# Tunable atomic collapse in mono- and bilayer graphene

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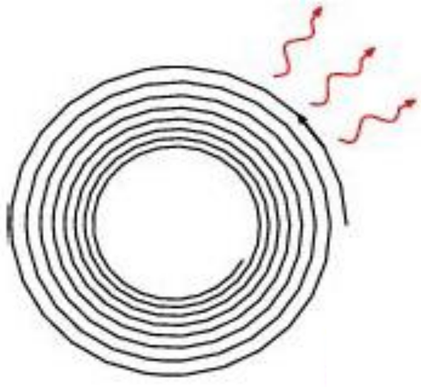
# **Atoms** (Kepler problem)

Classical physics
→ unstable planetary atoms



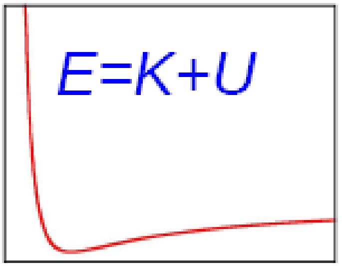
Quantum theory (Bohr, 1913)
→ stable orbits (wave nature of electrons)

Rydberg formula:

$$E_n = -\frac{me^4 Z^2}{2\hbar^2 n^2}$$

Lower bound: $E_1 = -\frac{me^4 Z^2}{2\hbar^2}$

Heisenberg (1926) : *uncertainty principle* $\Delta p \Delta x \geq \hbar/2$




$$K_{\mathrm{nr}} = \frac{p^2}{2m} \sim \frac{\hbar^2}{2mr^2} \gg U = -\frac{Ze^2}{r}$$

**Planetary atom stabilized by QM (zero point motion)**

# Scaling arguments

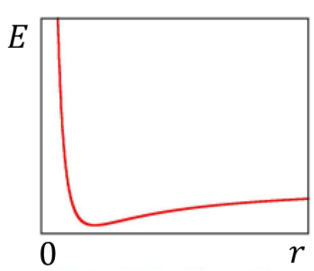
Usually:

$$U = -\frac{Ze^2}{r} \qquad U \propto -\frac{1}{r}$$

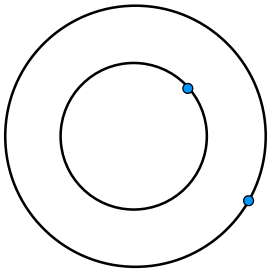
$$K = \frac{p^2}{2m} \sim \frac{\hbar^2}{2mr^2} \qquad K \propto \frac{1}{r^2}$$

$\boldsymbol{r \to 0}$:

- $|K| \gg |U|$
- $\mathrm{E} = K + U \to \infty$




stable orbitals



With relativity:

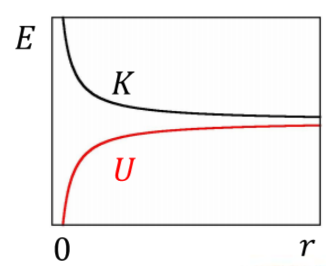
$$U = -\frac{Ze^2}{r} \qquad U \propto -\frac{1}{r}$$

$$K = cp \sim \frac{c\hbar}{r} \qquad K \propto \frac{1}{r}$$

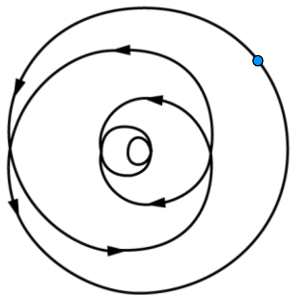
$\mathrm{Ze}^2 \leftrightarrow \mathrm{c}\hbar \quad \Rightarrow \quad \boldsymbol{\frac{Ze^2}{c\hbar} = Z\alpha}$

$\boldsymbol{r \to 0}$:

- $|K|$ ?? $|U|$
- $K + U =$ ??



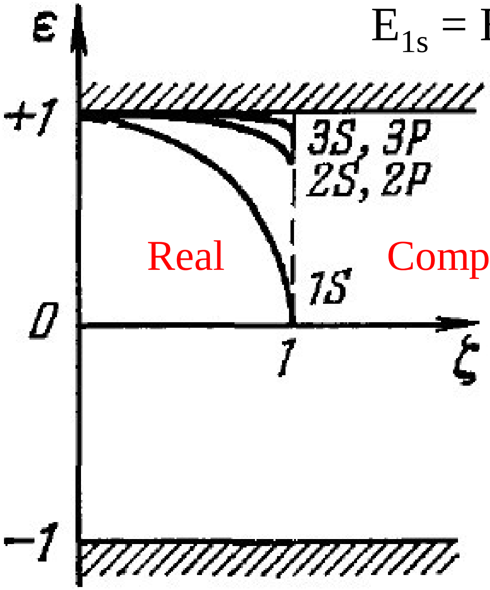

collapsing orbitals ?



# Dirac atoms can implode

***Dirac (1929)***

$E_{1s} = E_g\{1-(Z\alpha)^2\}^{1/2}$




$$\zeta \equiv \frac{Ze^2}{\hbar c} = Z\alpha$$

$\alpha = 1/137$

Subcritical (Z < 137)

Complex energies when $\zeta > 1$

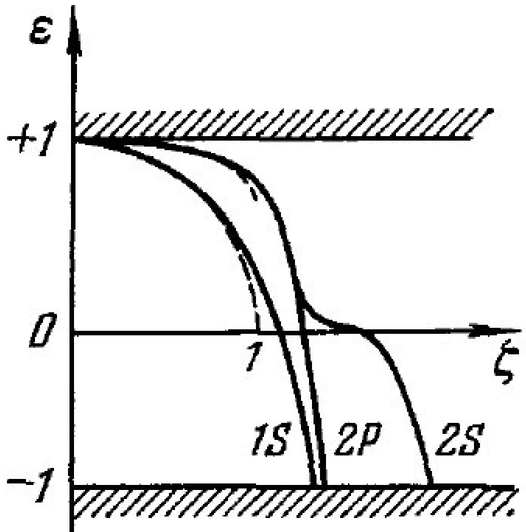
***Pomeranchuk & Smorodinskii (1945)***
***Werner and Wheeler (1957)***

Finite size of nucleus
Pomeranchuk nuclear form factor

$r_0 = 1.2\ 10^{-12}$ cm

1s level dives into Dirac sea at Z=170

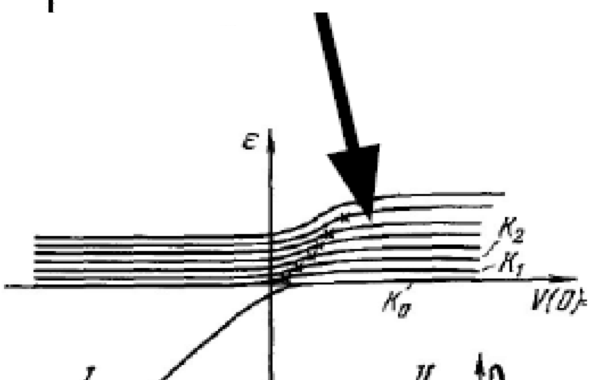
# Supercritical atoms




*Gershteyn, Zeldovich (1969)*
*Popov (1970)*



Z > $Z_c$=170 Collapse
→ vacuum reconstruction

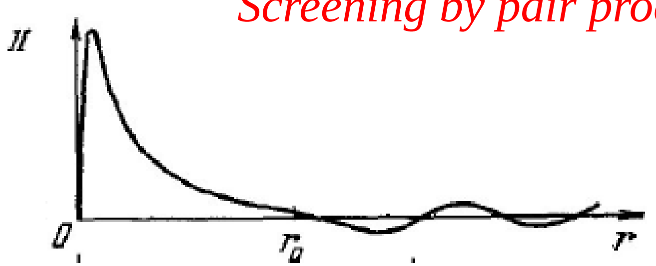



*Resonance states in the Dirac sea*

E → E=$E_0$ - iγ with γ ~ exp(-b/(Z-$Z_c$)$^{1/2}$)

Quasi-localized spatial structure of the resonance states

*Screening by pair production?*

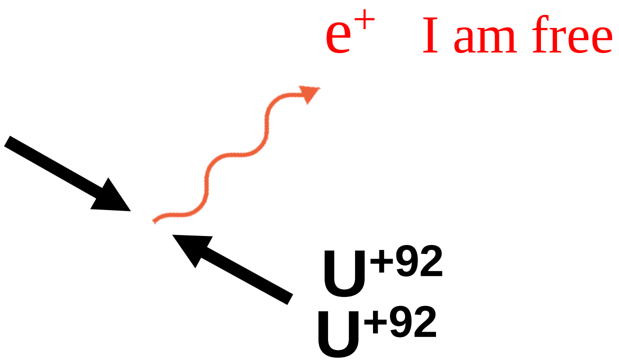



The electrons collapse into the nucleus, where they would then eject positrons, which would spiral outward and away.

# Experiment

Collision of heavy ions

- Darmstadt experiments (1980s & 1990s)
- Uranium (Z = 92)
- 3-6 MeV collisions




Signature of atomic collapse: **positron emission**

*>> No signature of supercritical emission <<*

# Graphene

3D atoms: $\zeta \equiv \frac{Ze^2}{\hbar c} = Z\alpha$ → critical value $Z\alpha \approx 1$ → $Z_c \sim 137$

$Z_c >$ existing nuclei ($Z_{max} < 120$)

Graphene: $c \rightarrow v_F$ $\alpha \rightarrow \alpha_{eff} = \alpha(c/v_F) \approx 2.5$ *Large effective fine structure constant*

$\varepsilon_0 \rightarrow \varepsilon$ (graphene + environment) $e^2 \rightarrow e^2/\varepsilon$

*Scaling of effective charge*

$\beta = \alpha_{eff}\,(Z/\varepsilon)$ → critical value $\beta_c \approx 1/2$ → $Z_c \approx \varepsilon/2\alpha_{eff} \sim 1$

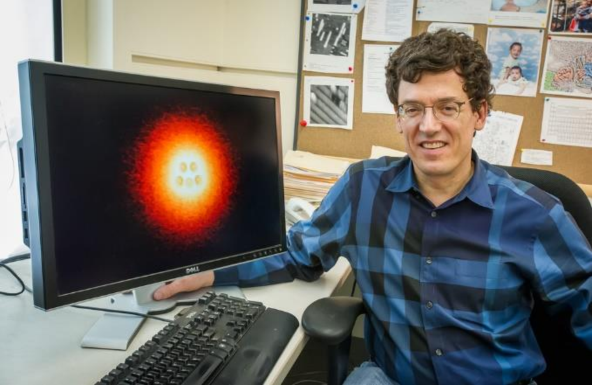
$m \rightarrow 0$ massless Dirac equation: continuum spectrum

