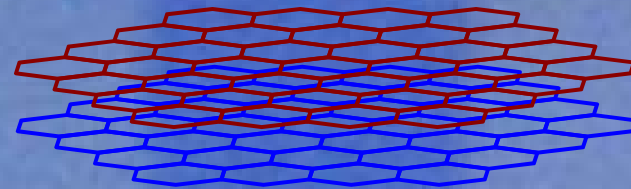


Diffusive Charge Transport in Graphene



Michael S. Fuhrer
Department of Physics and
Center for Nanophysics and Advanced Materials
University of Maryland

Outline

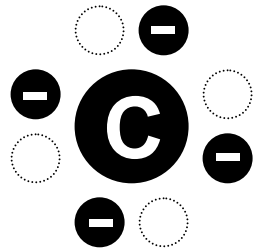
- I. Introduction to Graphene
 - “Massless” electrons
 - Pseudospin and Berry’s phase

- II. Fabrication and Characterization of Graphene on SiO₂
 - Micro-Raman spectroscopy
 - Cleaning graphene

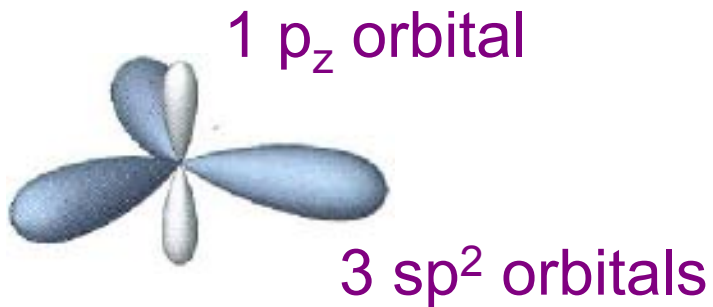
- III. Diffusive Transport in Graphene
 - Boltzmann Transport
 - Charged impurities
 - Charged impurities – minimum conductivity
 - Phonons
 - Dielectric Environment – Tuning Fine Structure Constant
 - Corrugations
 - Lattice defects

Carbon and Graphene

Carbon



4 valence electrons

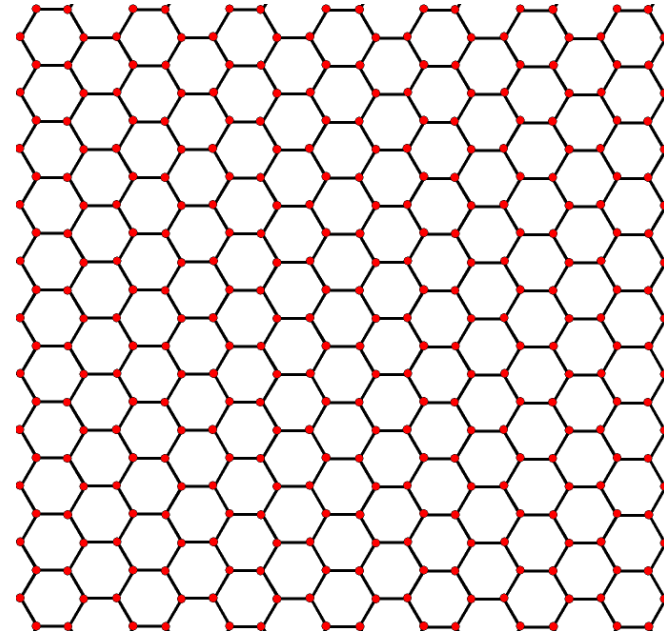


1 p_z orbital

3 sp² orbitals

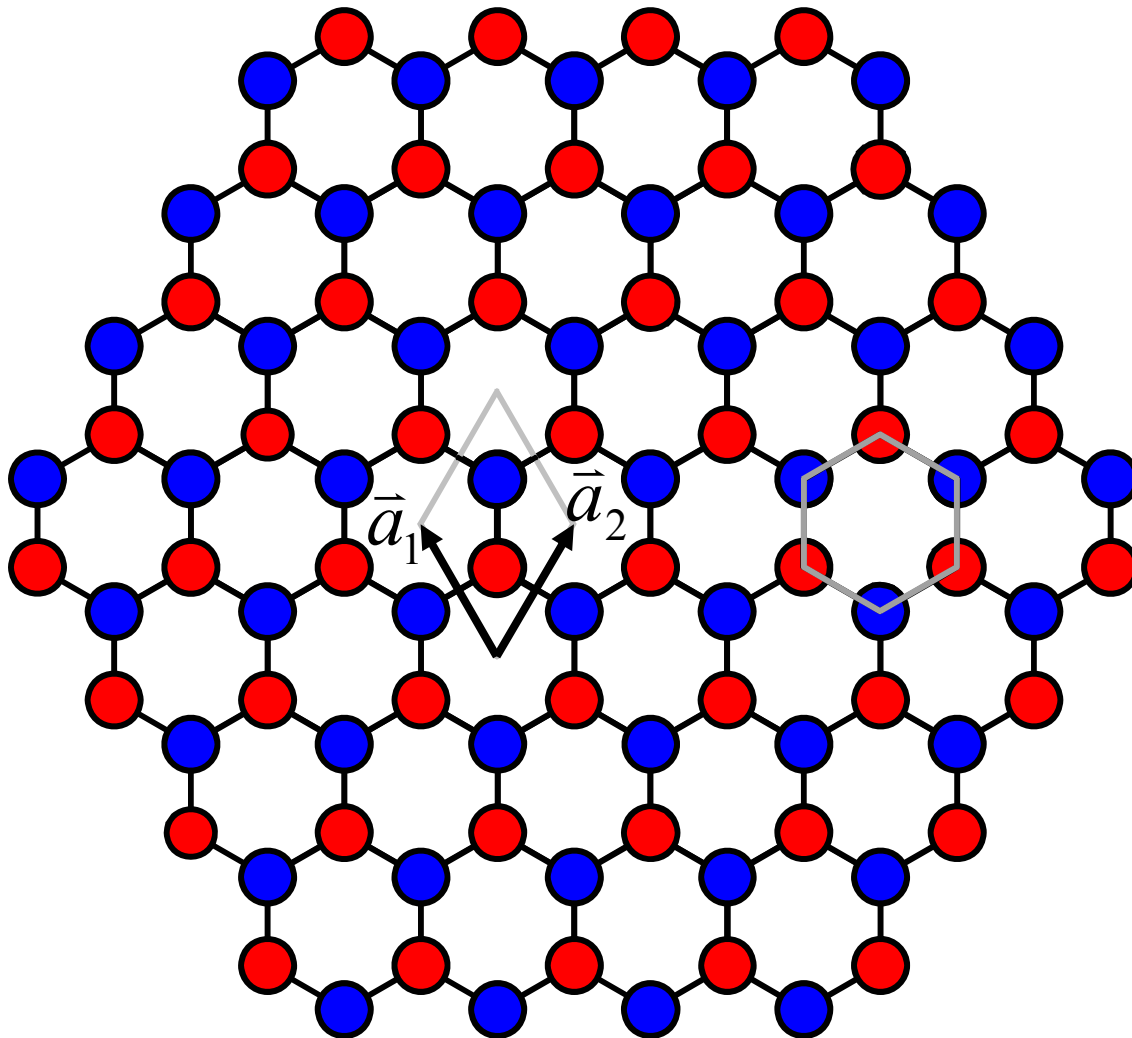
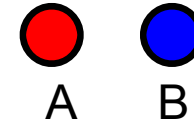
Graphene

Hexagonal lattice;
1 p_z orbital at each site

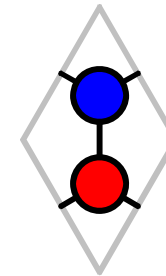


Graphene Unit Cell

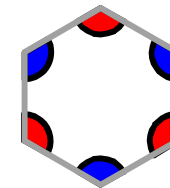
Two *identical* atoms in unit cell:



Two representations of unit cell:



Two atoms

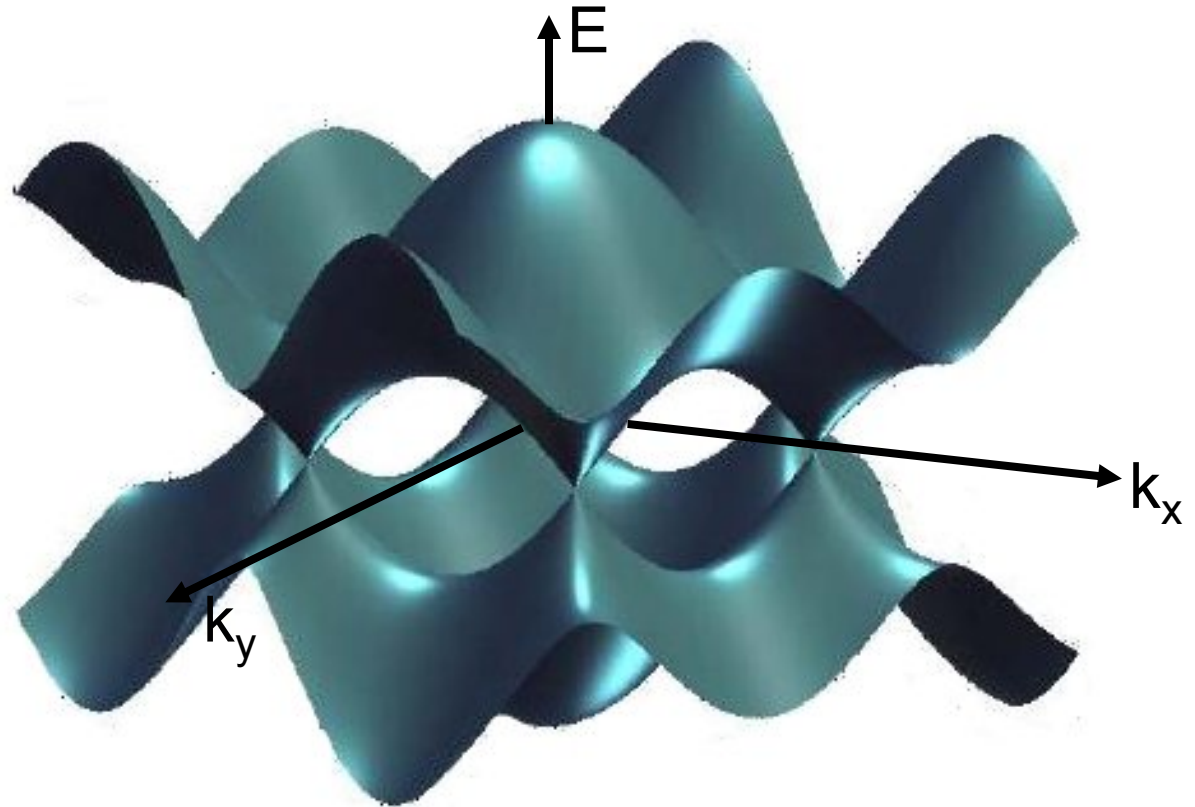
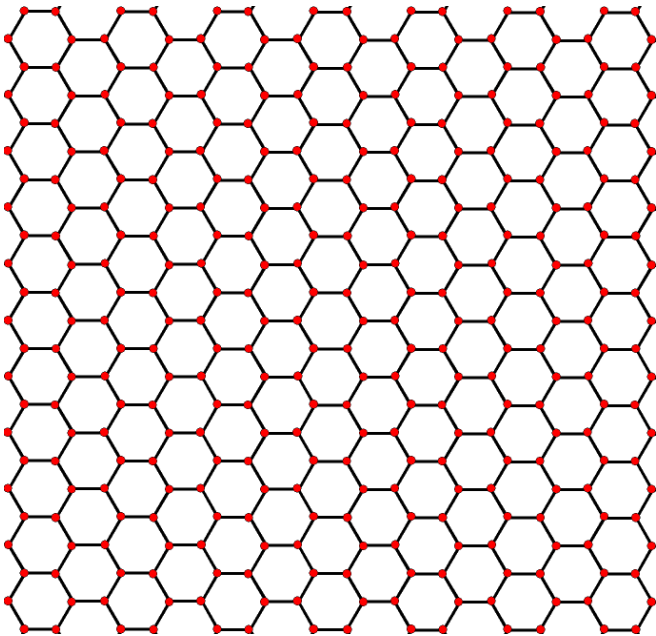


$1/3$ each of 6 atoms = 2 atoms

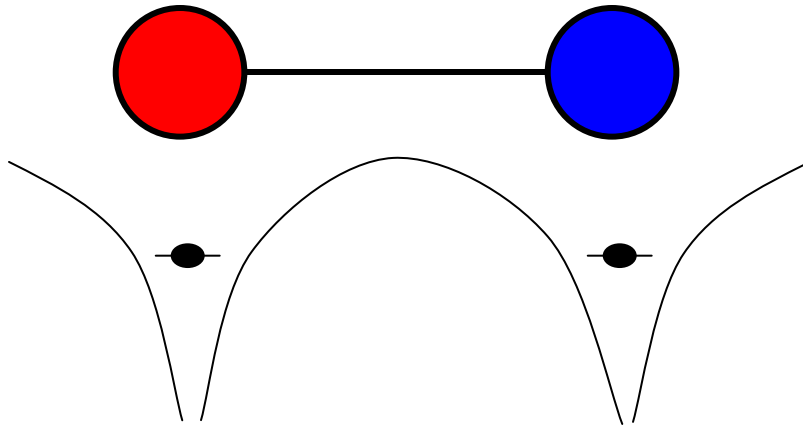
Band Structure of Graphene

Tight-binding model: P. R. Wallace, (1947)
(nearest neighbor overlap = γ_0)

$$E(\mathbf{k}) = E_F \pm \gamma_0 \sqrt{1 + 4 \cos\left(\frac{\sqrt{3}k_x a}{2}\right) \cos\left(\frac{k_y a}{2}\right) + 4 \cos^2\left(\frac{k_y a}{2}\right)}$$



Bonding vs. Anti-bonding



$$H = \begin{bmatrix} 0 & -\gamma_0 \\ -\gamma_0 & 0 \end{bmatrix} \quad E = \pm\gamma_0$$

γ_0 is energy gained per pi-bond

ψ

“anti-bonding”
anti-symmetric wavefunction

$$\psi_1 = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ -1 \end{bmatrix} \quad E_1 = +\gamma_0$$

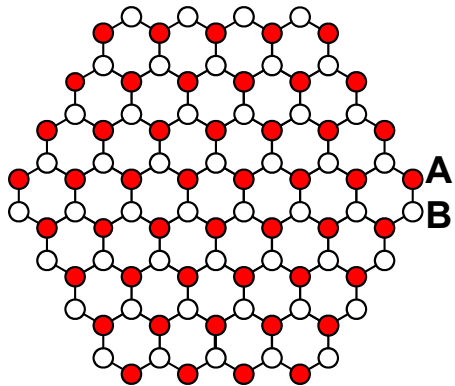
“bonding”
symmetric wavefunction

$$\psi_2 = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ 1 \end{bmatrix} \quad E_2 = -\gamma_0$$

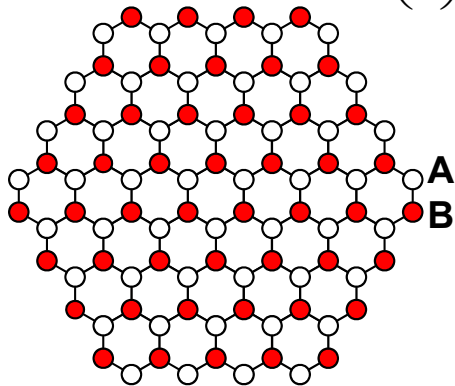
Band Structure of Graphene – Γ point ($k = 0$)

Bloch states:

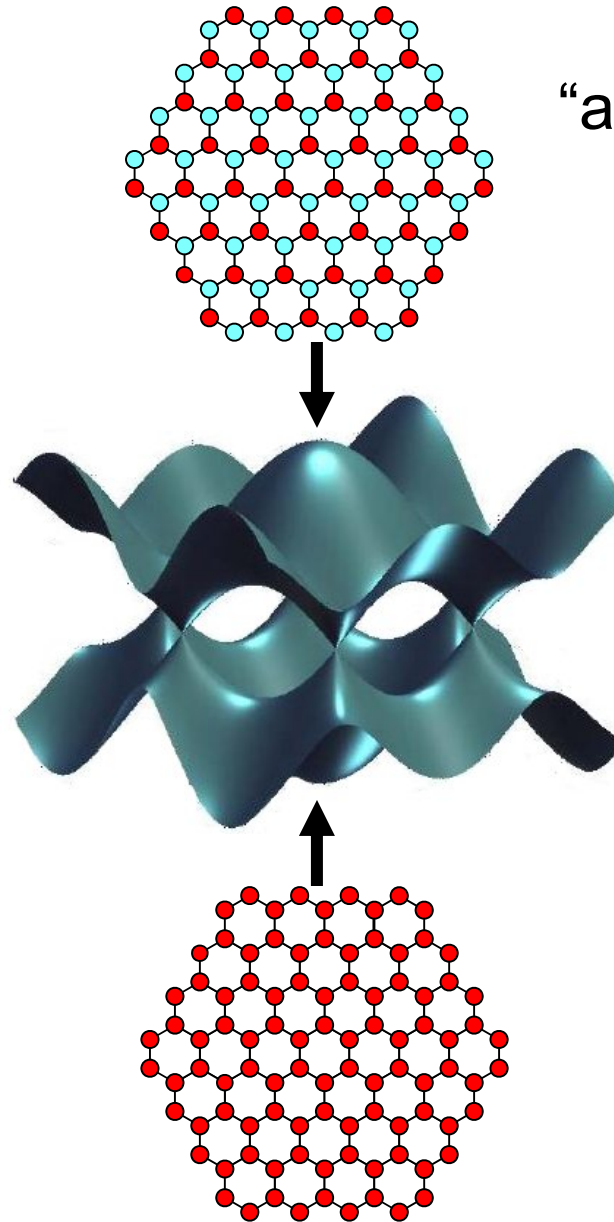
$$F_A(r), \quad \text{or} \quad \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$



$$F_B(r), \quad \text{or} \quad \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$



Γ point:
 $k = 0$



“anti-bonding”

$$E = +3\gamma_0$$

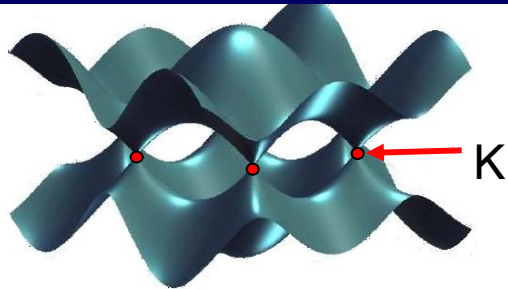
$$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -1 \end{pmatrix}$$

“bonding”

$$E = -3\gamma_0$$

$$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ 1 \end{pmatrix}$$

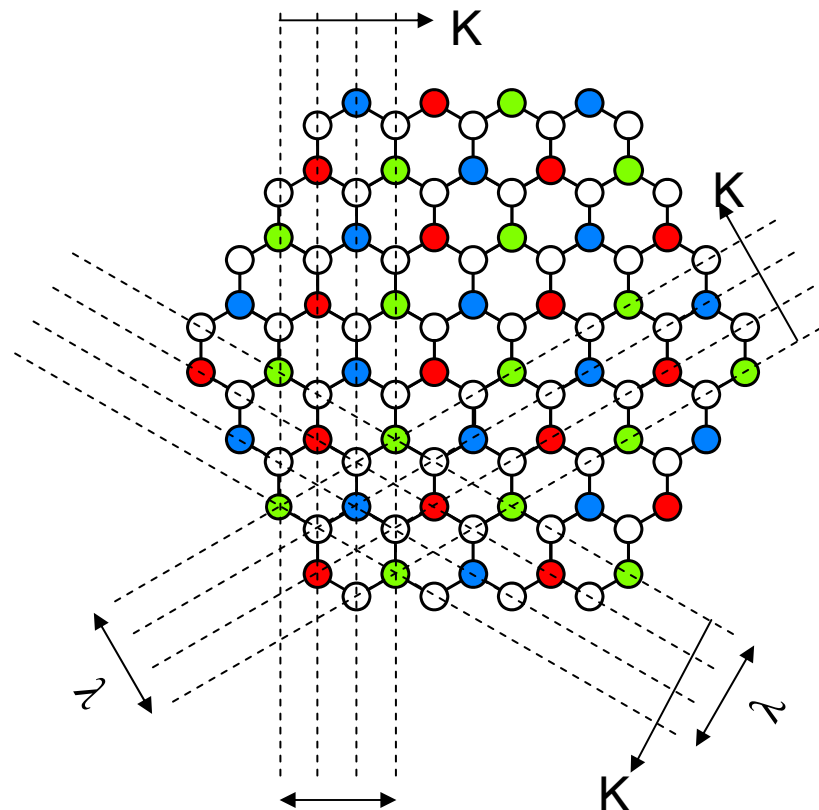
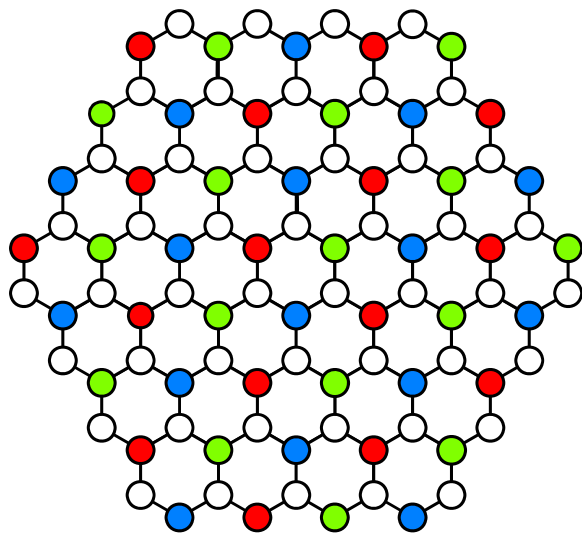
Band Structure of Graphene – K point






$$|\mathbf{K}| = \frac{4\pi}{3a} \Rightarrow \lambda = \frac{3a}{2}$$

$$F_A(r), \text{ or } \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

$$F_B(r), \text{ or } \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$





Phase:


-  1
-  $e^{i\frac{2\pi}{3}}$
-  $e^{i\frac{4\pi}{3}}$

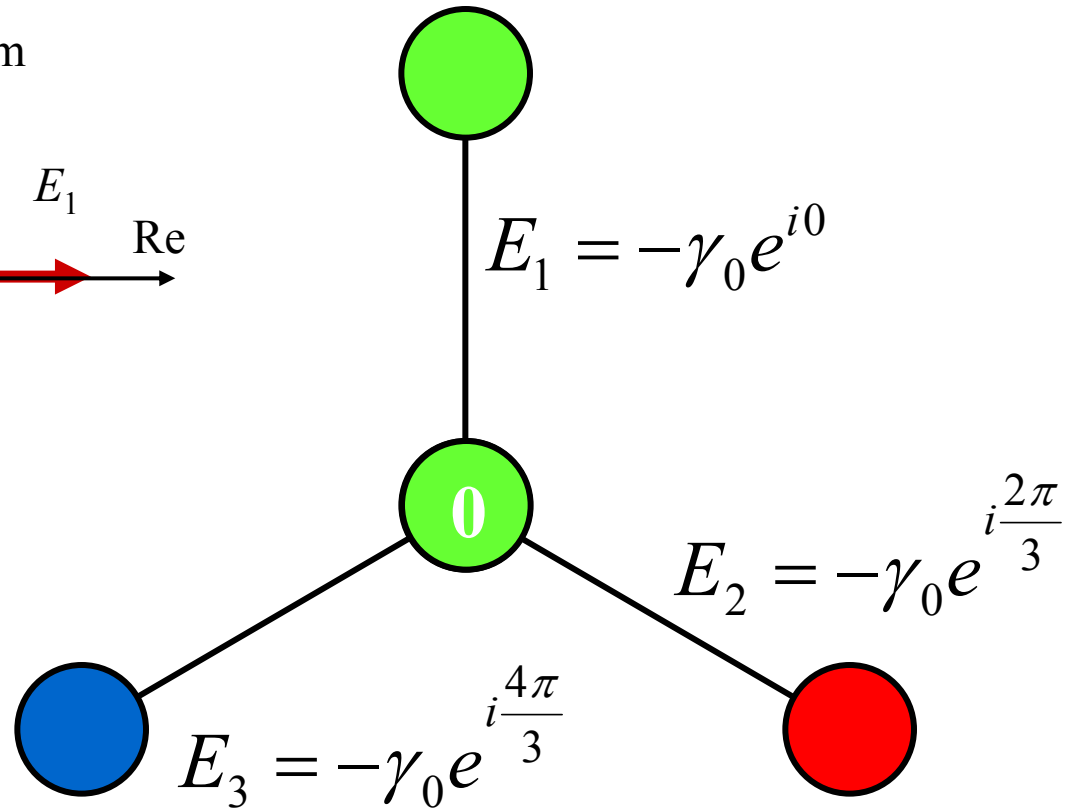
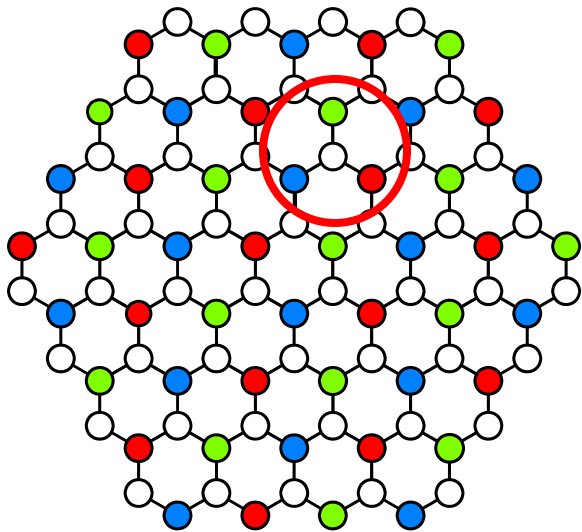
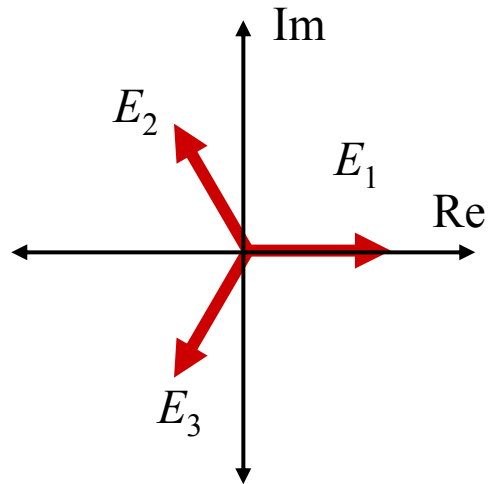
Bonding is Frustrated at K point

Phase:

 $e^{i0} = 1$

 $e^{i\frac{2\pi}{3}}$


 $e^{i\frac{4\pi}{3}}$





$$E = -\gamma_0 \left(e^{i0} + e^{i\frac{2\pi}{3}} + e^{i\frac{4\pi}{3}} \right) = 0$$

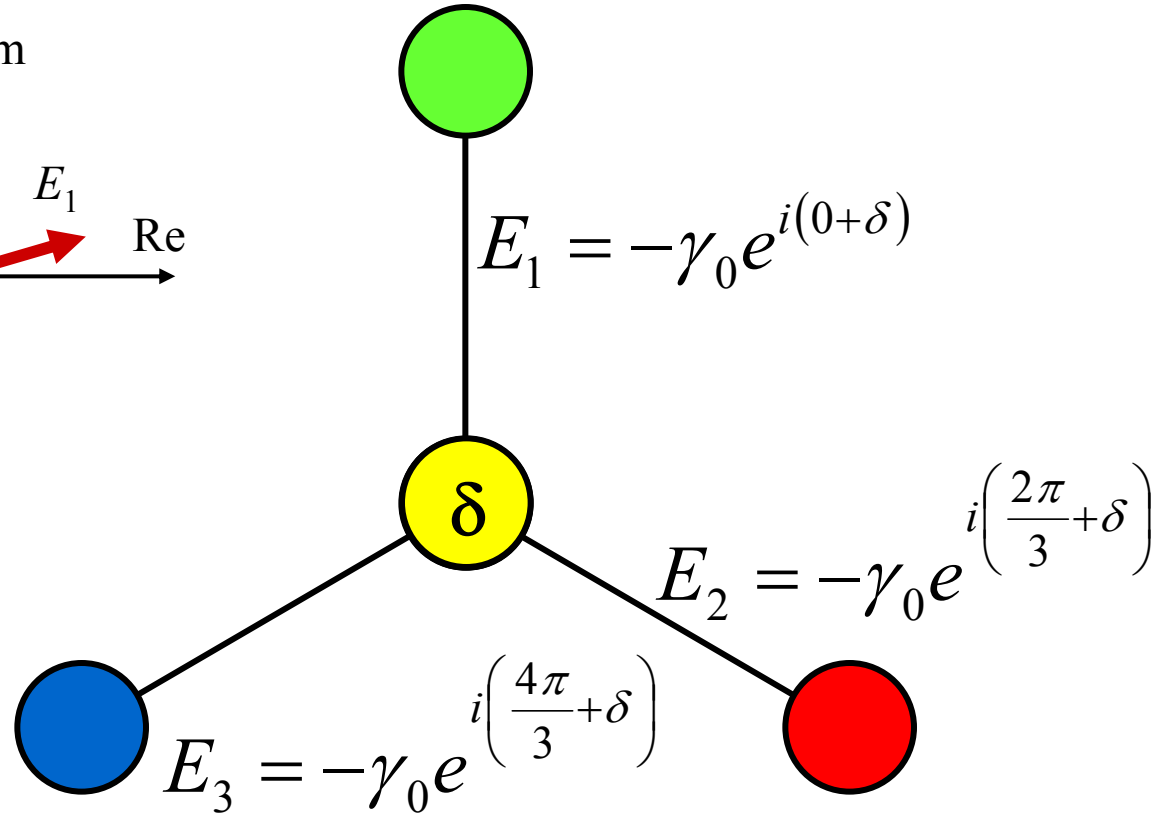
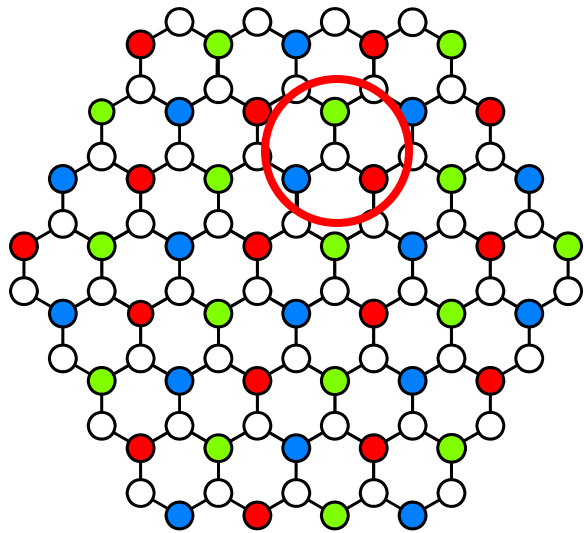
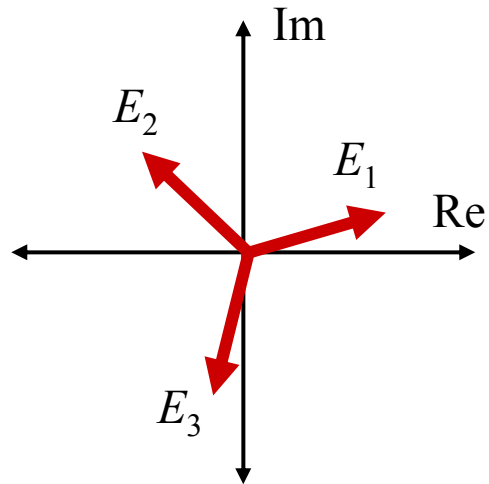
Bonding is Frustrated at K point

Phase:

 $e^{i0} = 1$

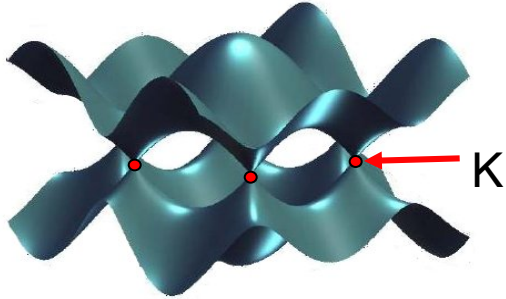
 $e^{i\frac{2\pi}{3}}$

 $e^{i\frac{4\pi}{3}}$



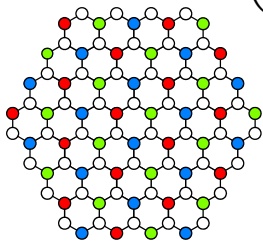
$$E = -\gamma_0 \left(e^{i(0+\delta)} + e^{i\left(\frac{2\pi}{3}+\delta\right)} + e^{i\left(\frac{4\pi}{3}+\delta\right)} \right) = 0e^{i\delta} = 0$$

Bonding is Frustrated at K point

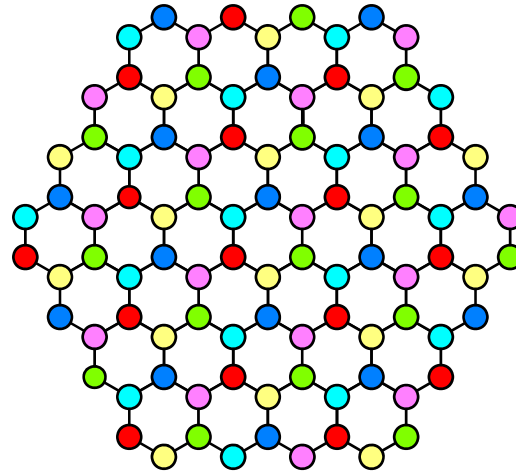
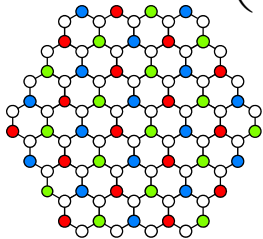


$$|\mathbf{K}| = \frac{4\pi}{3a} \implies \lambda = \frac{3a}{2}$$

$$F_A(r), \text{ or } \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$



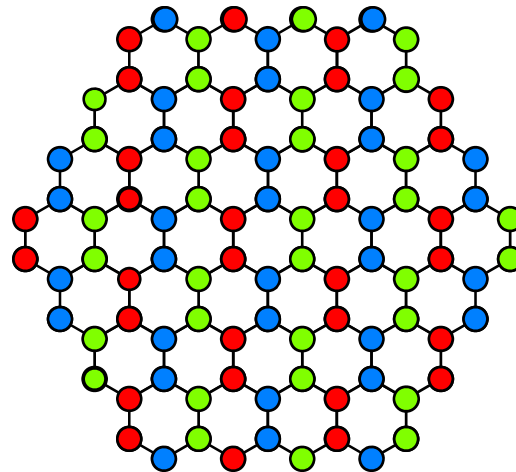
$$F_B(r), \text{ or } \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$



“anti-bonding”

$$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -1 \end{pmatrix}$$

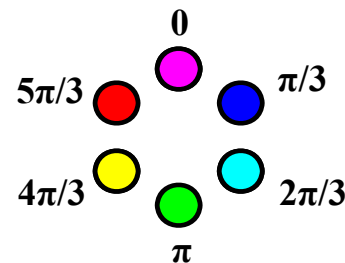
$$E = 0!$$



“bonding”

$$\frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ 1 \end{pmatrix}$$

$$E = 0!$$



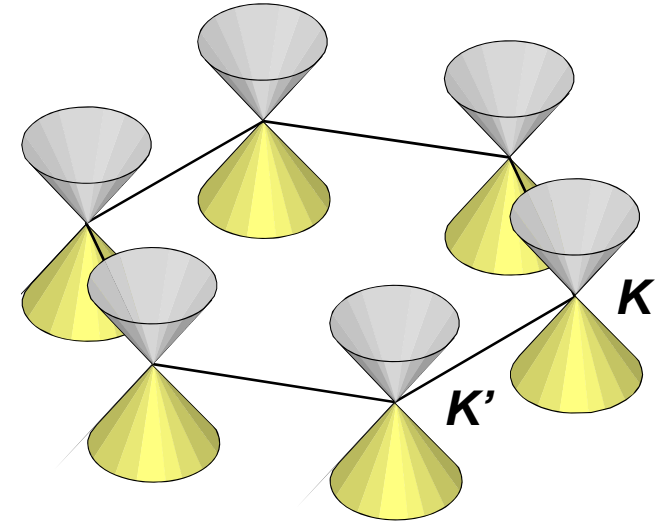
K point:
Bonding and anti-bonding
are degenerate!

Band Structure of Graphene: k-p approximation

Hamiltonian:

$$\hbar v_F \begin{pmatrix} 0 & k_x - ik_y \\ k_x + ik_y & 0 \end{pmatrix} \begin{pmatrix} F_A(r) \\ F_B(r) \end{pmatrix} = \varepsilon \begin{pmatrix} F_A(r) \\ F_B(r) \end{pmatrix}$$

$$\hbar v_F (\boldsymbol{\sigma} \cdot \mathbf{k}) F(r) = \varepsilon F(r)$$



Eigenvectors:

$$|k\rangle = \frac{1}{\sqrt{2}} e^{ik \cdot r} \begin{pmatrix} -ibe^{-i\theta_k/2} \\ e^{i\theta_k/2} \end{pmatrix};$$

θ_k is angle \mathbf{k} makes with y-axis
 $b = 1$ for electrons, -1 for holes

Energy:

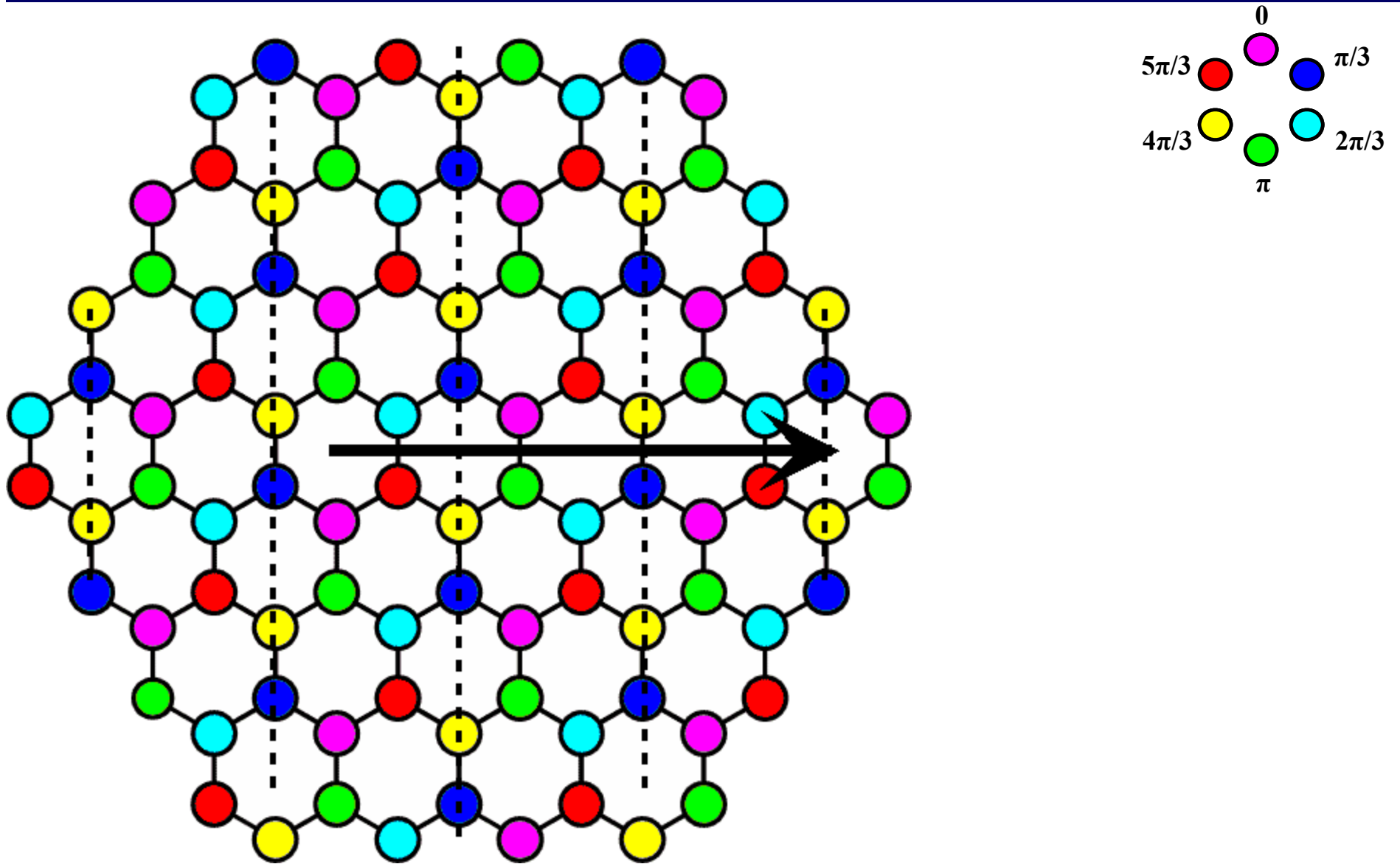
$$\varepsilon = b\hbar v_F |k|$$

linear dispersion relation
 “massless” electrons

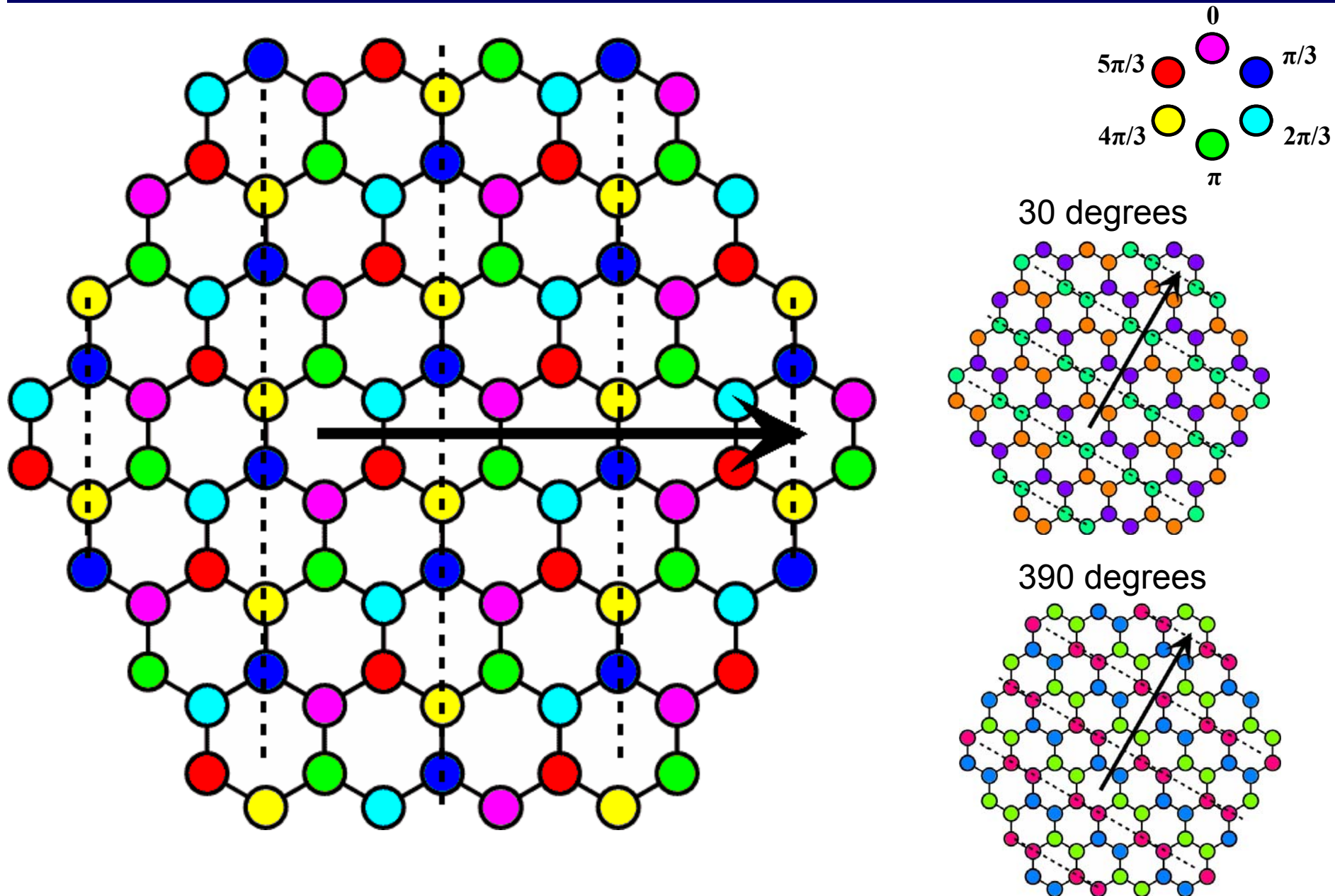
electron has “pseudospin”

