\mathbf{q}} \hat{a}_{\rm c,\mathbf{k}} (\hat{b}^{\dagger}_{-\mathbf{q},\boldsymbol{\lambda}} + \hat{b}_{\mathbf{q},\boldsymbol{\lambda}})$$

Strongly momentum-dependent electron-phonon coupling



important quantity:

$$|\gamma^{\lambda}_{cc}(\mathbf{k}=\mathbf{0},\mathbf{q})|$$

strongest coupling
to the highest-energy
optical phonon!

For this branch
$|\gamma^{\lambda}_{cc}(\mathbf{k}=\mathbf{0},\mathbf{q})|$
largest at $\mathbf{q}=\mathbf{0}$!

▸ RB

$\Longrightarrow$ **expect large mass enhancement!**

Comparison with other electron-phonon couplings

1. **momentum-independent couplings**

   Holstein-type (local) coupling: $\gamma(\mathbf{k},\mathbf{q}) = g = \text{const.}$

2. **momentum-dependent couplings**

   - SSH coupling on a square lattice:

     $\gamma_{\text{SSH}}(\mathbf{k},\mathbf{q}) \propto \sin(\mathbf{k}\cdot\mathbf{a}) - \sin\left[(\mathbf{k}+\mathbf{q})\cdot\mathbf{a}\right]$

     $\gamma_{\text{SSH}}(\mathbf{k}=\mathbf{0},\mathbf{q}) \propto |\mathbf{q}| \;\to\; 0 \quad (\mathbf{q}\to\mathbf{0})$

   - coupling to the "breathing" modes in cuprates:

     $\gamma(\mathbf{k},\mathbf{q}) = \gamma(\mathbf{q}) \propto \sqrt{\sin^2(q_x/2) + \sin^2(q_y/2)}$

     $\gamma(\mathbf{q}) \propto |\mathbf{q}| \;\to\; 0 \quad (\mathbf{q}\to\mathbf{0})$

Quasiparticle weight and effective mass

GENERAL: ratio of the effective ($m_{\rm eff}$) and bare band ($m_{\rm e}^*$) masses

$$\frac{m_{\rm eff}}{m_{\rm e}^*} = \lim_{\mathbf{k}\to\mathbf{0}} \frac{\varepsilon(\mathbf{k}) - \varepsilon(0)}{E(\mathbf{k}) - E(0)}$$

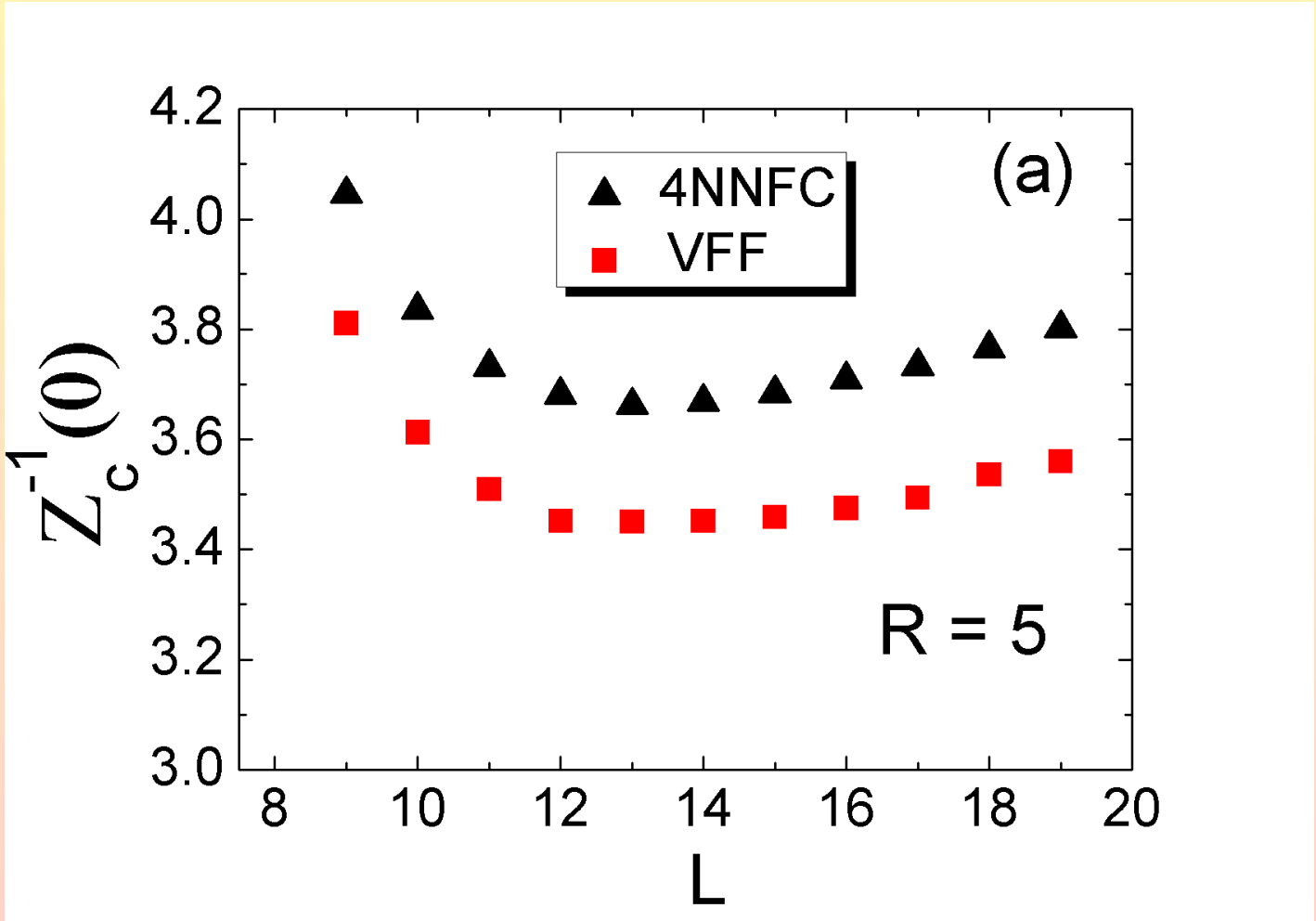
renormalized dispersion: $E(\mathbf{k}) = \varepsilon(\mathbf{k}) + \mathbf{Re}\,\Sigma[\mathbf{k}, E(\mathbf{k})]$