→ atomic-spectrum : no discrete spectrum

**Manifestion of collapse: formation of resonances (quasi-localized spatial structure)**



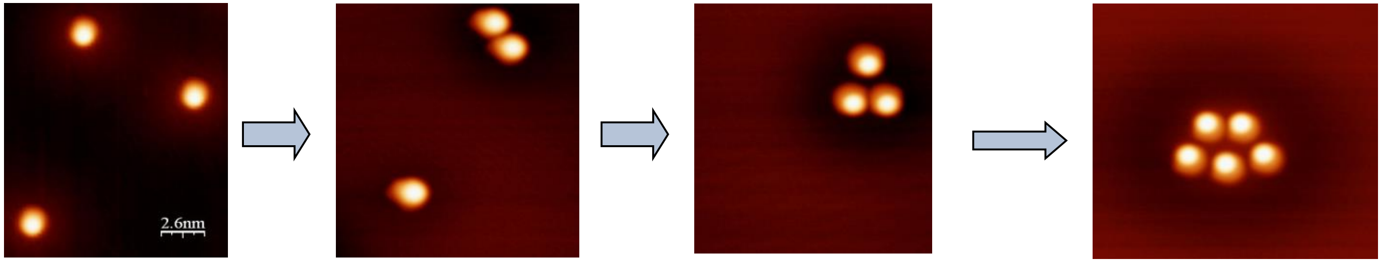
A.V. Shytov et al, PRL **99**, 246802 (2007)



# Atomic collapse: graphene

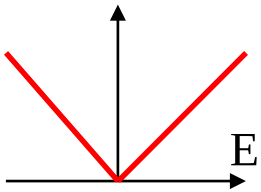
Y. Wang *et al*, Science **340**, 734 (2013)

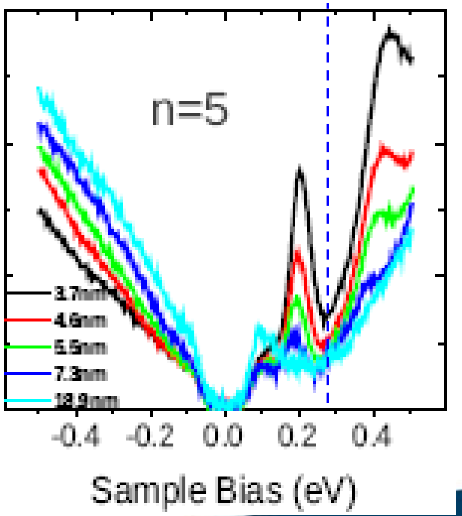
**Ca Dimers are moveable charge centers (M. Crommie group, Berkeley)**




STM → I(V)

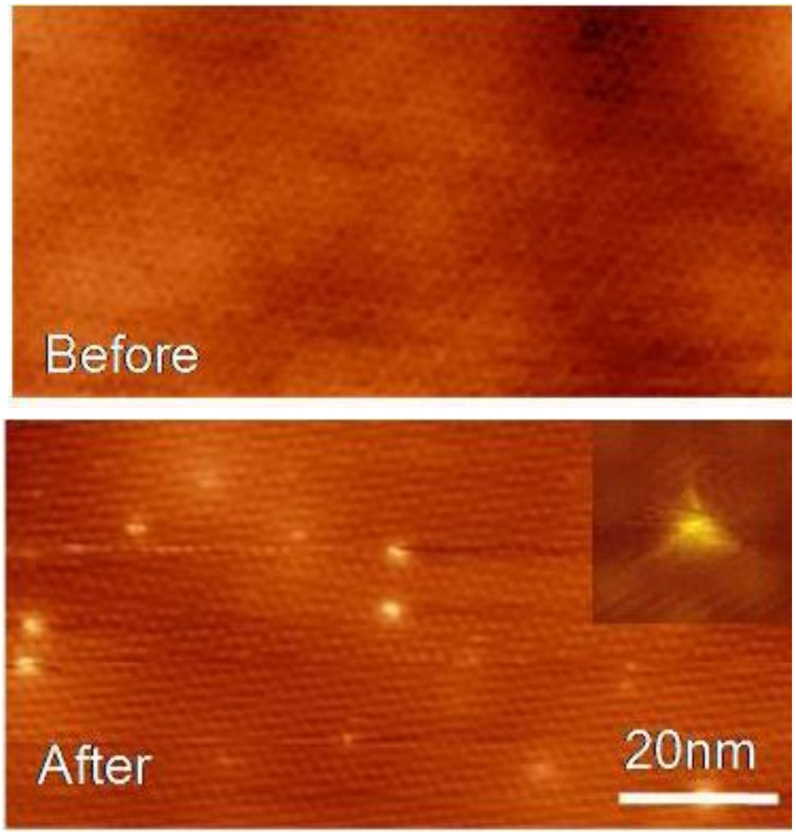
dI/dV ≈ DOS(E)

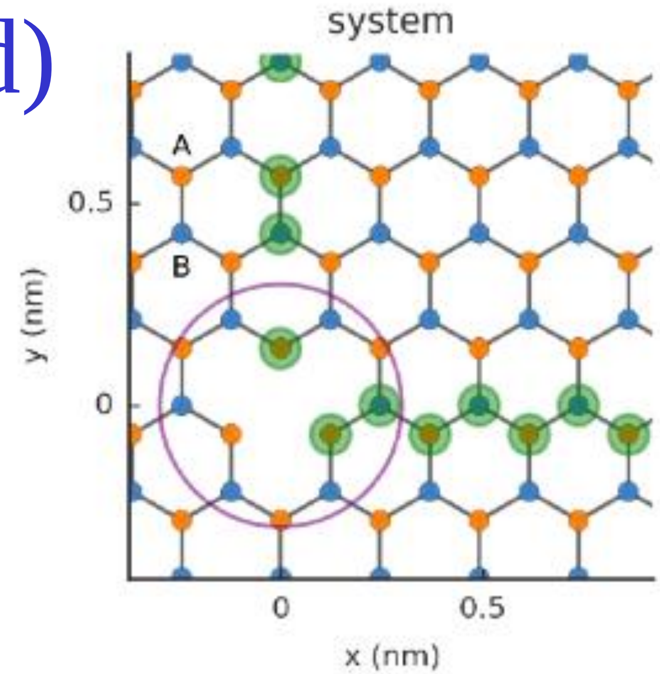


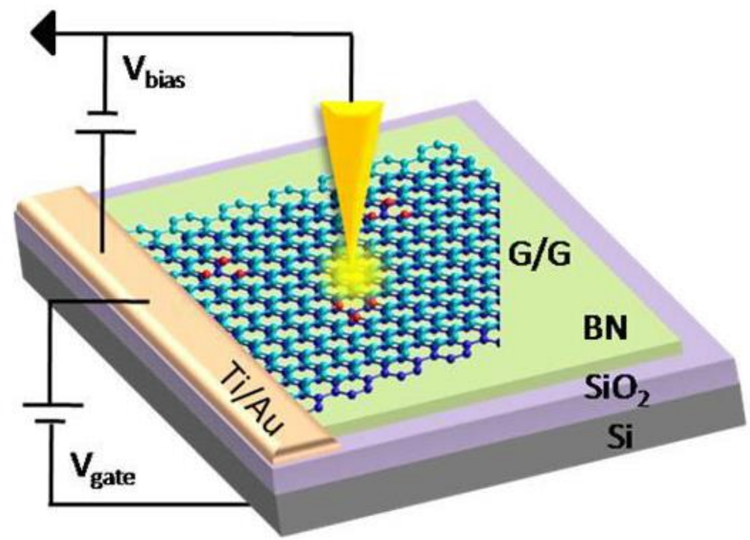




BUT: - nucleus has finite size
- nucleus vertical displaced from plane of electrons
- only one resonance

# Vacancy (charged)



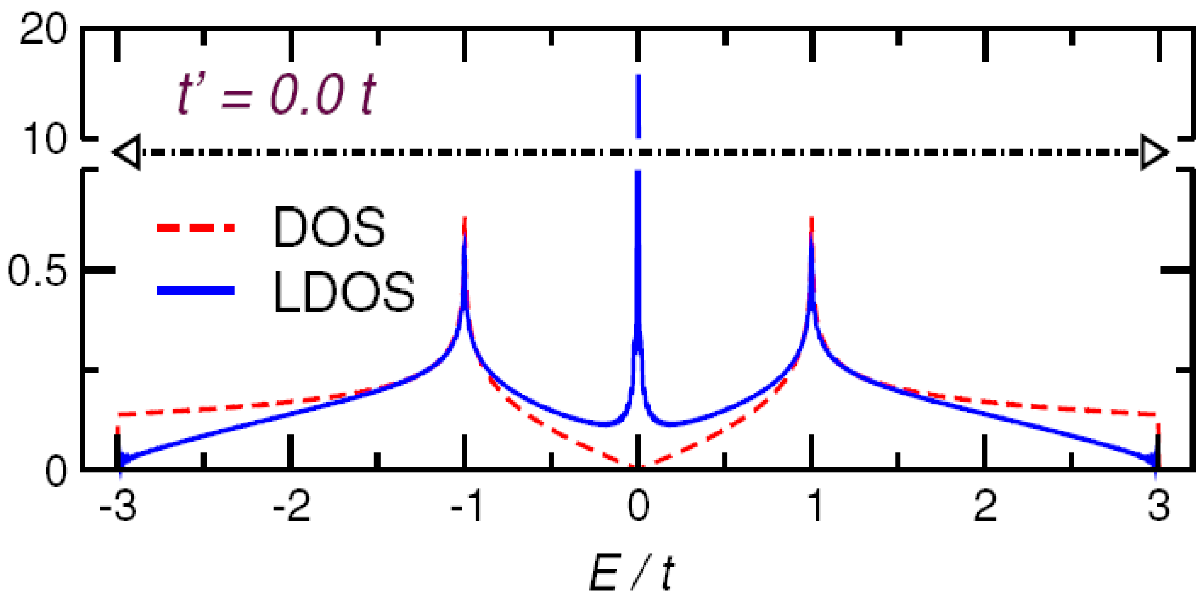




Rutgers University
(Andrei-group)