\mathbf{M} points parallel (anti-parallel) to momentum

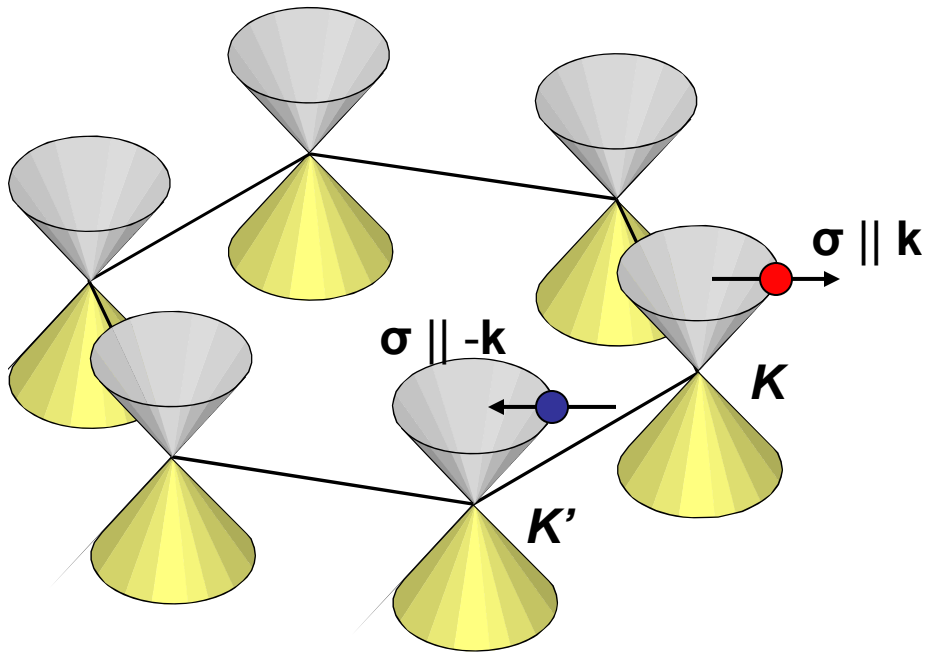
Visualizing the Pseudospin



Visualizing the Pseudospin



Pseudospin

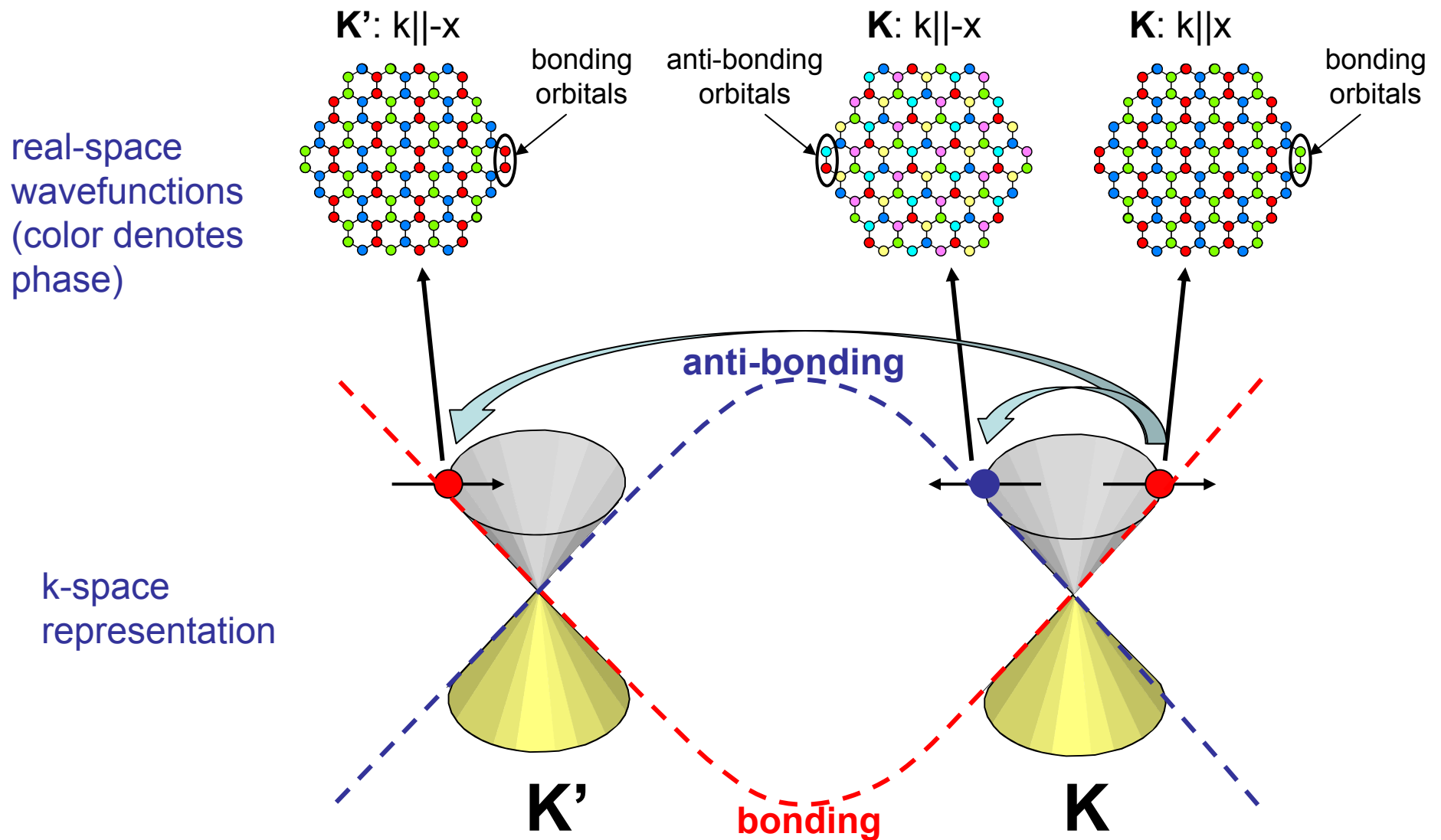


$$H_K = \hbar v_F \vec{\sigma} \cdot \vec{k} = \hbar v_F \begin{pmatrix} 0 & k_x - ik_y \\ k_x + ik_y & 0 \end{pmatrix}$$

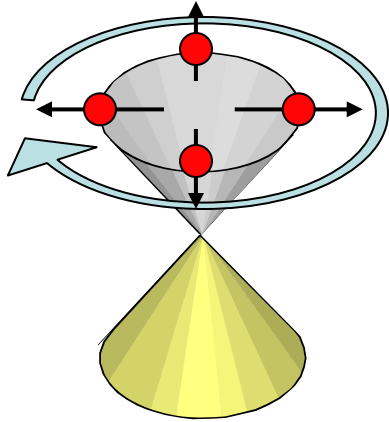
$$H_{K'} = \hbar v_F \vec{\sigma}' \cdot \vec{k}$$

- Hamiltonian corresponds to spin-1/2 “pseudospin”
Parallel to momentum (K) or anti-parallel to momentum (K')
- Orbits in k-space have Berry's phase of π

Pseudospin: Absence of Backscattering



“Pseudospin”: Berry’s Phase in IQHE

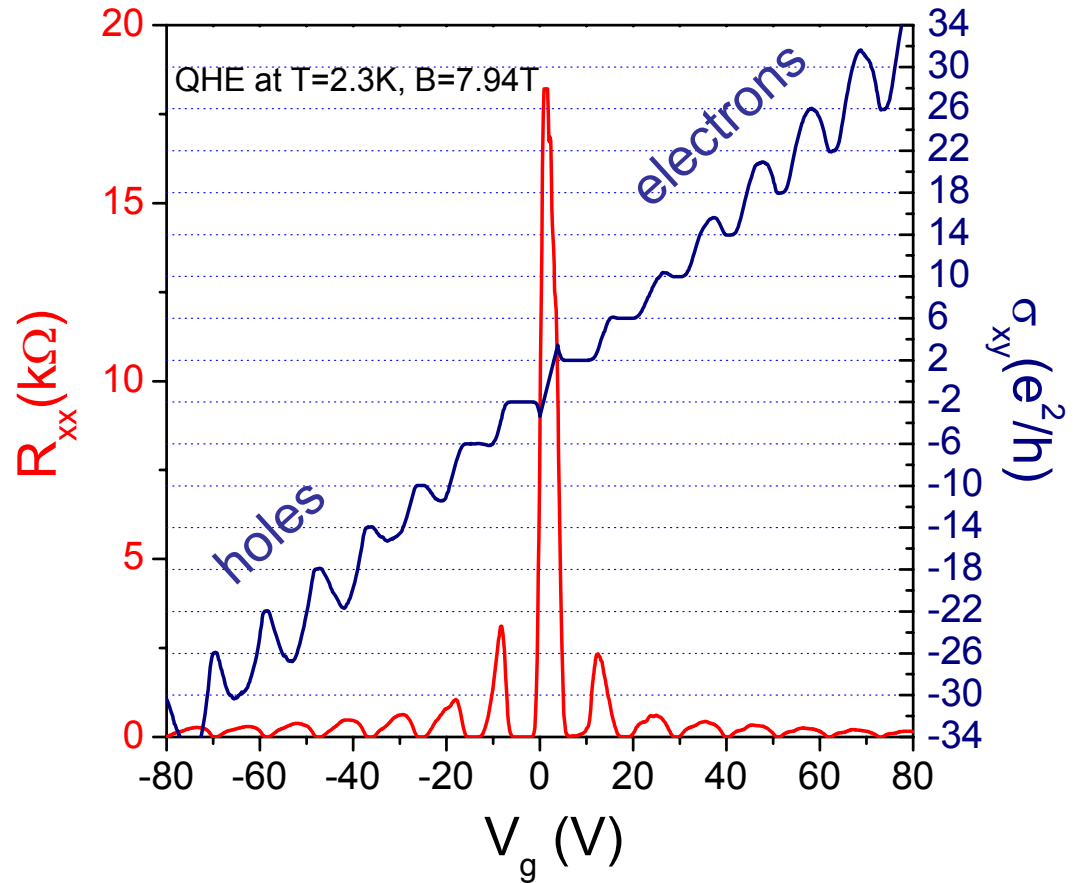


π Berry’s phase for electron orbits results in $\frac{1}{2}$ -integer quantized Hall effect

