

Disorder in bilayer and double layer graphene

David Abergel

with E. Rossi, R. Sensarma, M. Rodriguez-Vega, and S. Das Sarma.

Nordita

KTH Royal Institute of Technology and Stockholm University

September 19th, 2014



NORDITA



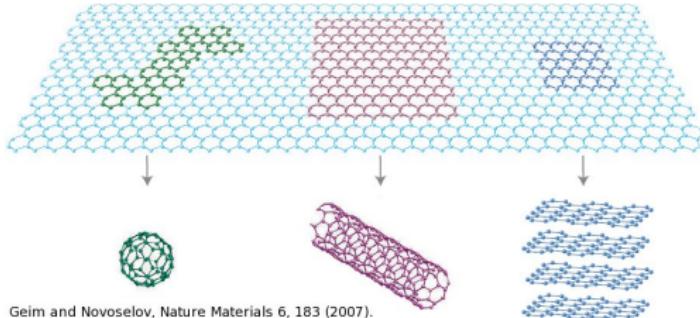
European Research Council

Established by the European Commission

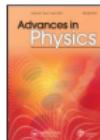
- 1 Graphene and disorder
- 2 Thomas-Fermi theory
- 3 Gapped bilayer graphene
- 4 Excitonic condensation in double layer graphene

Graphene and Disorder

Graphene



- Graphene is monolayer of carbon in honeycomb lattice.
- sp^2 bonding with p_z valence electrons.
- Massless Dirac-like low energy spectrum.
- Amazing physical characteristics.
- Many possible applications.



Many details in long review, Abergel et al., Adv. Phys. 59, 261 (2010).

Graphene is an exciting material to study because

- Rich fundamental physics.
- Vigorous experimental effort.
- Huge interest from many communities.
- ~ 5000 papers per year,
 ~ 200 on fundamental issues.

Possible applications in

- Touch screens,
- photonics and plasmonics,
- RF transistors,
- lasing,
- NEMS,
- biosensing,
- flexible electronics.

Graphene – Band structure

- Wave functions are two component spinors:

$$\psi = \begin{pmatrix} \psi_A \\ \psi_B \end{pmatrix}.$$

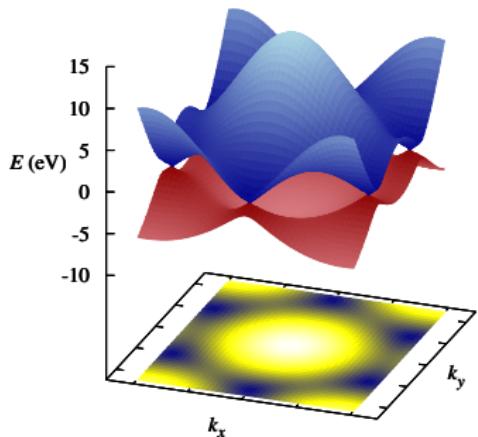
- Tight binding Hamiltonian:

$$H = \begin{pmatrix} H_{AA} & H_{BA} \\ H_{AB} & H_{BB} \end{pmatrix} = t_{AB} \begin{pmatrix} 0 & f(\mathbf{k}) \\ f^*(\mathbf{k}) & 0 \end{pmatrix},$$

$$f(\mathbf{k}) = e^{ik_y a/\sqrt{3}} + 2 \cos \frac{k_x a}{2} e^{ik_y a/2\sqrt{3}}.$$

- Schrödinger equation:

$$H\psi = E\psi.$$



Graphene – Band structure

- Wave functions are two component spinors:

$$\psi = \begin{pmatrix} \psi_A \\ \psi_B \end{pmatrix}.$$

- Tight binding Hamiltonian:

$$H = \begin{pmatrix} H_{AA} & H_{BA} \\ H_{AB} & H_{BB} \end{pmatrix} = t_{AB} \begin{pmatrix} 0 & f(\mathbf{k}) \\ f^*(\mathbf{k}) & 0 \end{pmatrix},$$

$$f(\mathbf{k}) = e^{ik_y a/\sqrt{3}} + 2 \cos \frac{k_x a}{2} e^{ik_y a/2\sqrt{3}}.$$

- Schrödinger equation:

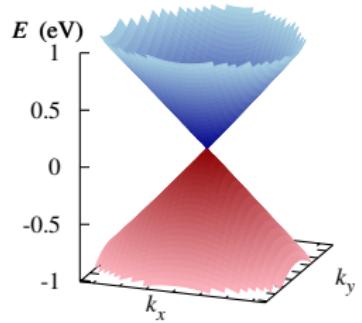
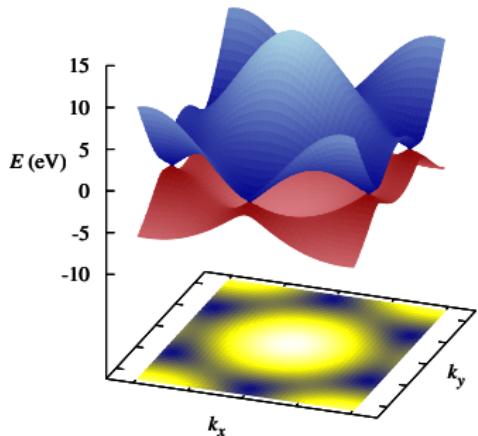
$$H\psi = E\psi.$$

- Expand near K points:

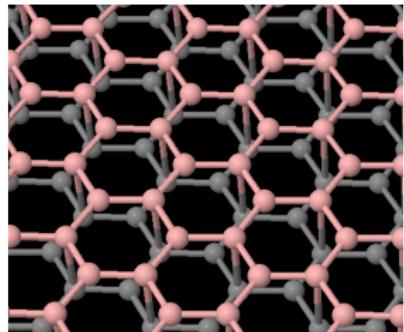
$$H_K = \begin{pmatrix} 0 & \hbar v_F(k_x - ik_y) \\ \hbar v_F(k_x + ik_y) & 0 \end{pmatrix}.$$

- Which yields the massless Dirac-like spectrum:

$$E_{\pm} = \pm \hbar v_F k.$$



Bilayer graphene

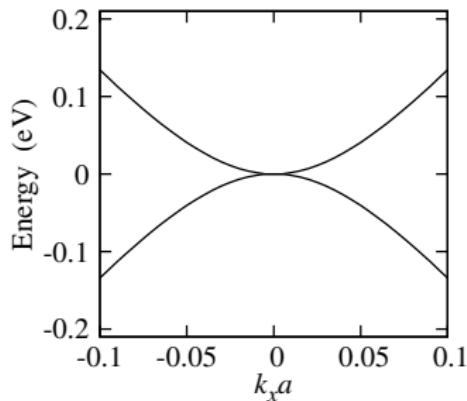


- Inter-layer bonds on alternate lattice sites.
- *AB* (Bernal) stacking preferable.
- Now four atoms per unit cell \Rightarrow four component wave function

$$\psi = \begin{pmatrix} \psi_{Au} \\ \psi_{Bl} \\ \psi_{Al} \\ \psi_{Bu} \end{pmatrix}.$$

$$H_K = \begin{pmatrix} 0 & 0 & 0 & v_F\pi^\dagger \\ 0 & v_F\pi^\dagger & v_F\pi & 0 \\ 0 & 0 & v_F\pi & \gamma_1 \\ v_F\pi & 0 & \gamma_1 & 0 \end{pmatrix}$$

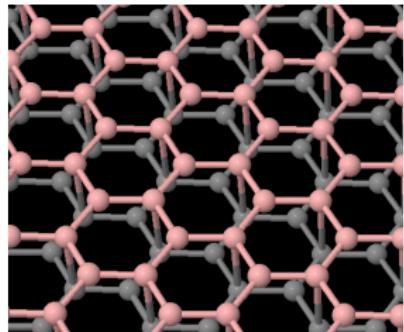
where $\pi = \hbar(k_x + ik_y)$.