PRL **96,** 036801 (2006) PHYSICAL REVIEW LETTERS week ending 27 JANUARY 2006

# Disorder Induced Localized States in Graphene

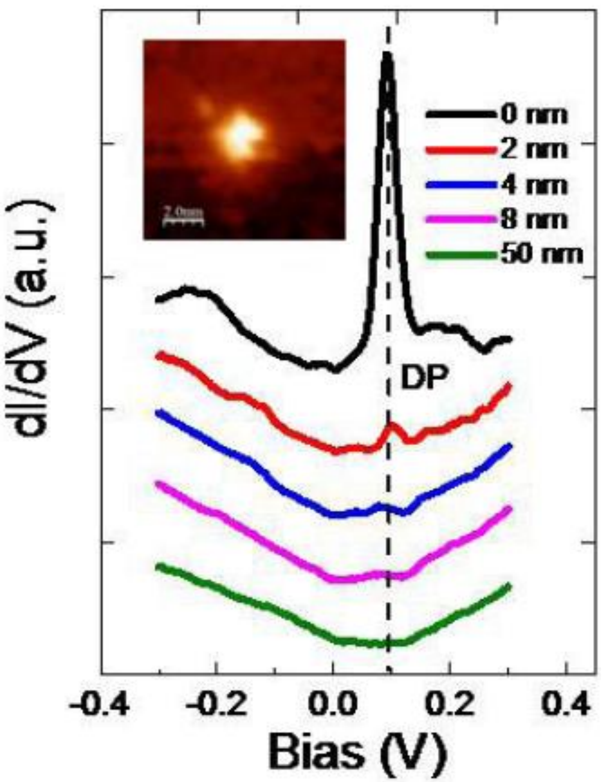
Vitor M. Pereira,[1,2] F. Guinea,[1,3] J. M. B. Lopes dos Santos,[2] N. M. R. Peres,[1,4] and A. H. Castro Neto[1]



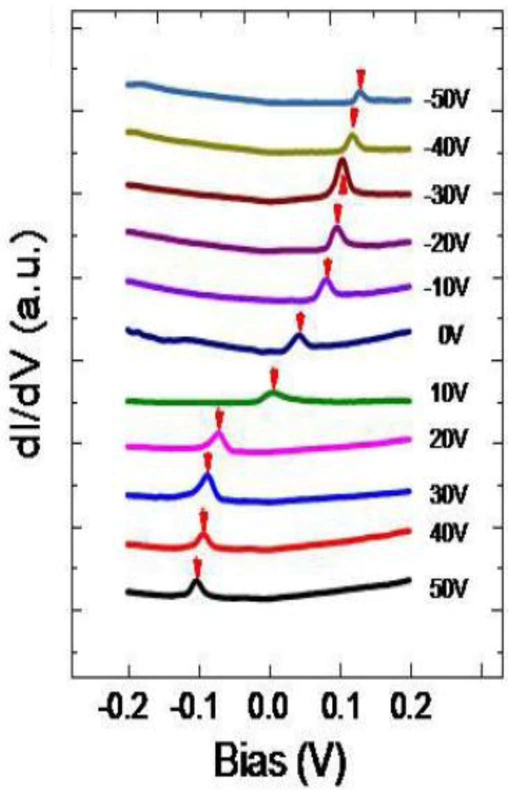

Vacancy → resonance close to the Fermi level
(half filled case)

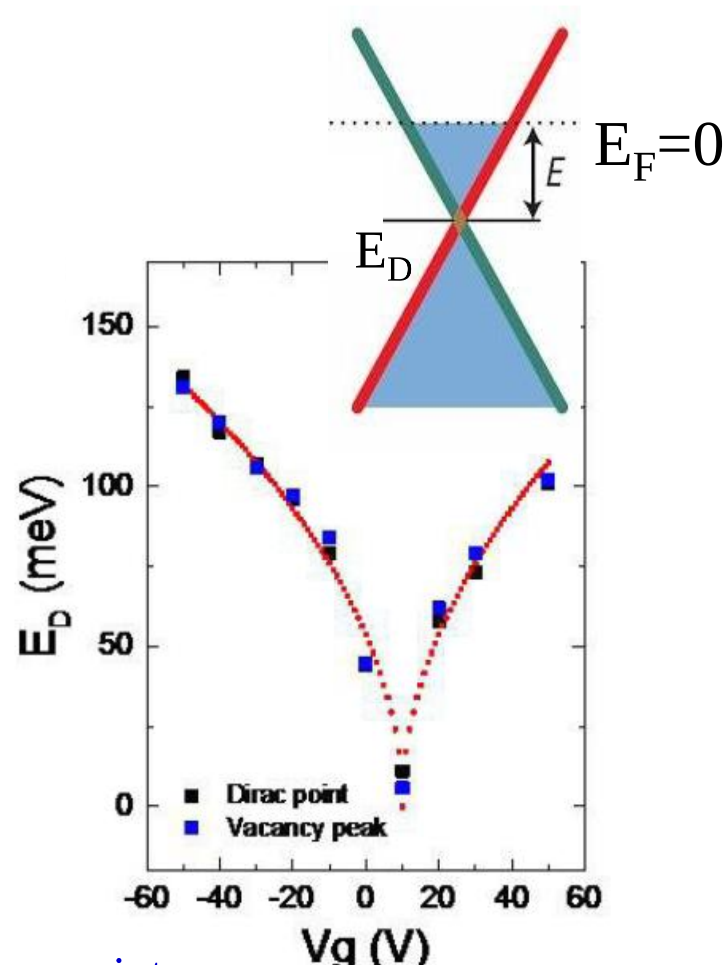
# Vacancy peak

Spatial dependence




Doping dependence

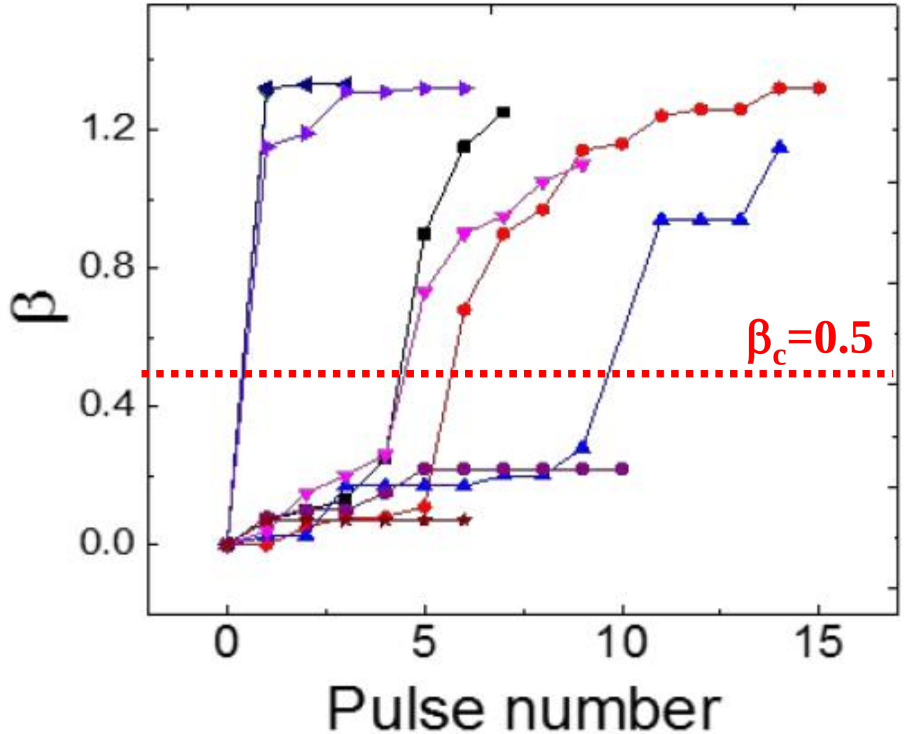


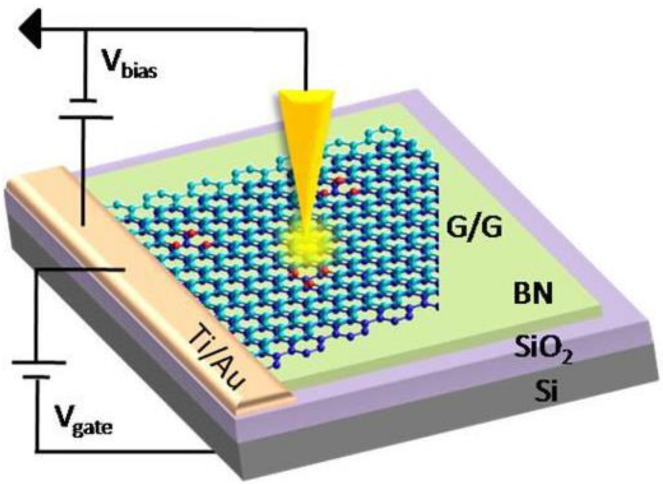




Localized on the vacancy site

It tracks the Dirac point

# Charging vacancy in graphene



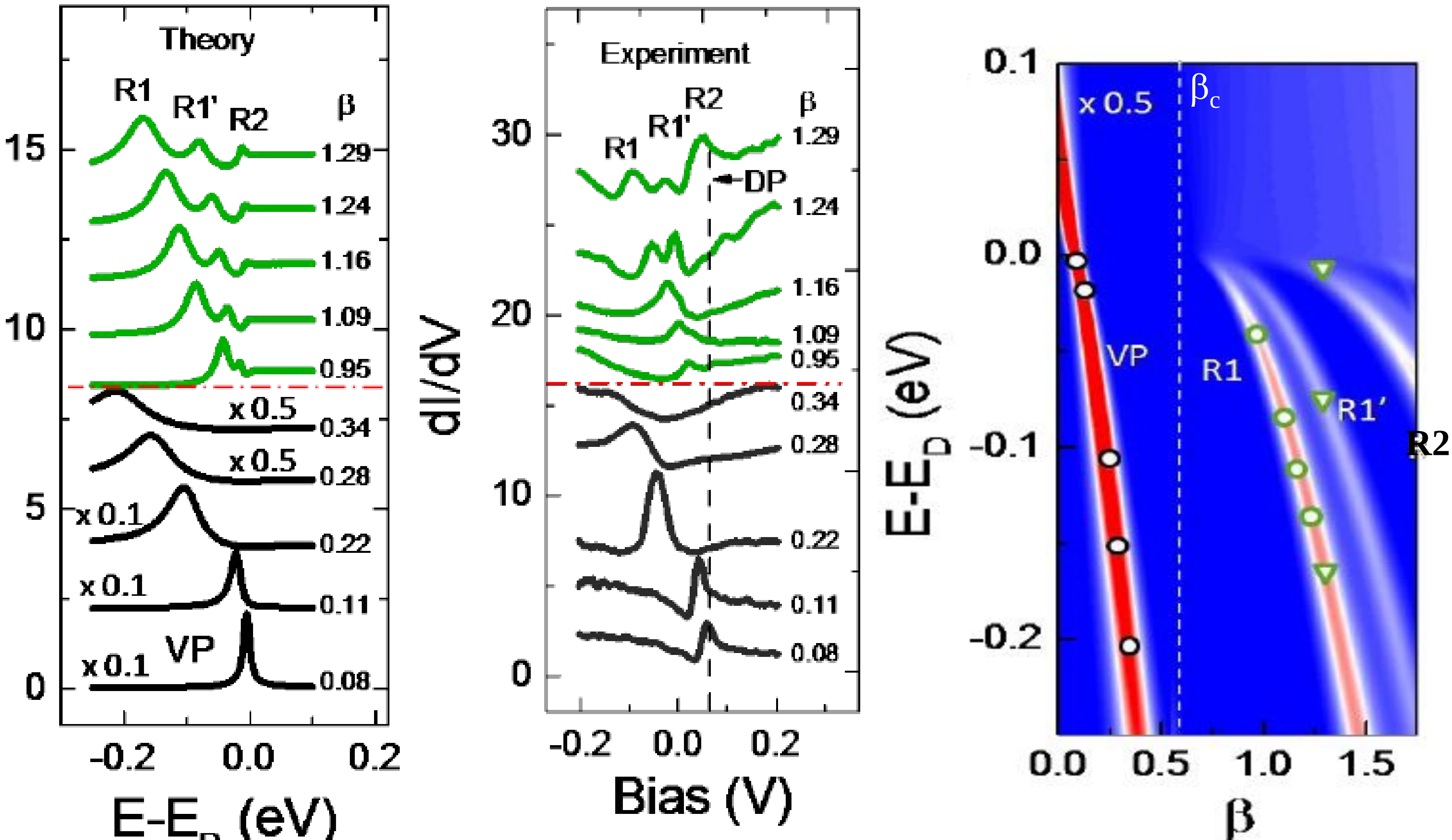




$\beta = \alpha(c/v_F)(Z/\varepsilon)$

# Spectrum

$$\beta = \frac{Z}{\kappa}\alpha_g = \frac{Z}{\kappa}\frac{1}{137}\frac{c}{v_F}$$