$$Z^{-1}(0) = 1 - \frac{\partial}{\partial\omega}\Big[\,\mathbf{Re}\,\Sigma(\mathbf{k},\omega)\Big]\Big|_{\mathbf{k}=0,\,\omega=E_0}$$

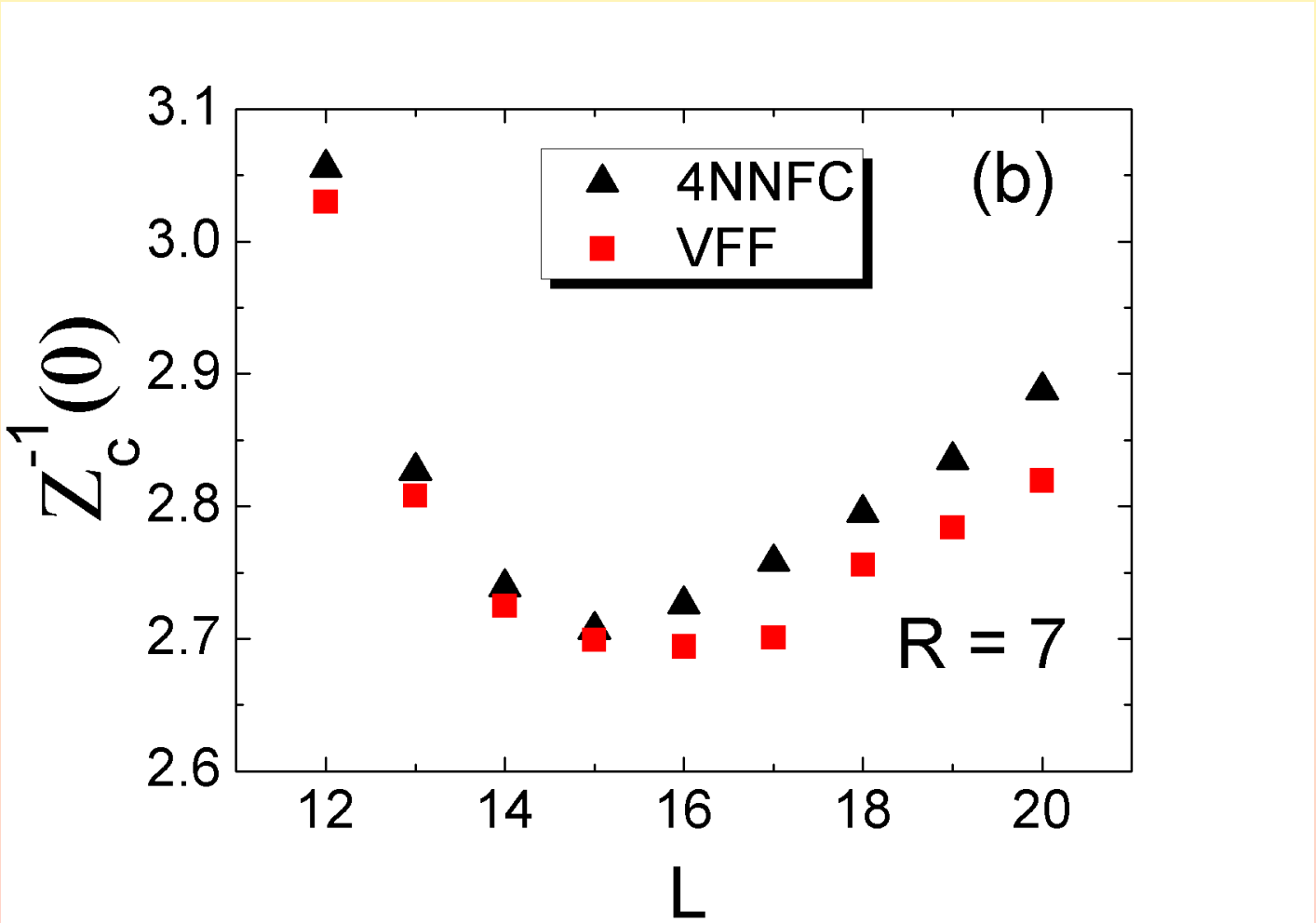
Rayleigh-Schrödinger perturbation theory:

$$Z_{\rm c}^{-1}(0) = 1 + \frac{1}{N}\sum_{\mathbf{q},\lambda} \frac{|\gamma_{\rm cc}^{\lambda}(\mathbf{k}=0,\mathbf{q})|^2}{\left[\varepsilon_{\rm c}(0) - \varepsilon_{\rm c}(\mathbf{q}) - \omega_\lambda\right]^2}$$

Strong phonon-induced renormalization (I)



Strong phonon-induced renormalization (II)



Large electron-phonon mass enhancement

$$\left(\frac{m_{\rm eff}}{m_{\rm e}^*}\right)_{\alpha} = \frac{1 - \dfrac{\partial}{\partial\omega}\,\mathrm{Re}\,\Sigma_{\rm c}(\mathbf{k},\omega)\big|_{\mathbf{k}=0,\omega=E_{\rm c}(0)}}{1 + \dfrac{\partial}{\partial\varepsilon_{\rm c}(\mathbf{k}_{\alpha})}\,\mathrm{Re}\,\Sigma_{\rm c}(\mathbf{k}_{\alpha},\omega)\big|_{\mathbf{k}_{\alpha}=0,\omega=E_{\rm c}(0)}}$$

$$\lambda_{\rm me}^{(\alpha)} = \frac{Z_{\rm c}^{-1}(0)}{1 + \dfrac{\partial}{\partial\varepsilon_{\rm c}(\mathbf{k}_{\alpha})}\,\mathrm{Re}\,\Sigma_{\rm c}(\mathbf{k}_{\alpha},\omega)\big|_{\mathbf{k}_{\alpha}=0,\omega=E_{\rm c}(0)}} - 1$$

example: {9,5} lattice – $\lambda_{\rm me}^{(x)} = 2.411$, $\lambda_{\rm me}^{(y)} = 2.448$

Are there polarons in this system?

- Standard criteria for polaron formation not amenable to testing;
- short-range, nonpolar e-ph coupling (covalently-bonded systems)
  $\Rightarrow$ carriers are quasifree electrons or small polarons!
- interface with polar substrates (SiC, SiO_2):
  interplay of Peierls-type and Fröhlich-type coupling

  G. De Filippis, V. Cataudella, S. Fratini, and S. Ciuchi, arXiv:1005.2476 (2010)

Conclusions and Outlook

- Graphene antidot lattices have completely/nearly flat bands as a result of bipartite structure and sublattice site imbalance
- Phonon-induced mass renormalization in graphene antidot lattices is very strong – onset of polaronic behavior
- Study more realistic graphene antidot lattices using simplified (continuum) approaches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