See Abergel, Solid State Comm. **152**, 1383 (2012).

Effective mass is $m^* = \frac{\gamma_1}{2v_F^2}$.

Bilayer graphene

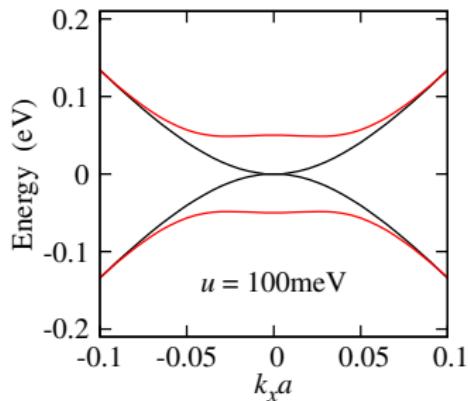


- Inter-layer bonds on alternate lattice sites.
- *AB* (Bernal) stacking preferable.
- Now four atoms per unit cell \Rightarrow four component wave function

$$\psi = \begin{pmatrix} \psi_{Au} \\ \psi_{Bl} \\ \psi_{Al} \\ \psi_{Bu} \end{pmatrix}.$$

$$H_K = \begin{pmatrix} \frac{u}{2} & 0 & 0 & v_F\pi^\dagger \\ 0 & -\frac{u}{2} & v_F\pi & 0 \\ 0 & v_F\pi^\dagger & -\frac{u}{2} & \gamma_1 \\ v_F\pi & 0 & \gamma_1 & \frac{u}{2} \end{pmatrix}$$

where $\pi = \hbar(k_x + ik_y)$.

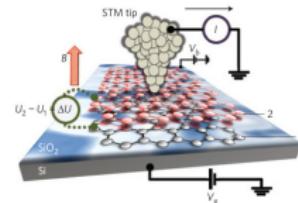


See Abergel, Solid State Comm. **152**, 1383 (2012).

Effective mass is $m^* = \frac{\gamma_1}{2v_F^2}$.

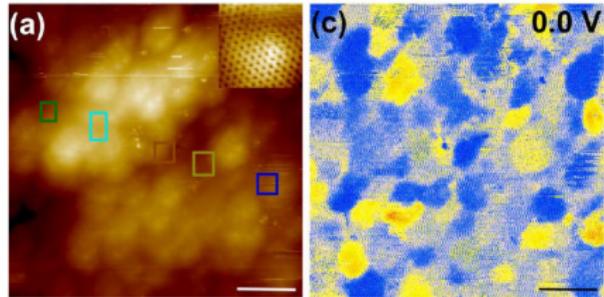


- STM can reveal atomic-scale structure of crystal.
- Also resolve the Dirac point,
- Which can be used to extract the local charge density.



Picture credit: Rutter et al., Nat. Phys. 7.

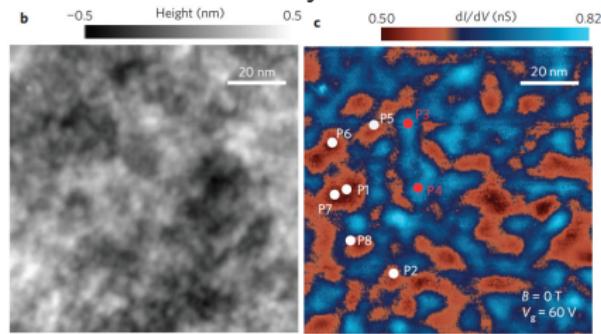
Monolayer:



Deshpande et al., Phys. Rev. B 79, 205411 (2009).

Scale bar is 8nm.

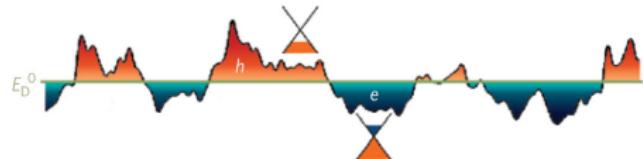
Bilayer:



Rutter et al., Nat. Phys. 7, 649 (2011).

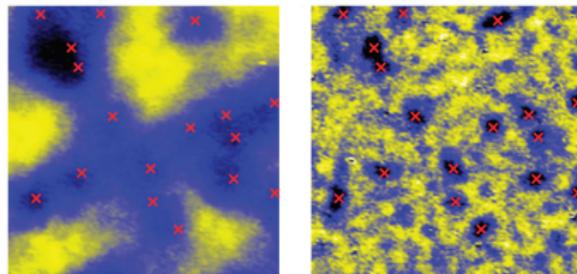
Scale bar is 20nm.

Scalar potential acts as a local shift in the chemical potential:



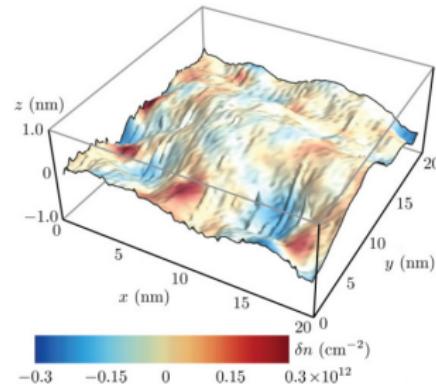
Picture credit: Beidenkopf *et al.*, Nat. Phys. 7.

Charged impurities:



Zhang *et al.*, Nat. Phys. 5, 722 (2009).

Ripples, corrugations, and strain:



Gibertini *et al.* Phys. Rev. B 85, 201405(R) (2012).

Thomas-Fermi theory

- Broken translational symmetry makes it impossible to analytically calculate exact density distribution for random disorder.
- We employ a numerical method: Thomas-Fermi theory.
- Functional method (à la DFT).
- The kinetic energy operator is also replaced by a functional of the density.
- This restricts the applicability to the regime where $|\nabla n/n| < k_F$, which is satisfied for double layer graphene.

- Energy functional includes contributions from disorder potential, and electron-electron interactions:

$$\begin{aligned} E[n] = E_K[n(\mathbf{r})] + \frac{e^2}{2\kappa} \int d\mathbf{r}' \int d\mathbf{r} \frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \\ + \frac{e^2}{\kappa} \int d\mathbf{r} V_D(\mathbf{r})n(\mathbf{r}) - \mu \int d\mathbf{r} n(\mathbf{r}). \end{aligned}$$

- Includes density-density interactions, and **external disorder potential**.
- Ground state density landscape is found by numerically minimizing the energy functional with respect to the density distribution.
- Density distribution can be translated into a local chemical potential.

Gapped bilayer graphene

D.S.L. Abergel, E. Rossi, and S. Das Sarma, Phys. Rev. B **86**, 155447 (2012).

- Measurements of the electronic compressibility can reveal much information about the **ground state properties** of electrons in a material.
- Methods include capacitance measurements, SET microscopy, and tunneling spectroscopy (STM).
- Therefore it's important to have a full theoretical understanding of the compressibility.