LDOS

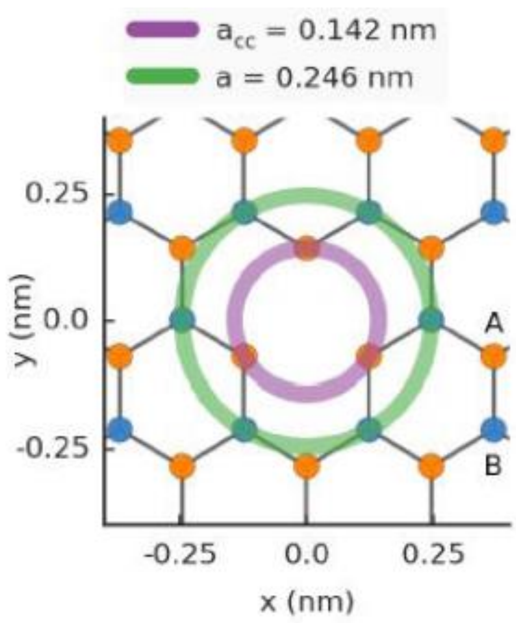


# Modelling electronic properties

# Theory

$$H = t\sum_i (a_i^\dagger b_i + H.c.) + \sum_i (V(r_i^A)a_i^\dagger a_i + V(r_i^B)b_i^\dagger b_i)$$

Tight-binding hamiltonian (including next nearest neighbor interactions)

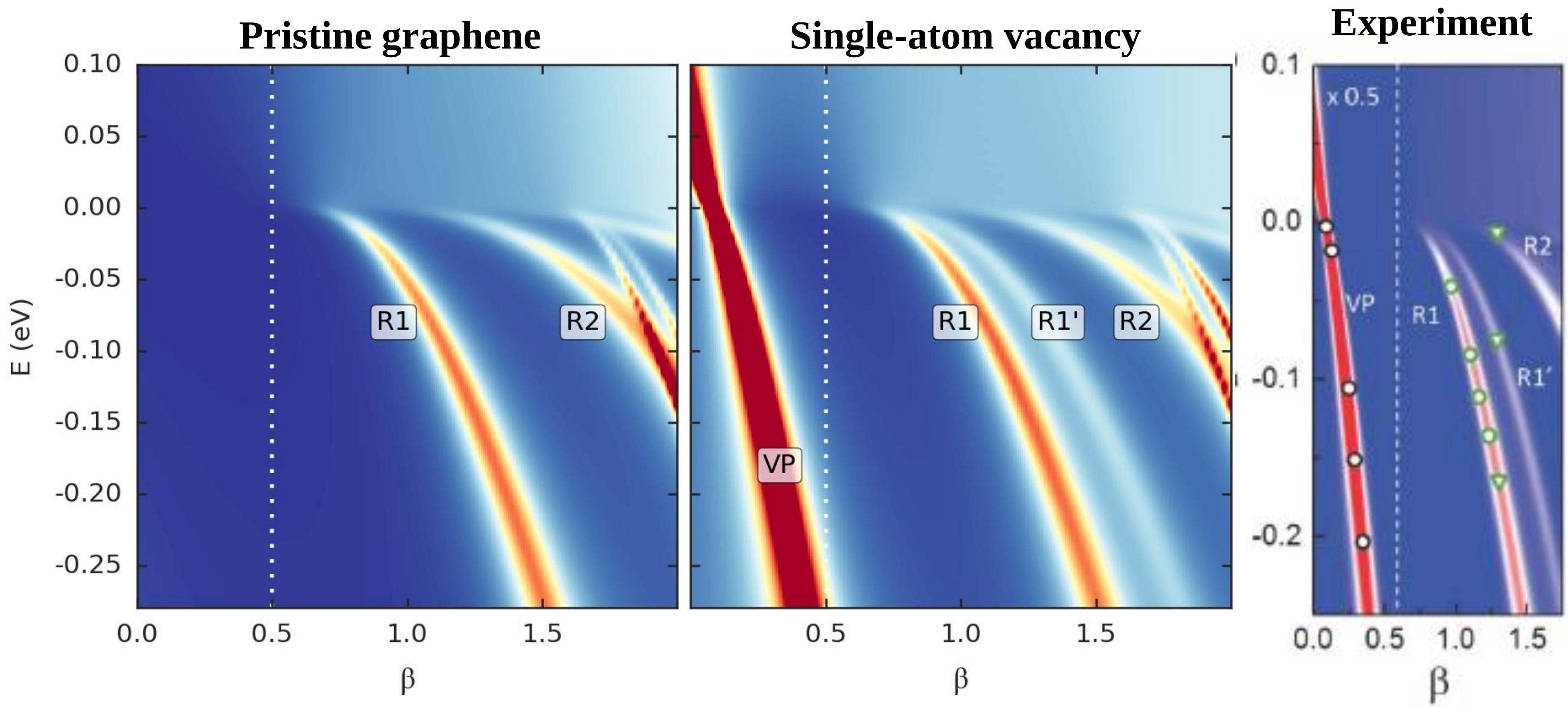



$$V(r) = \begin{cases} -\hbar v_F \frac{\beta}{r_0}, \text{ if } r \leq r_0 \\ -\hbar v_F \frac{\beta}{r}, \text{ if } r > r_0 \end{cases}$$

$r_0 = 0.5$ nm

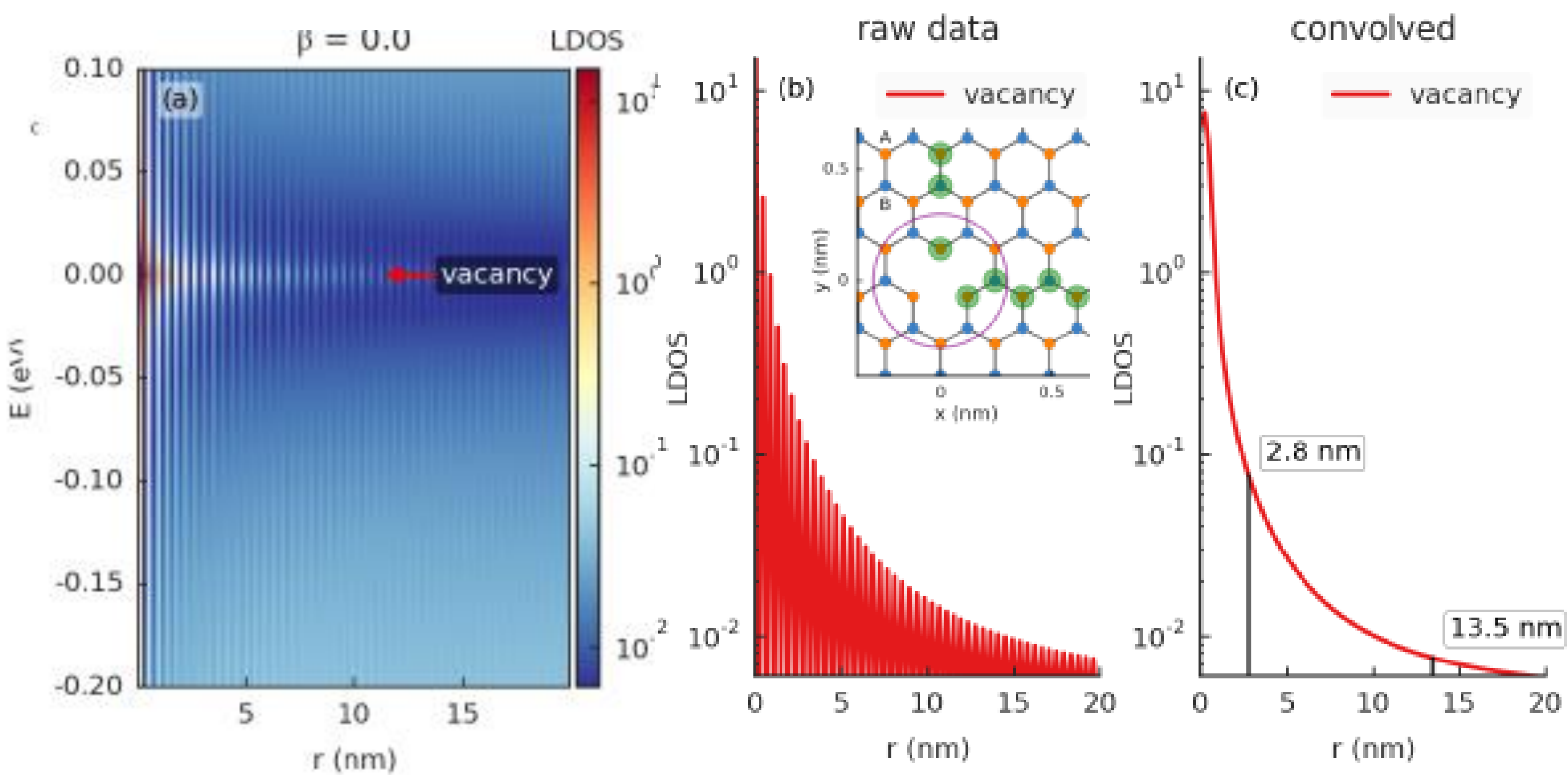
→ Exact numerical solution for a hexagonal sample