- Measurements of the electronic compressibility can reveal much information about the **ground state properties** of electrons in a material.
- Methods include capacitance measurements, SET microscopy, and tunneling spectroscopy (STM).
- Therefore it's important to have a full theoretical understanding of the compressibility.
- Compressibility linked to $\frac{d\mu}{dn}$:

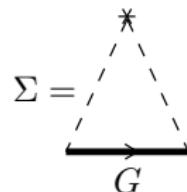
$$\frac{1}{K} = n^2 \frac{d\mu}{dn}$$

NB Thermodynamic density of states is $dn/d\mu$.

- In the clean, non-interacting case, $\frac{d\mu}{dn}$ can be calculated analytically.

- Standard methods of theoretical condensed matter physics include methods of calculating the effects of the scattering of electrons by impurities.
- In diagrammatic perturbation theory (DPT), one can compute the self-energy of the electron–impurity interaction.
- In the self-consistent Born approximation:

$$\Sigma_\alpha(\mathbf{k}, E) = n_i \sum_{\mathbf{k}'\alpha'} \frac{|V_D(\mathbf{k} - \mathbf{k}')|^2 F_{\alpha,\alpha'}(\mathbf{k}, \mathbf{k}')}{E - E_{\mathbf{k}'\alpha'} - \Sigma_{\alpha'}(\mathbf{k}', E)}.$$

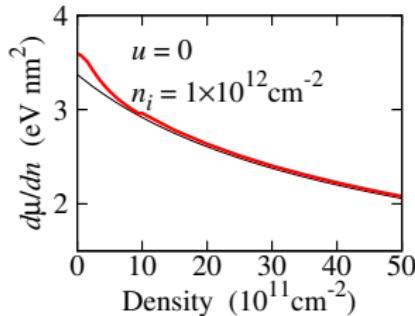


- Electron–impurity interaction is screened Coulomb potential:

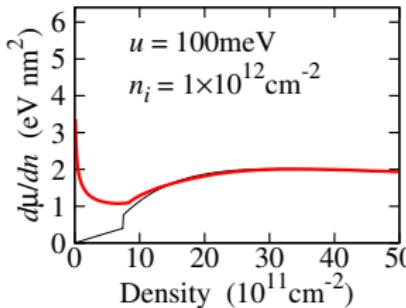
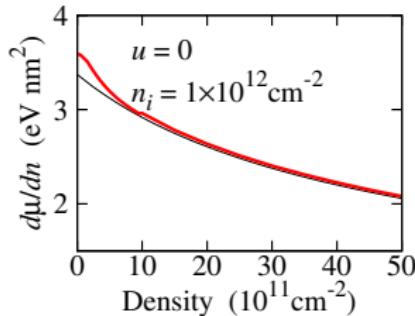
$$V_D(\mathbf{q}) = \frac{2\pi e^2}{\kappa(q + q_s)}, \quad q_s = \frac{2\pi e^2}{\kappa} \rho_0(\mu).$$

- The self-energy may be included in the electron Green's function and the DOS calculated from it:

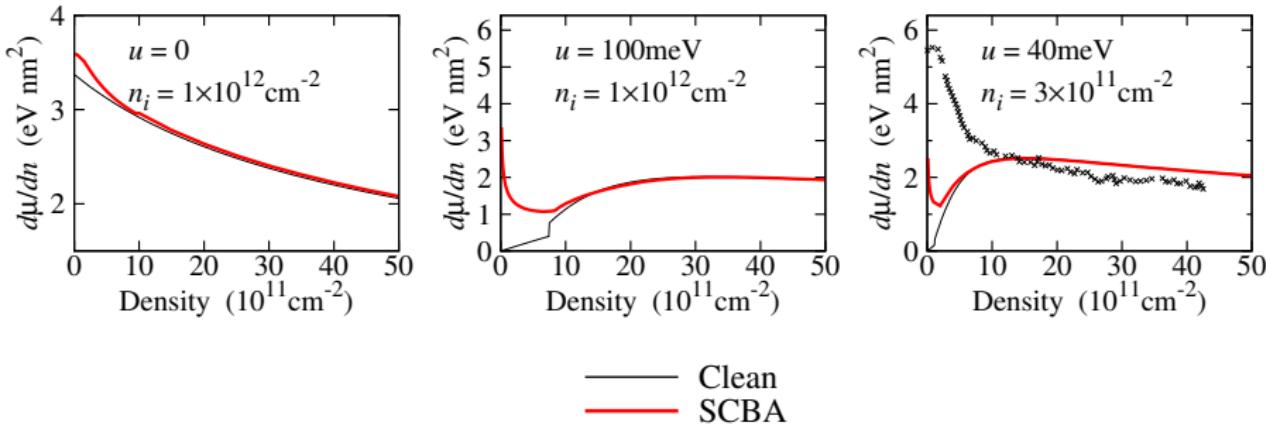
$$G_\alpha(\mathbf{k}, E) = \frac{1}{E - E_{\mathbf{k}\alpha} - \Sigma_\alpha(\mathbf{k}, E)}, \quad \frac{dn}{d\mu} = -\frac{g_s g_v}{\pi} \sum_\alpha \int \frac{d^2\mathbf{k}}{4\pi^2} G_\alpha(\mathbf{k}, E).$$



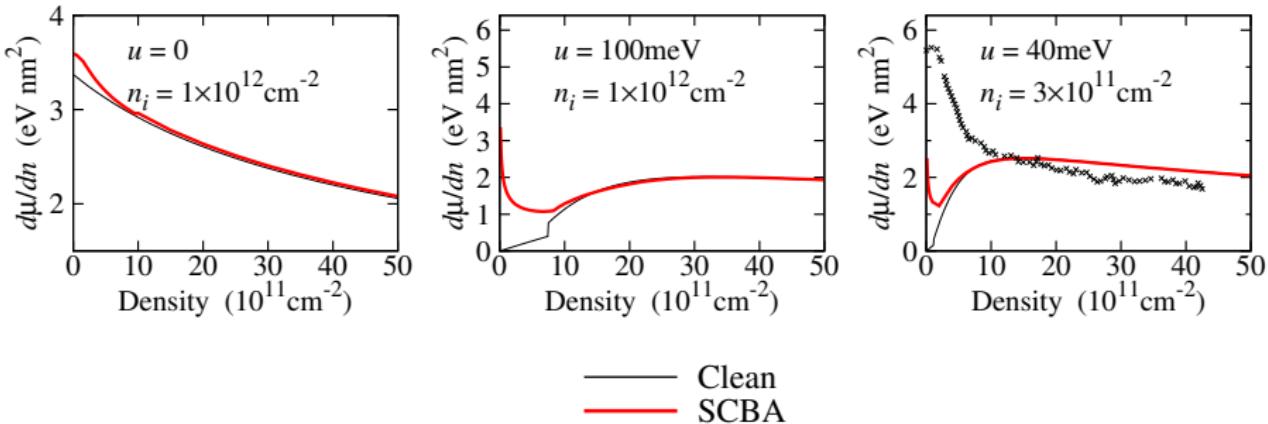
— Clean
— SCBA



— Clean
— SCBA



- Experimental data from Henriksen *et al.* Phys. Rev. B **82**, 041412 (2010).
- Qualitatively different behavior between DPT and experiment at low density.
- There are **two** places in the DPT procedure where **homogeneity** are imposed:
 - ▶ The average over disorder configurations in the derivation of $\Sigma_\alpha(\mathbf{k}, E)$,
 - ▶ the use of a static screening wave vector q_s .