# Dirac versus TB approach



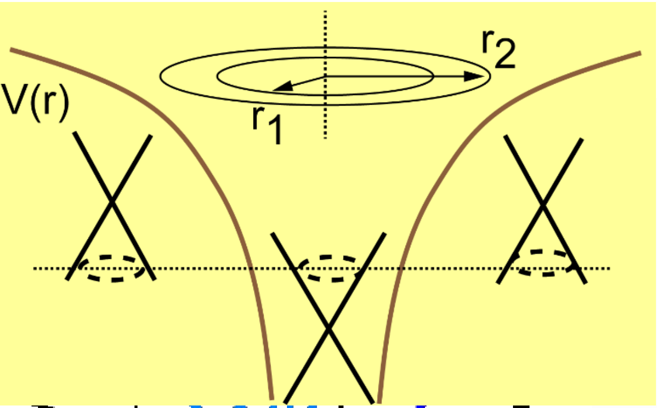

Qualitative differences: - VP-state
- R1’ - state

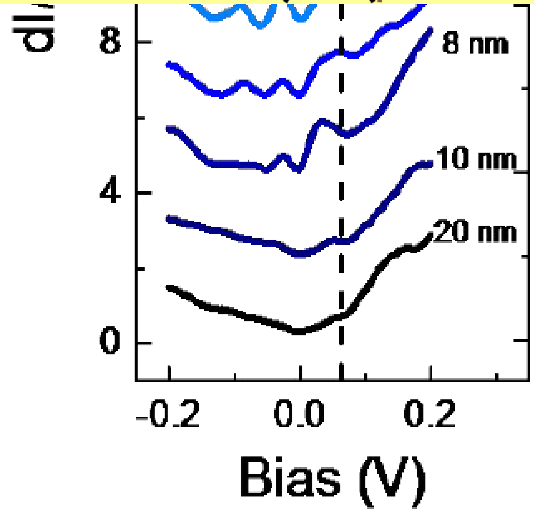
# Vacancy peak




Vacancy: atom from sublattice B is removed → sublattice symmetry is broken
VP-state is localized on sublattice A (and LDOS is zero on sublattice B)

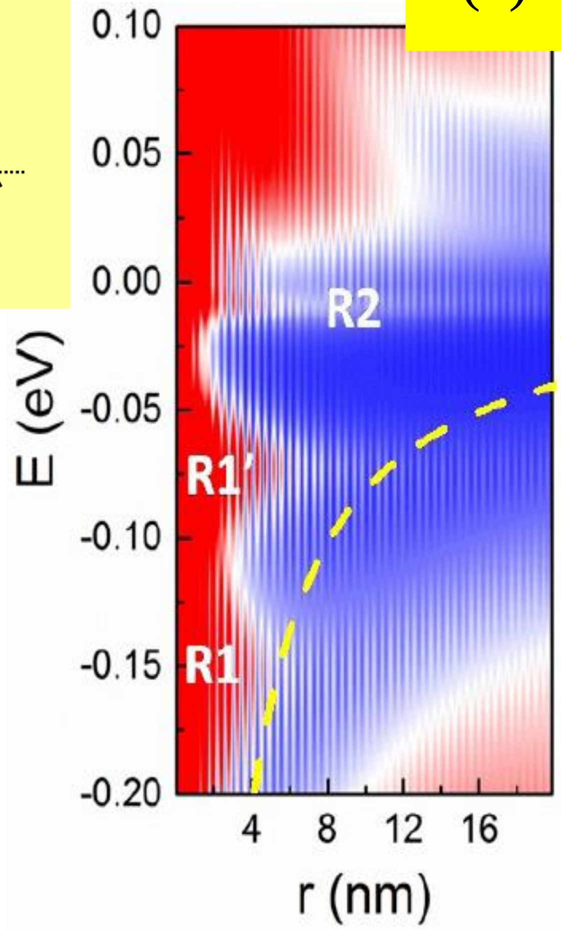
# Spatial dependence

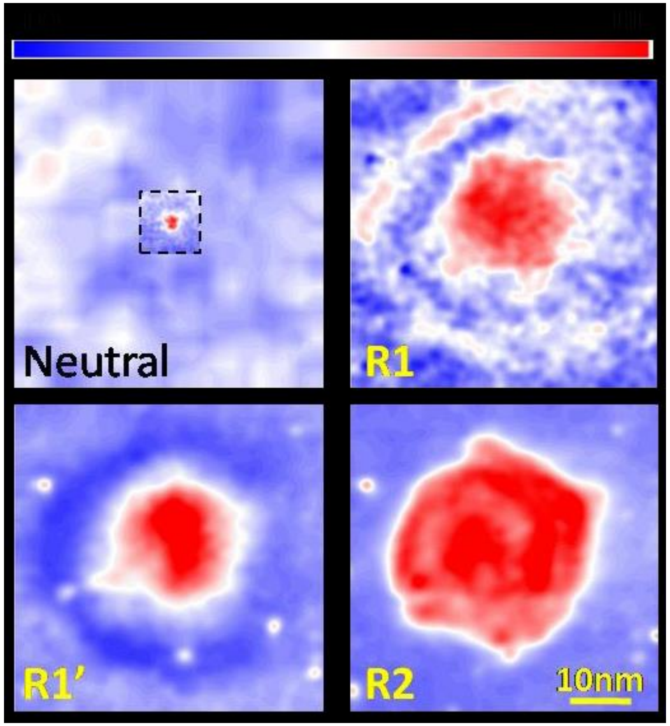



$$U(r) = \beta \frac{h v_F}{r}$$

➔ Local Dirac point

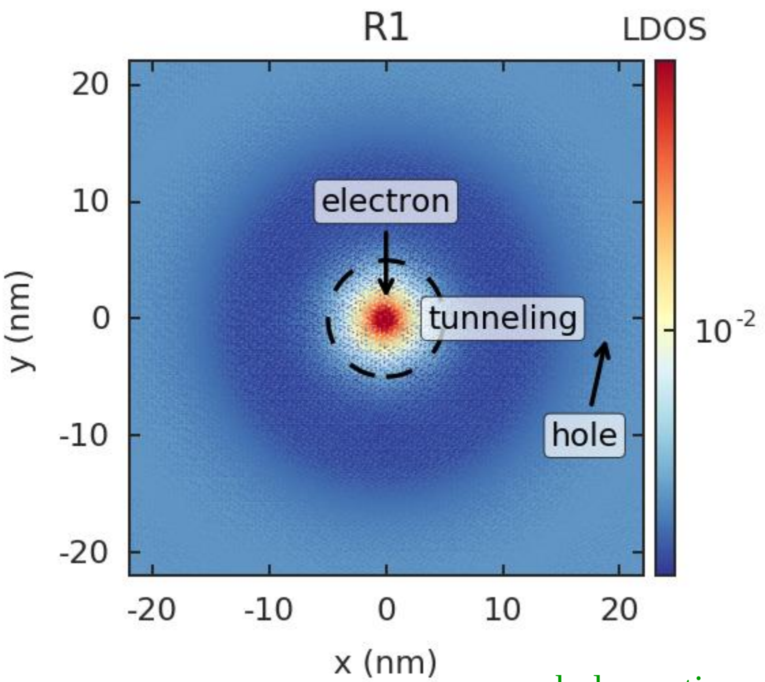



Collapsed states extend far beyond the vacancy site

Vacuum polarization

# LDOS for collapse resonance R1

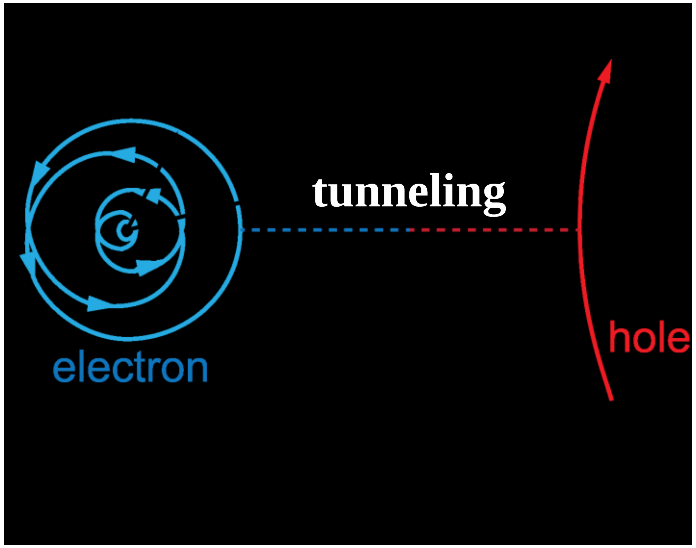



hole continuum

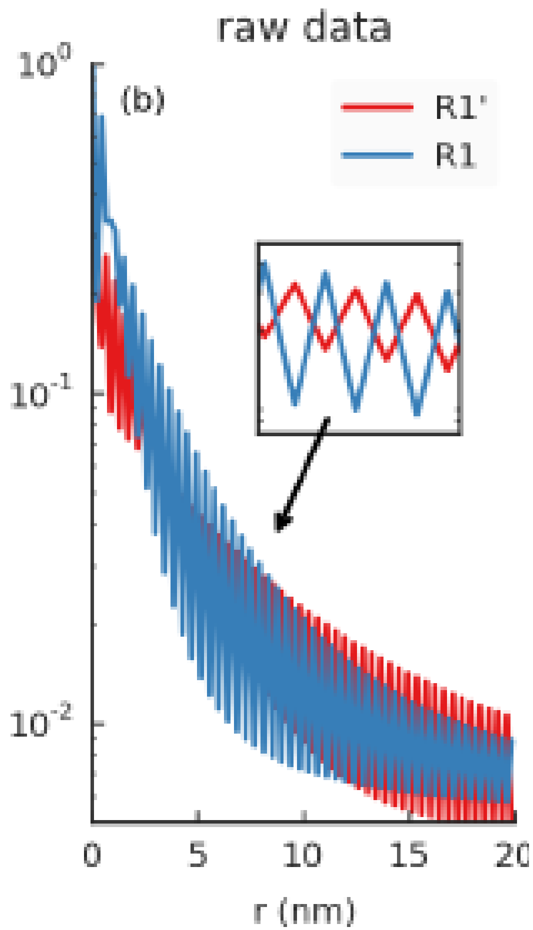
quasi-bound electron inside Coulomb pot.

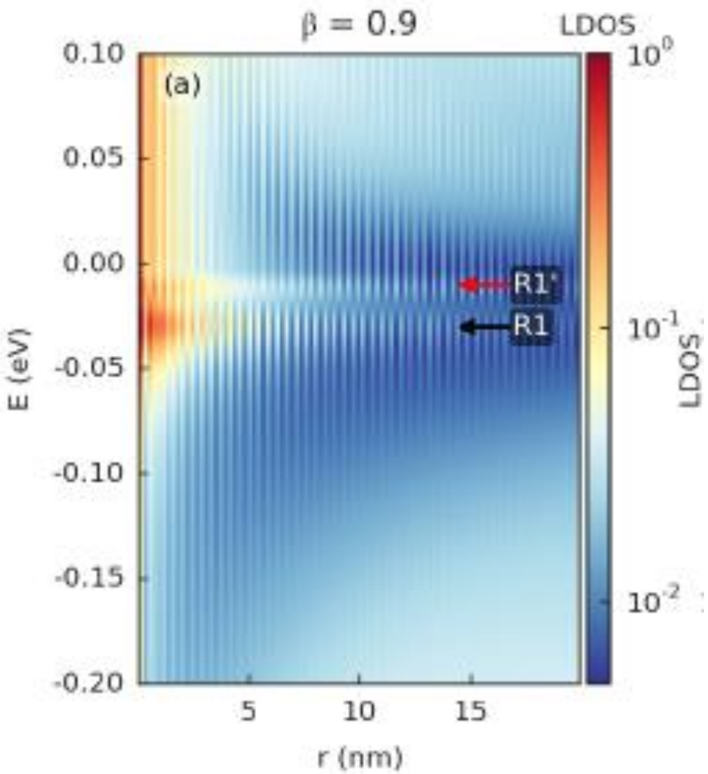
low intensity ring

**Quasi-classical orbit**

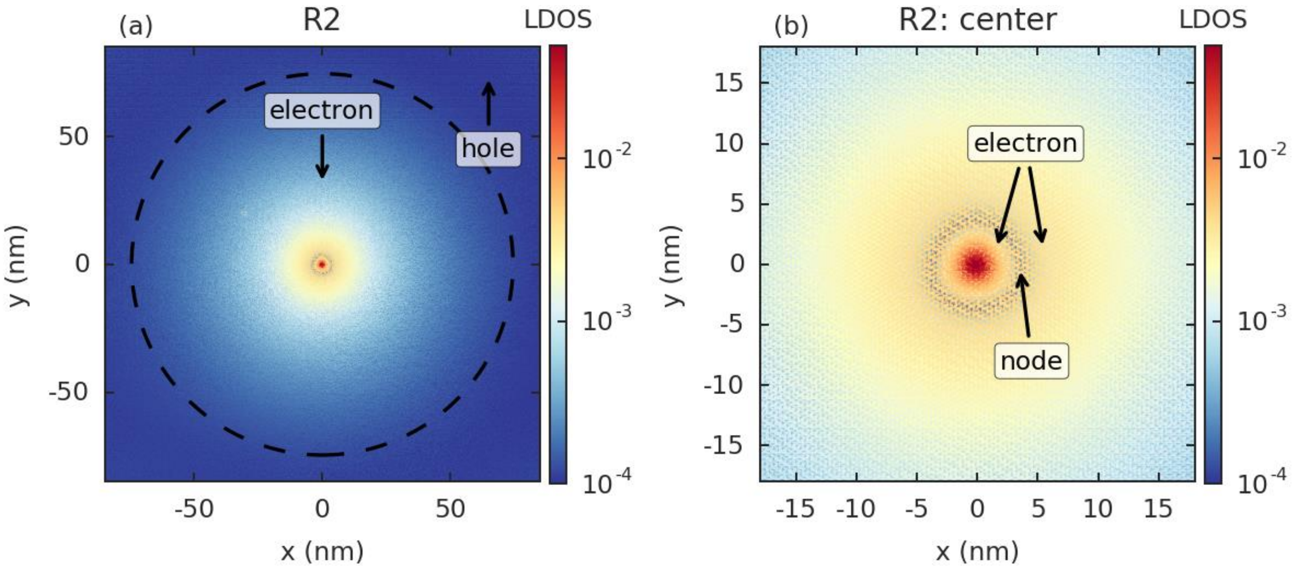


# R1 ←→ R1’

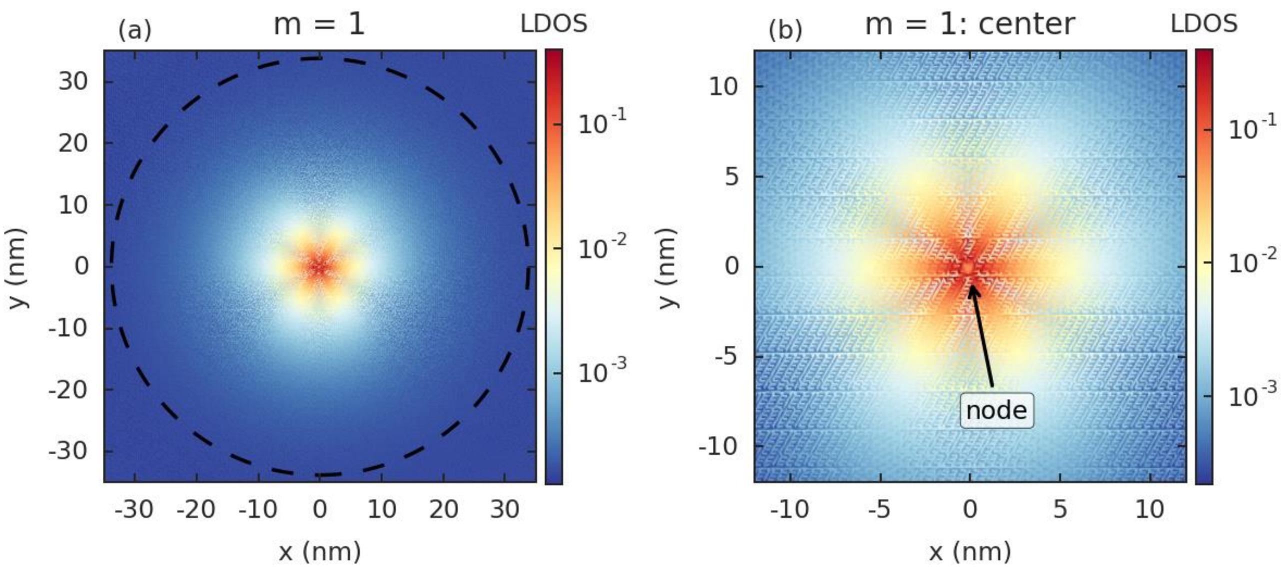



- R1 is more localized than R1’
- R1 has higher probability to be on sublattice B (= vacancy)
- R1’ is more localized on sublattice A