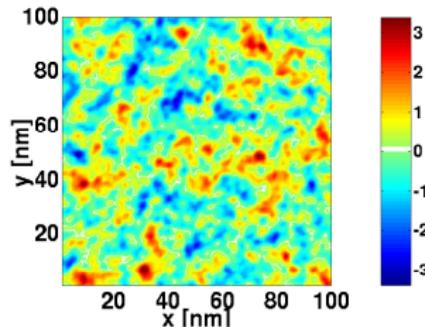


- Experimental data from Henriksen *et al.* Phys. Rev. B **82**, 041412 (2010).
- Qualitatively different behavior between DPT and experiment at low density.
- There are **two** places in the DPT procedure where **homogeneity** are imposed:
 - ▶ The average over disorder configurations in the derivation of $\Sigma_\alpha(\mathbf{k}, E)$,
 - ▶ the use of a static screening wave vector q_s .

How can we go beyond DPT?



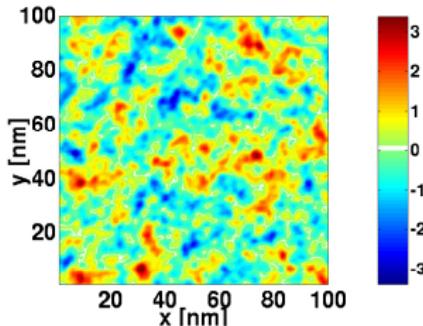
$u = 0, \langle n \rangle = 0.$



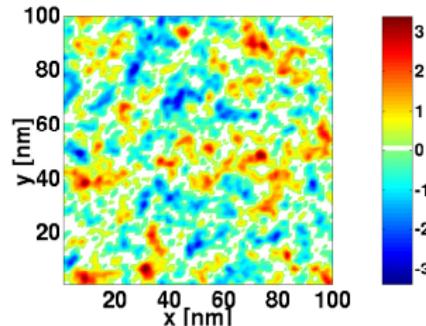


NORDITA TFT: Local n – effect of gap and $\langle n \rangle$

$u = 0, \langle n \rangle = 0.$



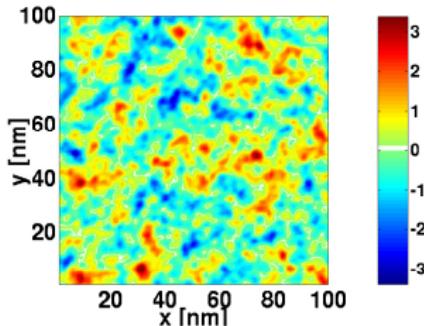
$u = 40\text{meV}, \langle n \rangle = 0.$



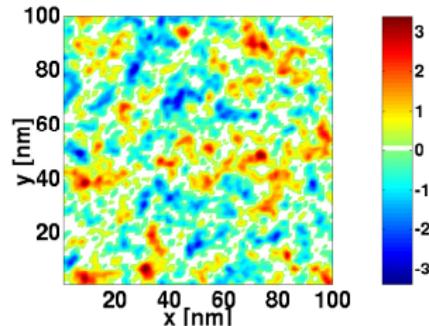


NORDITA TFT: Local n – effect of gap and $\langle n \rangle$

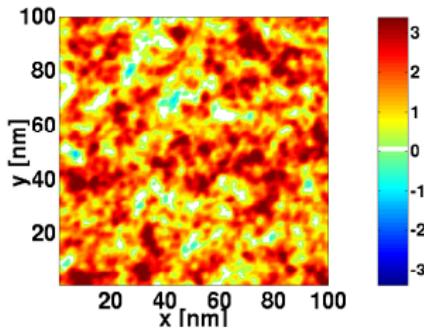
$$u = 0, \langle n \rangle = 0.$$



$$u = 40\text{meV}, \langle n \rangle = 0.$$



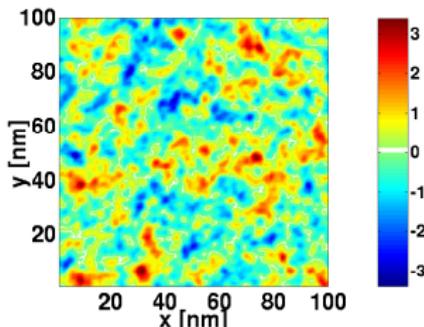
$$u = 40\text{meV}, \mu = 50\text{meV}, \\ \langle n \rangle \approx 1.2 \times 10^{12} \text{cm}^{-2}$$



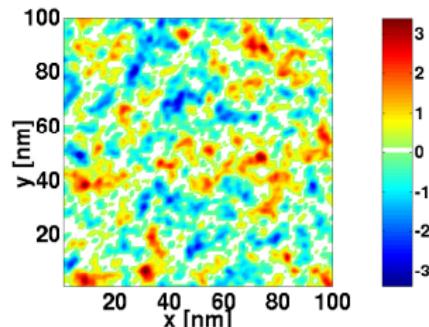


NORDITA TFT: Local n – effect of gap and $\langle n \rangle$

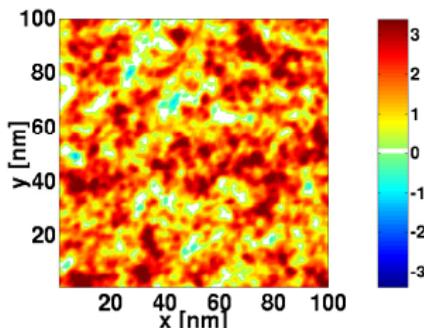
$$u = 0, \langle n \rangle = 0.$$



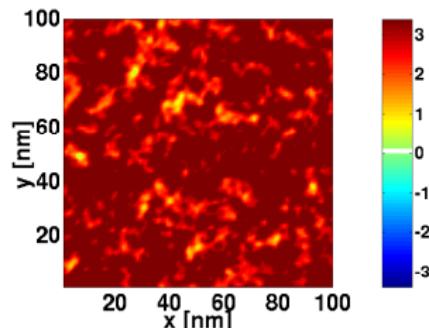
$$u = 40\text{meV}, \langle n \rangle = 0.$$



$$u = 40\text{meV}, \mu = 50\text{meV}, \\ \langle n \rangle \approx 1.2 \times 10^{12} \text{cm}^{-2}$$



$$u = 40\text{meV}, \mu = 100\text{meV}, \\ \langle n \rangle \approx 2.4 \times 10^{12} \text{cm}^{-2}$$



To compute the averaged $\frac{d\mu}{dn}$:

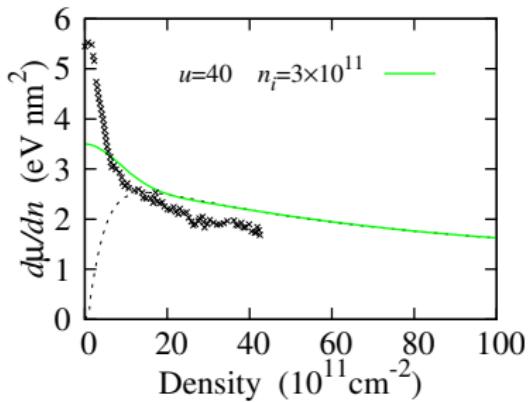
- μ is parameter of the theory.
- $P(n)$ is distribution function for local density.
[Actual functional dependence is $P(n, n_i, \mu, u)$.]
- Global density $\langle n \rangle = \int dn' n' P(n')$.
- Averaged compressibility then