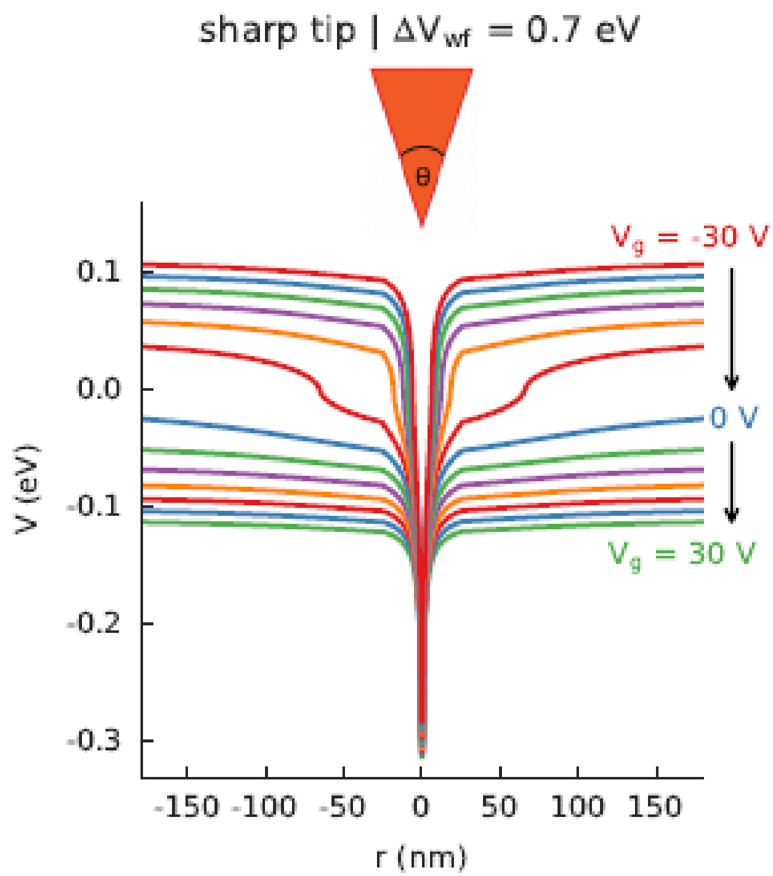

**like 2s-state**

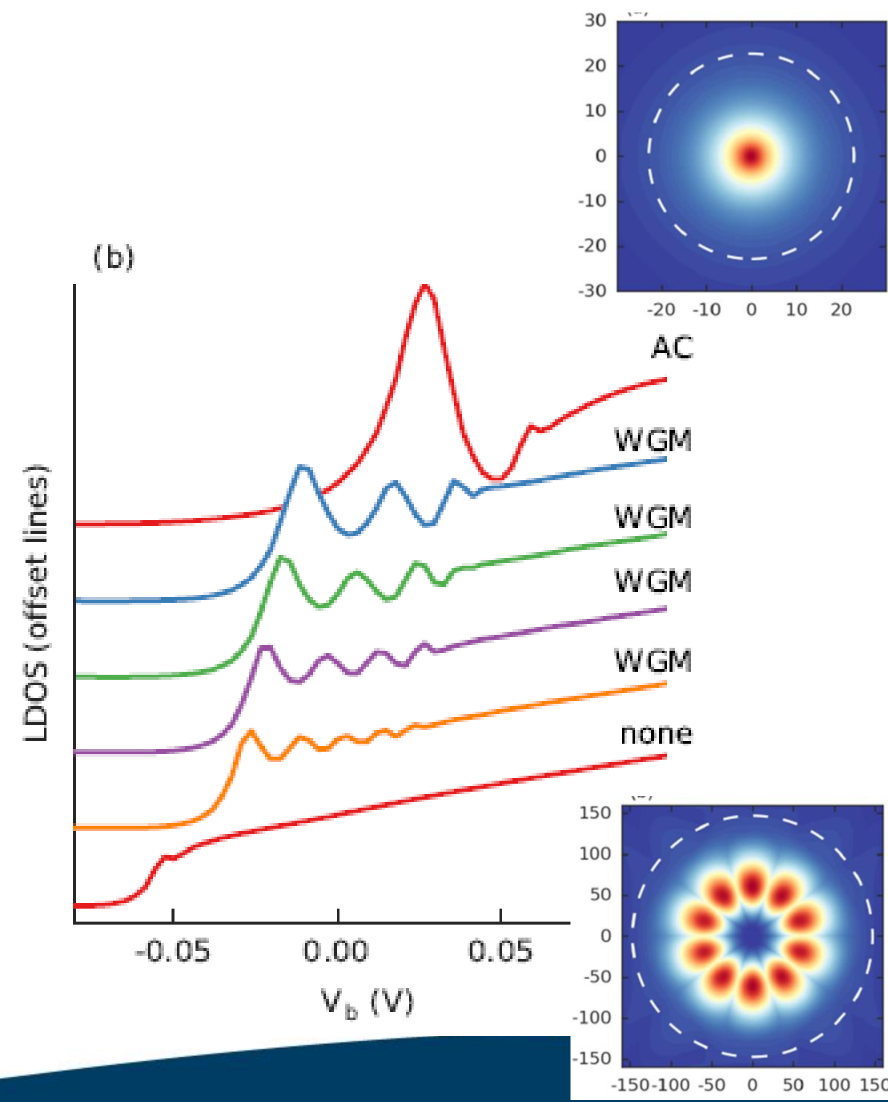



**like 2p-state**
(not affected by vacancy type)

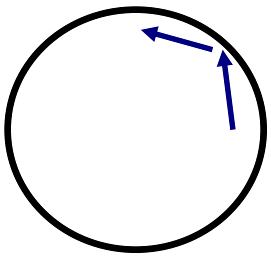
# STM-tip induced collapse state

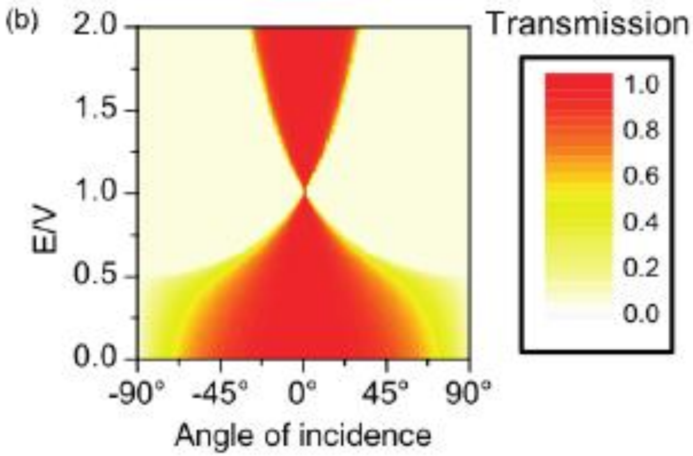
# Tuning from AC to WGM

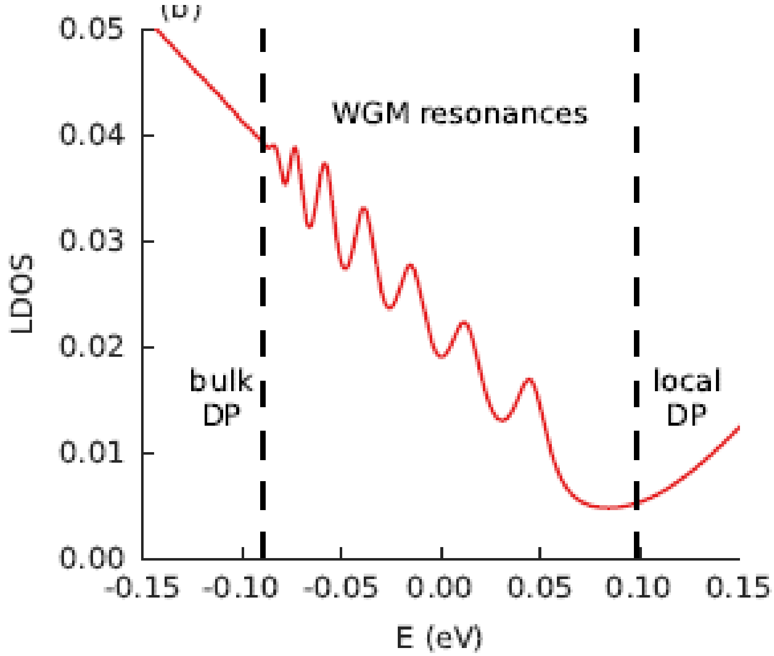


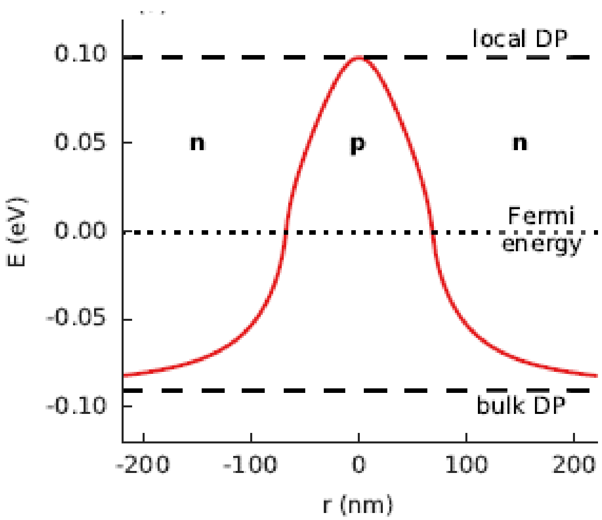



# Whispering gallery modes






- Radius pn-dot changes with $E_F$ (from nm to μm)
- m-large: electron hits pn-boundary under large angle θ → T(θ) << 1
- Length of closed loop = nλ → resonance
- quasi-confinement closer to the circumference of the junction → WGM



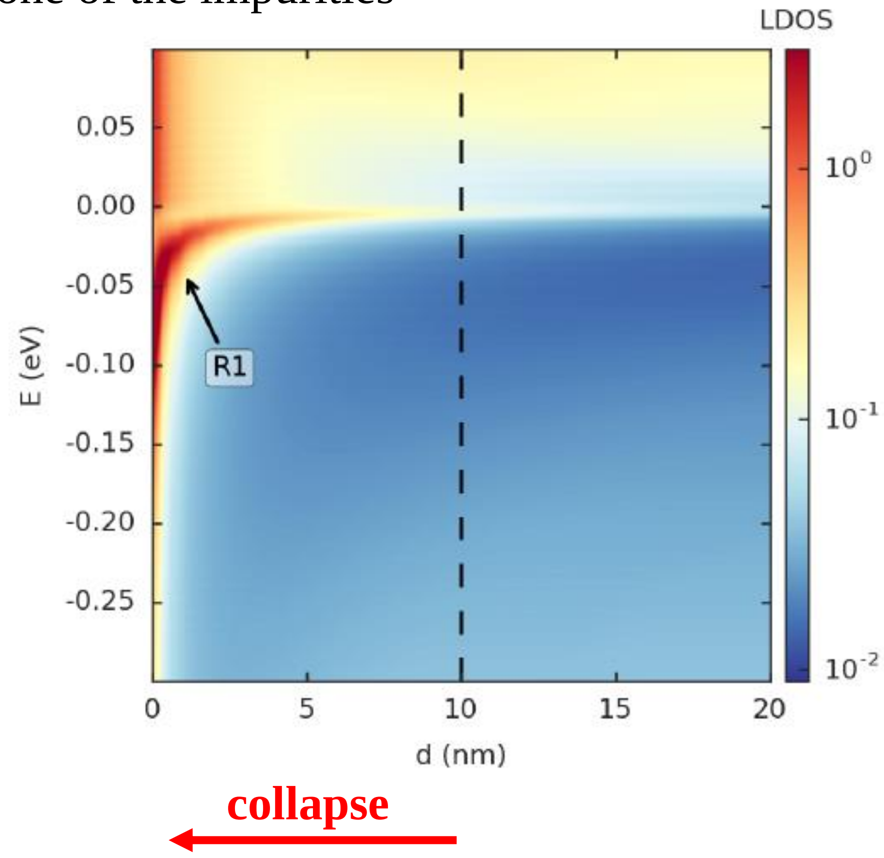
Y. Zhao et al, Science **348,** 672 (2015)

# Outlook

# Molecular collapse

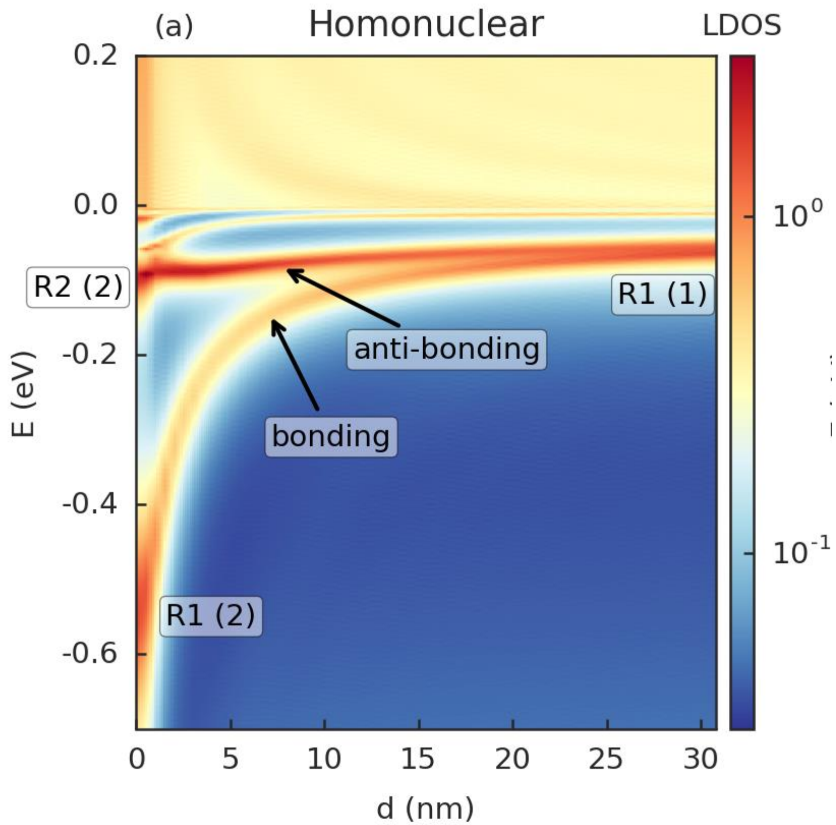
$\beta_1$=$\beta_2$=0.4 → two subcritical impurities

LDOS in center of one of the impurities



# Molecular collapse

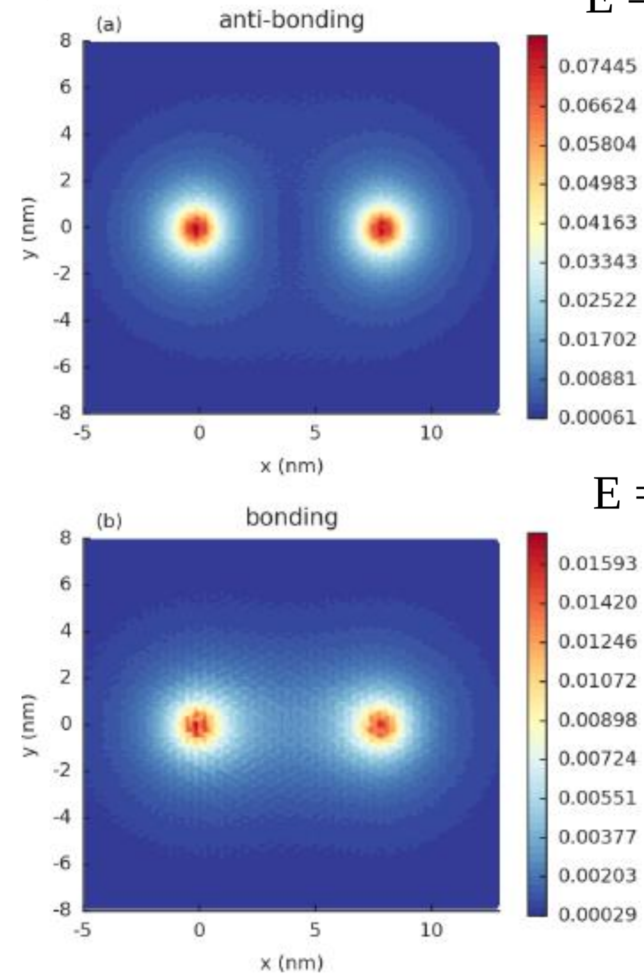
**2 impurities with $\beta_1=\beta_2=0.9$**




d = 8nm

E = -77meV

E = -135 meV



# Magnetic field dependence

→ *Has B any effect on the critical charge?*
→ *What is the signature of collapse (as function of B)?*

Prediction: in **3D** adding a magnetic field will *enhance* the effect of collapse → *reduce* the critical charge [V.N. Oraevskii, A.I. Rex, and V.B.Semikoz, Sov. Phys. JETP **45**, 428 (1977)].