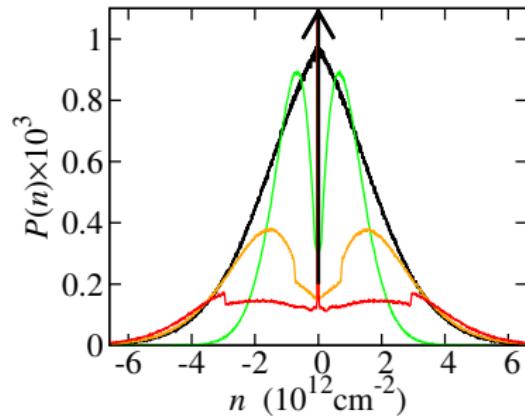
$$\left\langle \frac{d\mu}{dn} \right\rangle = \frac{d\mu}{d\langle n \rangle}.$$

To compute the averaged $\frac{d\mu}{dn}$:

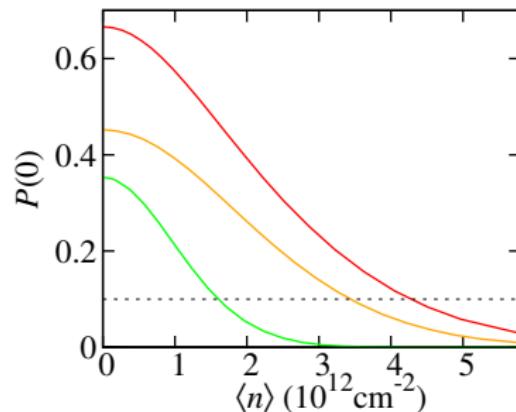
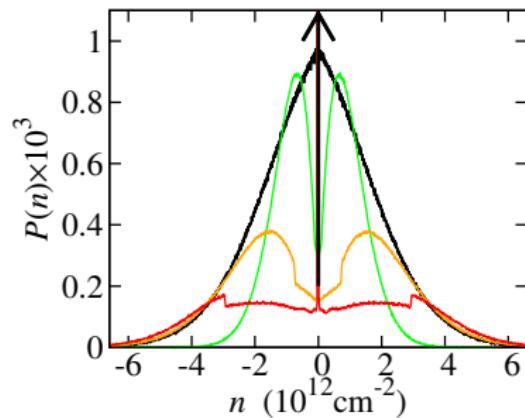
- μ is parameter of the theory.
- $P(n)$ is distribution function for local density.
[Actual functional dependence is $P(n, n_i, \mu, u)$.]
- Global density $\langle n \rangle = \int dn' n' P(n')$.
- Averaged compressibility then

$$\left\langle \frac{d\mu}{dn} \right\rangle = \frac{d\mu}{d\langle n \rangle}.$$

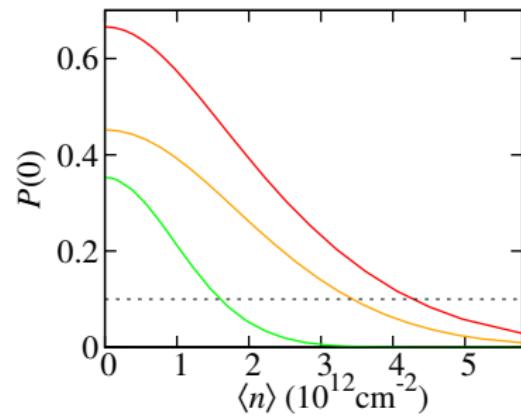
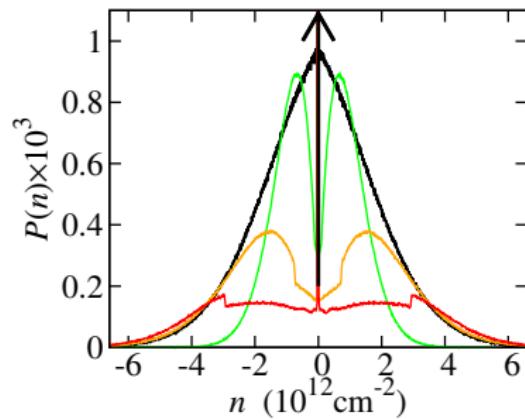




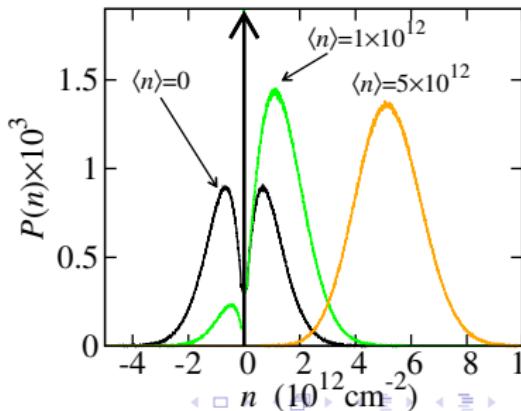
—	$u = 0$	$n_i = 1 \times 10^{12}$
—	$u = 40\text{meV}$	$n_i = 3 \times 10^{11}$
—	$u = 100\text{meV}$	$n_i = 1 \times 10^{12}$
—	$u = 200\text{meV}$	$n_i = 1 \times 10^{12}$

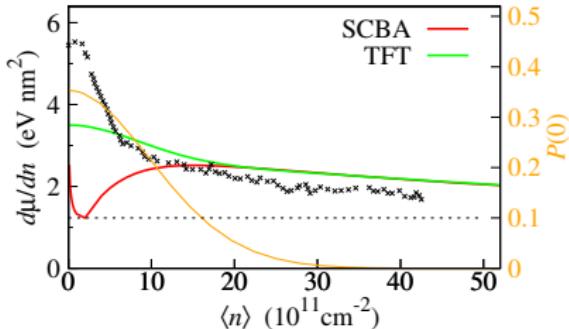


—	$u = 0$	$n_i = 1 \times 10^{12}$
—	$u = 40 \text{ meV}$	$n_i = 3 \times 10^{11}$
—	$u = 100 \text{ meV}$	$n_i = 1 \times 10^{12}$
—	$u = 200 \text{ meV}$	$n_i = 1 \times 10^{12}$



—	$u = 0$	$n_i = 1 \times 10^{12}$
—	$u = 40 \text{ meV}$	$n_i = 3 \times 10^{11}$
—	$u = 100 \text{ meV}$	$n_i = 1 \times 10^{12}$
—	$u = 200 \text{ meV}$	$n_i = 1 \times 10^{12}$

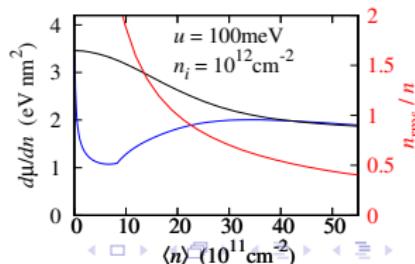
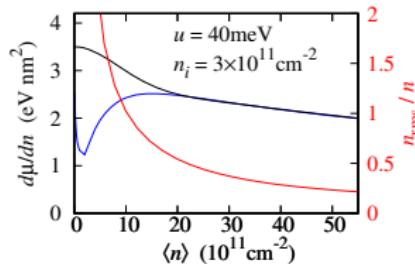
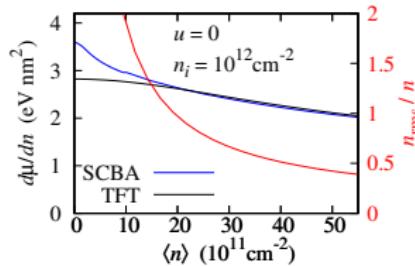




DPT deviate from experiment and TFT when $P(0) \sim 0.1$,

$P(0)$ is the measure of incompressible regions.

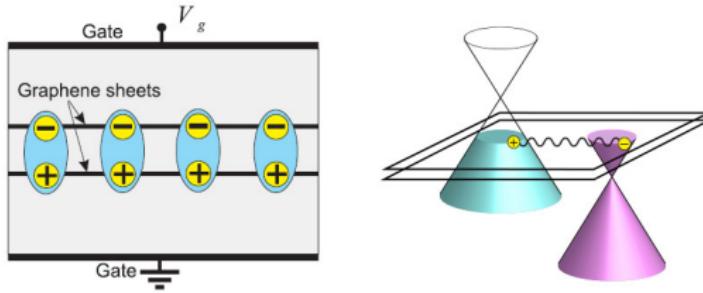
or when $n_{\text{rms}}/n \sim 1$.



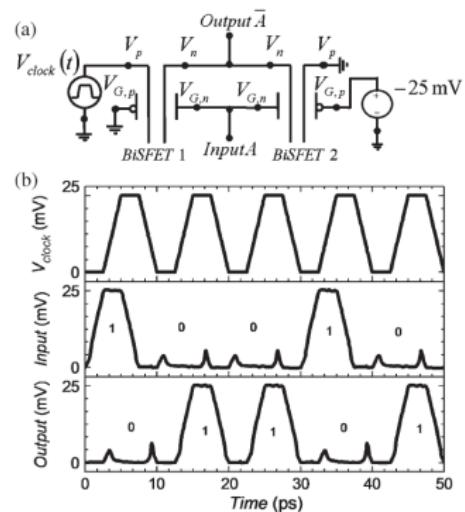
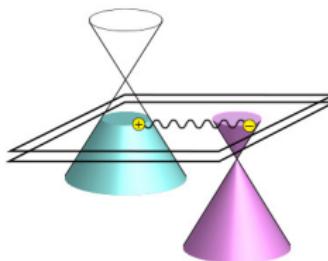
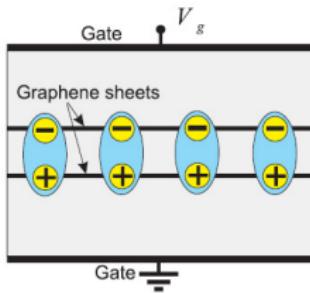
Excitonic condensation in double layer graphene

- D. S. L. Abergel, M. Rodriguez-Vega, E. Rossi, and S. Das Sarma, Phys. Rev. B **88**, 235402 (2013).
D. S. L. Abergel, R. Sensarma, and S. Das Sarma, Phys. Rev. B **86**, 155447(R) (2012).

- Double layer graphene predicted to support excitonic condensate.



- Double layer graphene predicted to support excitonic condensate.
- Proposal for dispersionless FET.



IEEE ELECTRON DEVICE LETTERS, VOL. 30, NO. 2, FEBRUARY 2009

Bilayer PseudoSpin Field-Effect Transistor (BiSFET): A Proposed New Logic Device

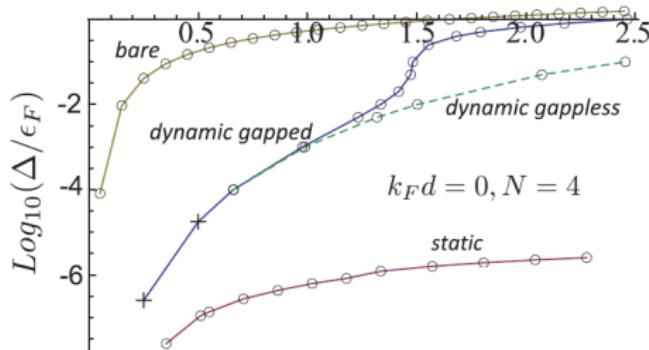
Sanjay K. Banerjee, *Fellow, IEEE*, Leonard F. Register, *Senior Member, IEEE*, Emanuel Tutuc, *Member, IEEE*, Dharmendar Reddy, and Allan H. MacDonald

- The condensate has yet to be observed despite several experimental attempts.
- Question is: Why?

- The condensate has yet to be observed despite several experimental attempts.
- Question is: Why?

Possibility 1: Excitonic gap is too small.

The form of the inter-layer screening used in the calculation of the gap is crucial:



Sodemann *et al.*, Phys. Rev. B **85**, 195136 (2012).

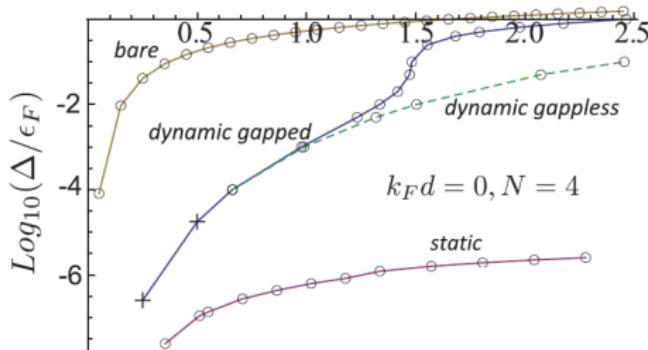
For SiO_2 or BN substrates, $\alpha = \frac{e^2}{\kappa \hbar v_F} \approx 0.5$.
 For vacuum (suspended graphene), $\alpha = 2.2$.

- Unscreened interaction \Rightarrow room temperature condensate!!!
- Static screening \Rightarrow vanishing gap.
- Dynamic screening \Rightarrow ???

- The condensate has yet to be observed despite several experimental attempts.
- Question is: Why?

Possibility 1: Excitonic gap is too small.

The form of the inter-layer screening used in the calculation of the gap is crucial:



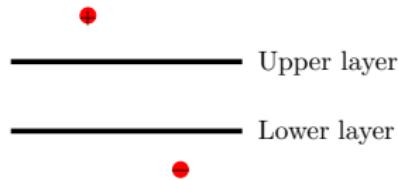
Sodemann *et al.*, Phys. Rev. B **85**, 195136 (2012).

For SiO₂ or BN substrates, $\alpha = \frac{e^2}{\kappa \hbar v_F} \approx 0.5$.
 For vacuum (suspended graphene), $\alpha = 2.2$.

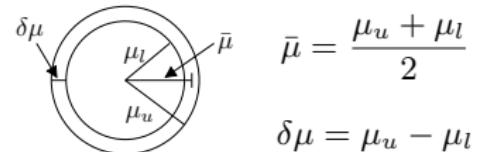
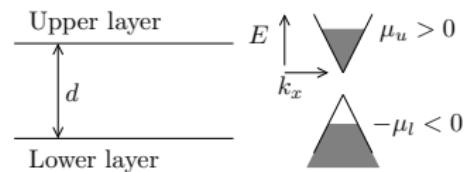
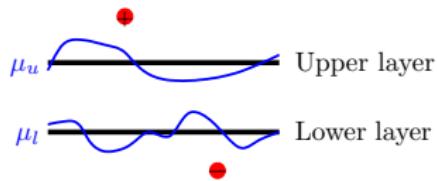
- Unscreened interaction \Rightarrow room temperature condensate!!!
- Static screening \Rightarrow vanishing gap.
- Dynamic screening \Rightarrow ???