Magnetic field confines electrons → bring it closer to the nucleus → $Z_c\downarrow$ when $B\uparrow$

In 2D conflicting predictions of the effect of B on $Z_c$ (from continuum theory)

All experiments up to now are in the subcritical regime

# The critical charge $Z_c$ → 0 for B # 0.

[O. V. Gamayun, E. V. Gorbar, and V. P. Gusynin, Phys. Rev. B **83**, 235104 (2011)].

# $Z_c$ independent of B

Y. Zhang, Y. Barlas, and K. Yang, Phys. Rev. B **85**, 165423 (2012)
T. Maier and H. Siedentop, J. Math. Phys. **53**, 095207 (2012)
S.C. Kim and S.-R. Eric Yang, Ann. Phys. (N.Y.) **347**, 21 (2014)

Problem: B-field → singular perturbation → no analytic solutions are known

Solutions are qualitatively different from B=0 case:

1) All energies are discrete
2) No complex energy solutions
3) Wavefunctions are normalizable

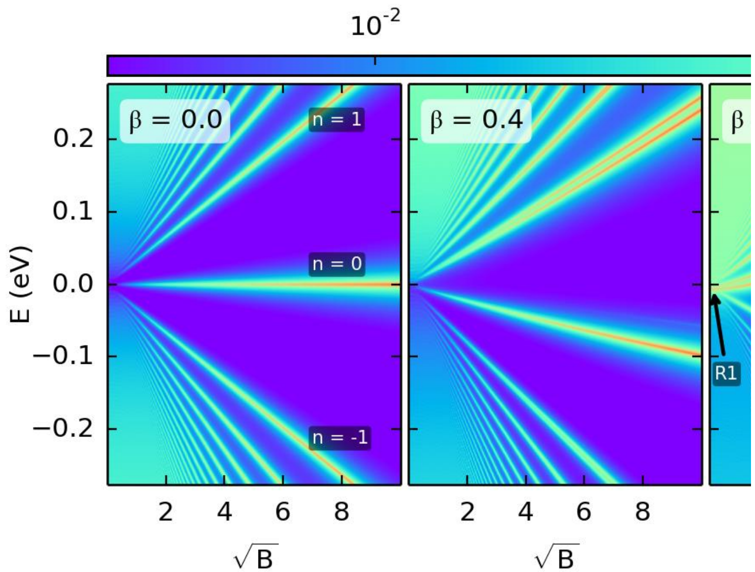
$$\boxed{t_{ij} \rightarrow t_{ij} e^{i2\pi\Phi_{ij}}}$$

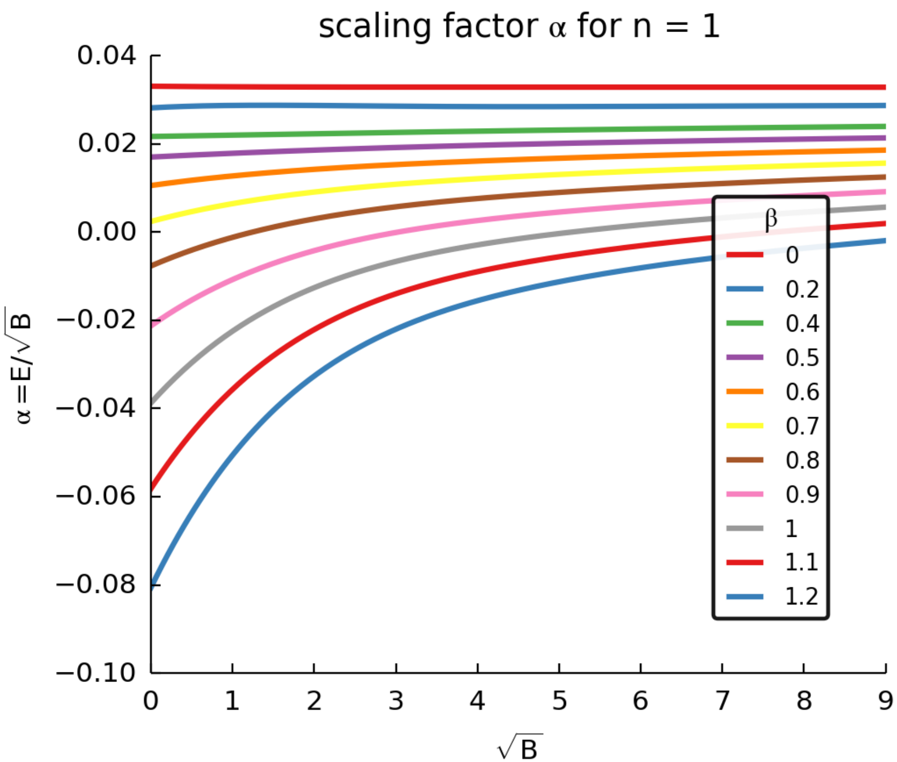
Renormalization length is introduced and single valley → here: lattice model

# Scaling of Landau levels ?

k ➔ $1/l_B \sim \sqrt{B}$

$$E(B) = v_F\sqrt{2n\hbar}\sqrt{B} = \nu\sqrt{B},$$



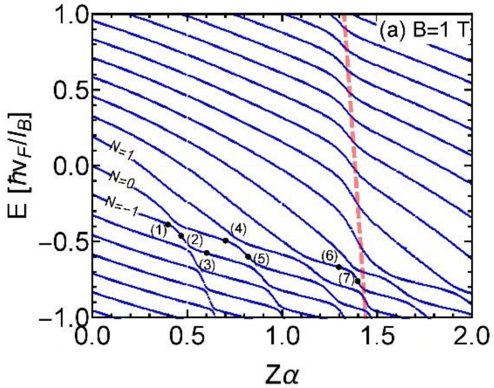



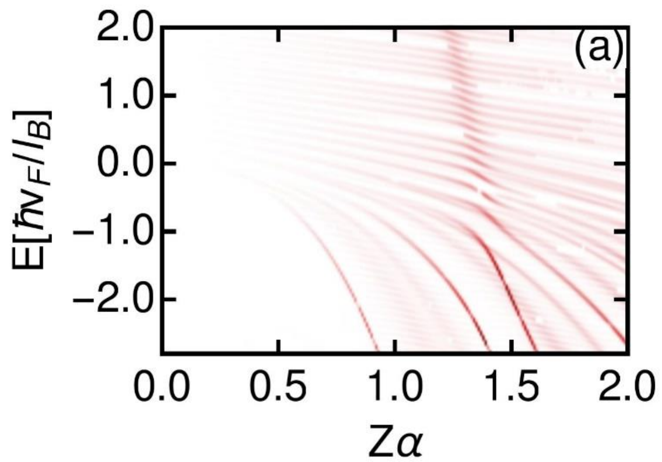

All levels show $B^{1/2}$ scaling

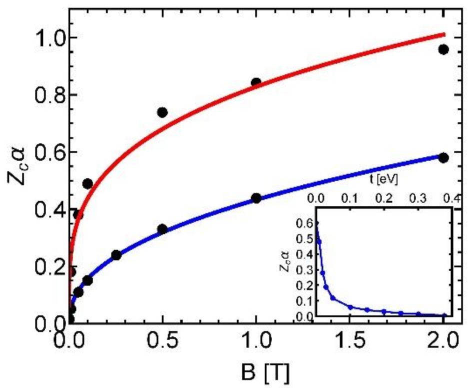
**No scaling**
**of atomic collapsed atates**

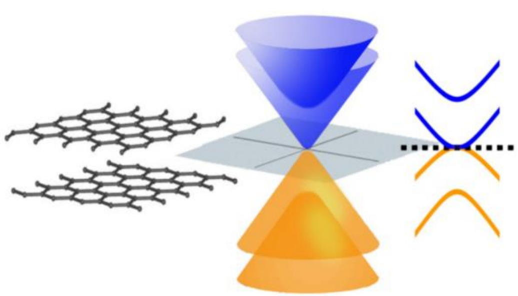
**Waiting for experimental confirmation**

# Bilayer graphene



## Bilayer graphene

| | 2-band | 4-band | mono |
|---|---|---|---|
| Dispersion relation | E ~ $k^2$ | E ~ $k^2$ , k-small<br>E ~ k , k-large | E ~ k |
| Complex energies | X | X | X |
| Fall-to-center | - | X | X |
| $Z_c\alpha$ | 0 | 0 | 0.5 |

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# Conclusions

⇒ Graphene as a lab. for investigating ***relativistic quantum mechanical*** effects (which have not been observed with 'real' particles):

- Klein paradox
- Atomic collapse

⇒ Kepler problem: $H = H_{kin} - Ze^2/\varepsilon r$

- Classical: $H_{kin} = \mathbf{p}^2/2m$ → unstable orbits
- Quantum mechanical: $H_{kin} = -\hbar^2\Delta/2m$ → Rydberg spectrum
- Dirac-Weyl (ultra-relativistic): $H_{kin} = v_F \boldsymbol{\sigma}\cdot\mathbf{p}$ → atomic collapse

⇒ Graphene: effects due to A/B sublattices are observable (R1/R1' collapsed states)

Nature Nanotechnology **12**, 1045 (2017)

Nature Physics **12**, 545 (2016)

⇒ STM-tip: transition from quantum (AC-states) to classical (WGM)

⇒ Outlook: - molecular collapsed states: bonding / anti-bonding states
- magnetic fields: non-scaling of Landau levels when in collapsed state
  critical charge independent of magnetic field

→ Bilayer: $Z_c=0$, AC-state due to second subband

2D Materials **5**, 015017 (2018)

*experiment*

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**THE END**