Possibility 2: Disorder

- Main question: Does charge inhomogeneity affect the formation of the condensate?



- Main question: Does charge inhomogeneity affect the formation of the condensate?



- This is similar to magnetic disorder in superconductivity.

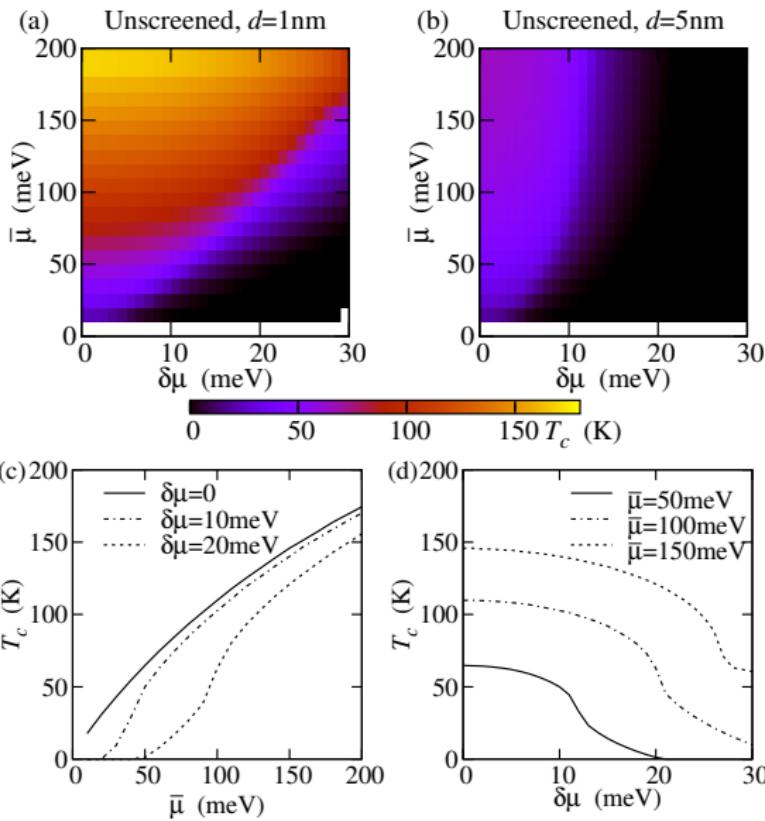
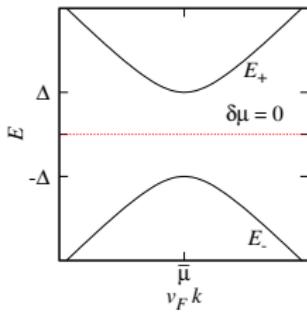
There are three stages to the calculation:

- ① Theory for homogeneous unbalanced system.
 - ▶ Temporarily ignore inhomogeneity, calculate effect of imperfectly nested Fermi surfaces.
- ② Analysis of realistic inhomogeneity.
 - ▶ Calculate statistics for $\delta\mu(\mathbf{r})$ in situations corresponding to contemporary experiments.
- ③ Combine these two results to assess impact of inhomogeneity on condensate formation.

Unscreened interaction:

$$V(q) = \frac{2\pi e^2}{\epsilon q}$$

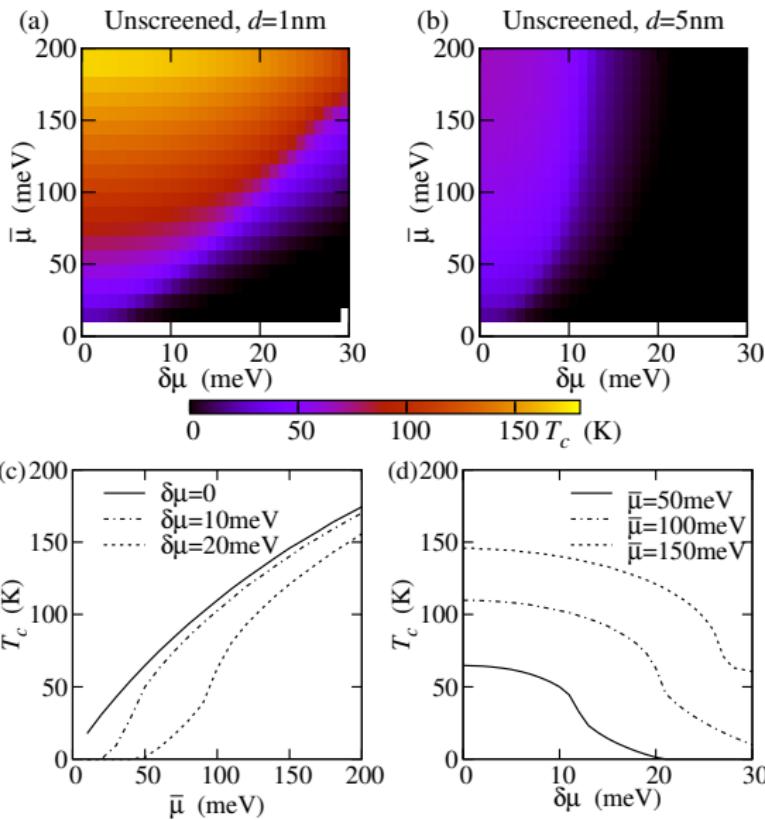
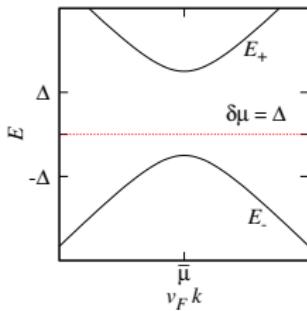
- $\Delta(\delta\mu)$ unchanged for $\delta\mu < 2\Delta(0)$.
- Equivalent to Clogston–Chandrasekhar limit.
- No evidence of FFLO state.



Unscreened interaction:

$$V(q) = \frac{2\pi e^2}{\epsilon q}$$

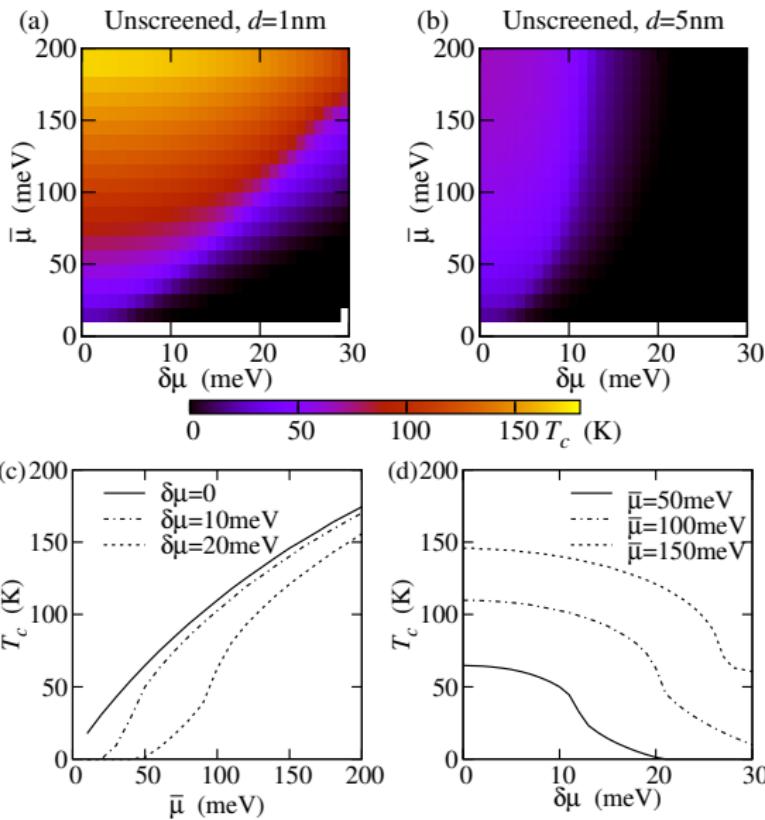
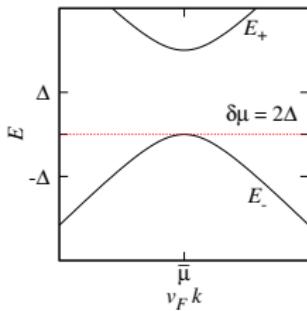
- $\Delta(\delta\mu)$ unchanged for $\delta\mu < 2\Delta(0)$.
- Equivalent to Clogston–Chandrasekhar limit.
- No evidence of FFLO state.



Unscreened interaction:

$$V(q) = \frac{2\pi e^2}{\epsilon q}$$

- $\Delta(\delta\mu)$ unchanged for $\delta\mu < 2\Delta(0)$.
- Equivalent to Clogston–Chandrasekhar limit.
- No evidence of FFLO state.



- Energy functional is

$$E[n_u, n_l] = E_u[n_u(\mathbf{r})] + E_l[n_l(\mathbf{r})] + \frac{e^2}{2\kappa} \iint d^2\mathbf{r} d^2\mathbf{r}' \frac{n_u(\mathbf{r})n_l(\mathbf{r}')}{\sqrt{|\mathbf{r} - \mathbf{r}'|^2 + d^2}}$$

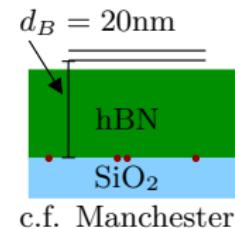
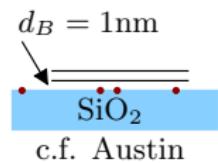
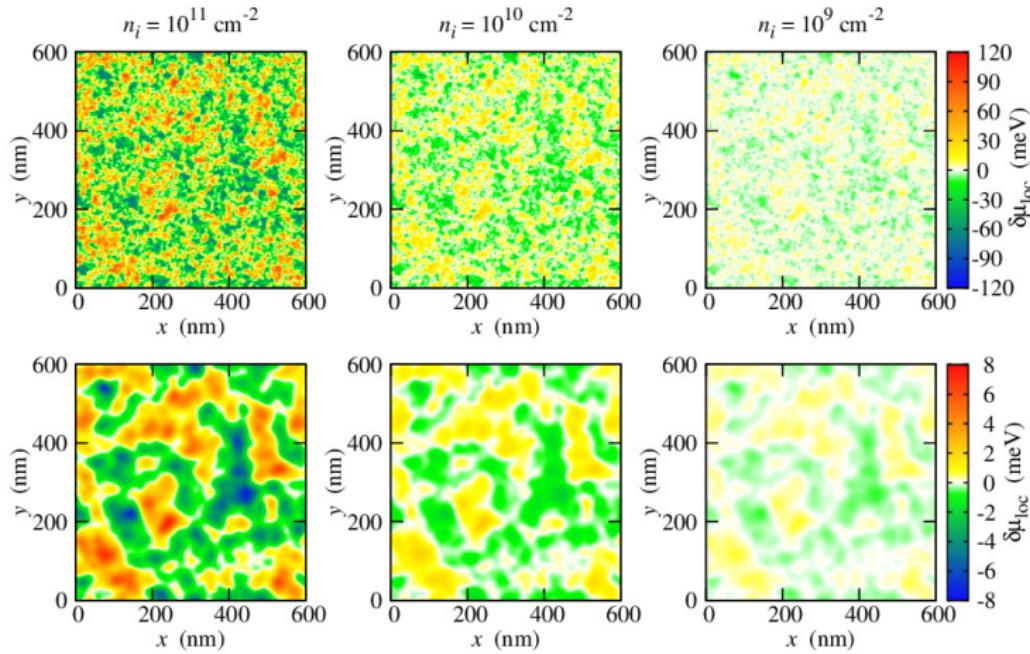
- Energy functional includes contributions from disorder potential, and electron-electron interactions:

$$\begin{aligned} E[n] = E_K[n(\mathbf{r})] &+ \frac{e^2}{2\kappa} \int d\mathbf{r}' \int d\mathbf{r} \frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \\ &+ \frac{e^2}{\kappa} \int d\mathbf{r} V_D(\mathbf{r})n(\mathbf{r}) - \mu \int d\mathbf{r} n(\mathbf{r}). \end{aligned}$$

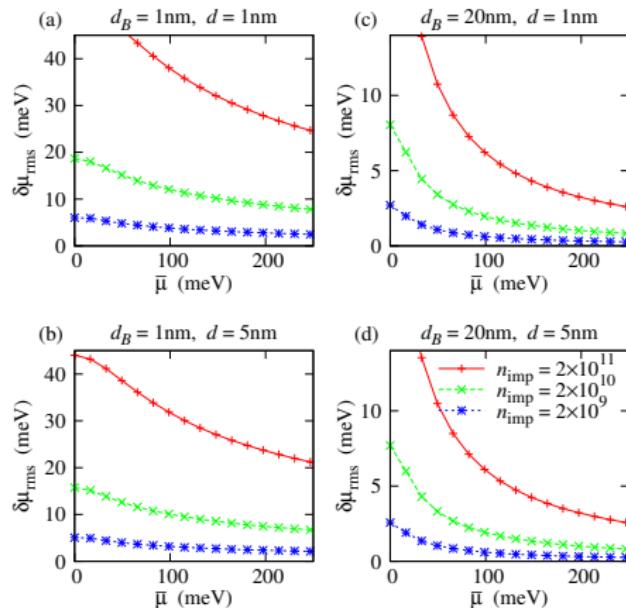
- Ground state density landscape is found by numerically minimizing the energy functional with respect to the density distribution.
- Density distribution gives local chemical potential for each layer, and hence the local $\delta\mu$.



- Using TFT, we calculate the spatial profile of $\delta\mu$ for a given manifestation of charged impurity disorder:



- We can perform this calculation for many (≈ 600) disorder realizations and collect **statistics** for the distribution of $\delta\mu$.
- This distribution characterized by it's root-mean-square (rms) value.



Predictions for Δ from BCS theory:

- Unscreened: $\Delta \sim 30\text{meV}$,
- Static screening: $\Delta \sim 0.01\text{meV}$,
- Dynamic screening: $\Delta \sim 1\text{meV}$.