$$\sigma_{xy} = \nu \frac{e^2}{h} \quad \nu = 4 \left(n + \frac{1}{2} \right)$$

$$g_s g_v = 2 \times 2 = 4$$

Berry’s phase = π



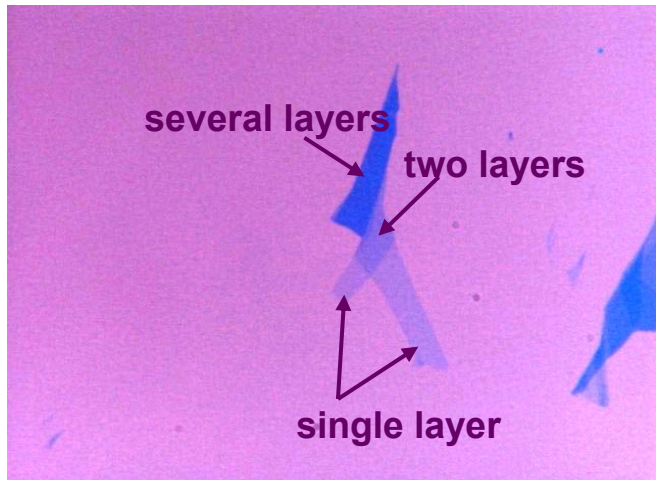
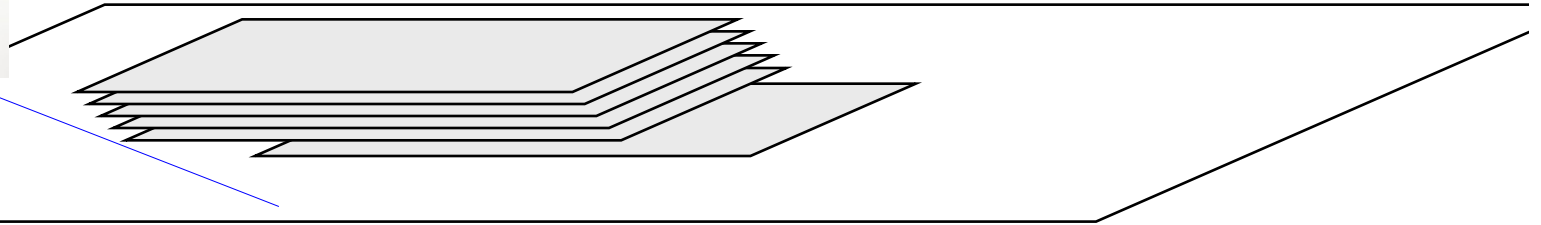
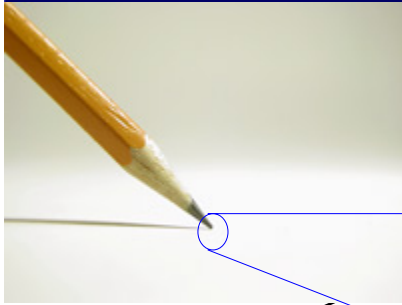
Outline

- I. Introduction to Graphene
 - “Massless” electrons
 - Pseudospin and Berry’s phase

- II. Fabrication and Characterization of Graphene on SiO₂
 - Micro-Raman spectroscopy
 - Cleaning graphene

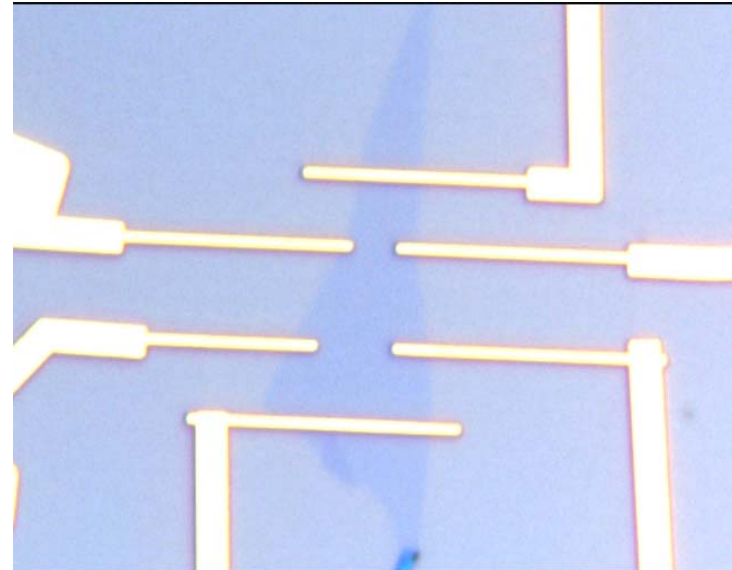
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Graphene – Fabrication



Optical micrograph (layer thickness verified by AFM)

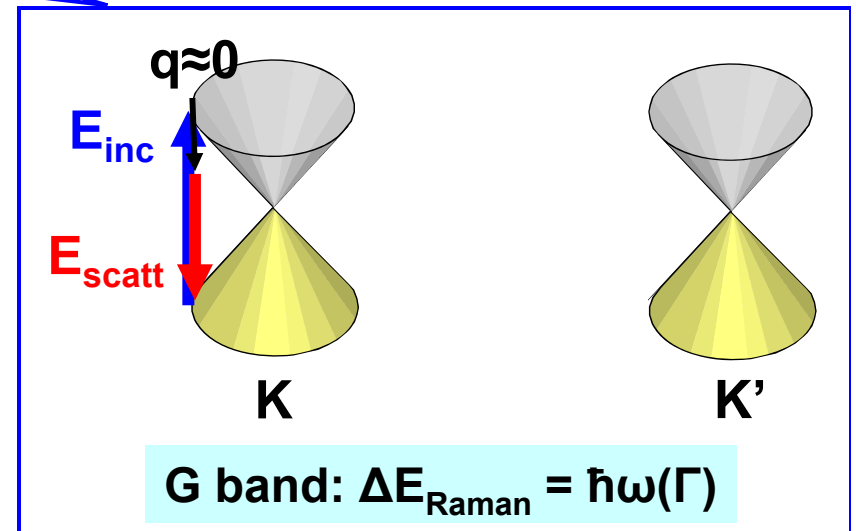
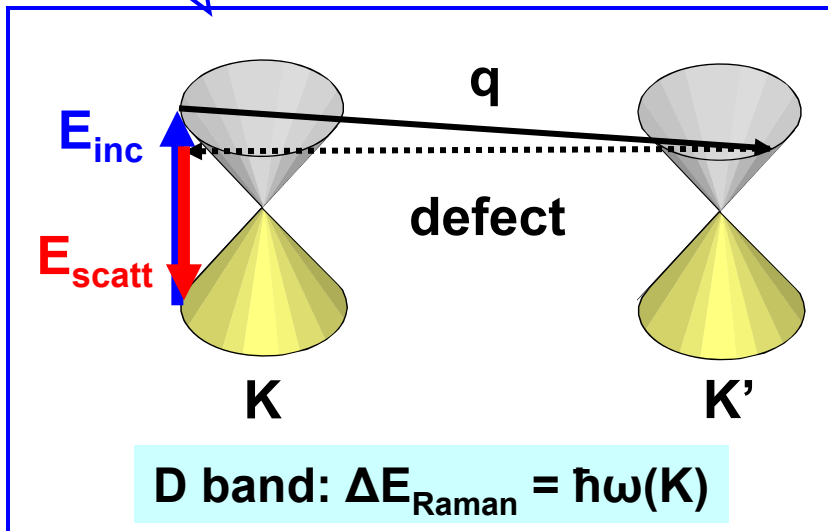
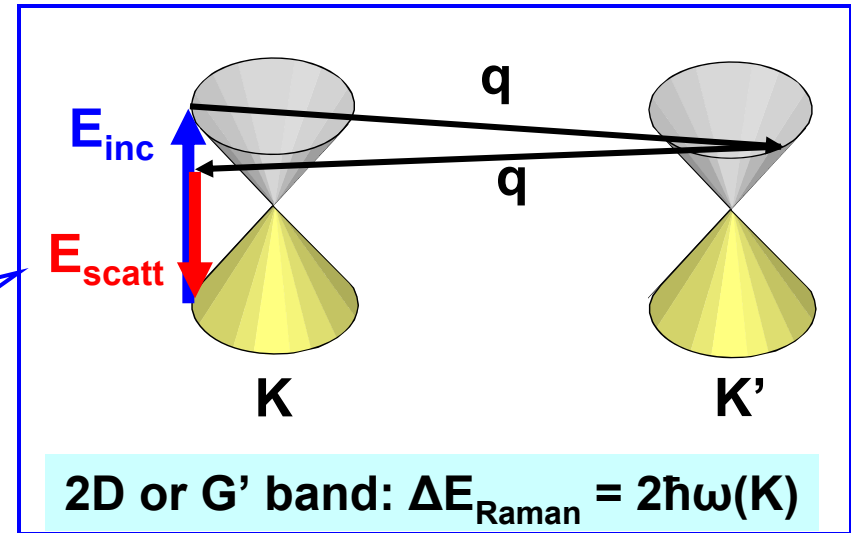
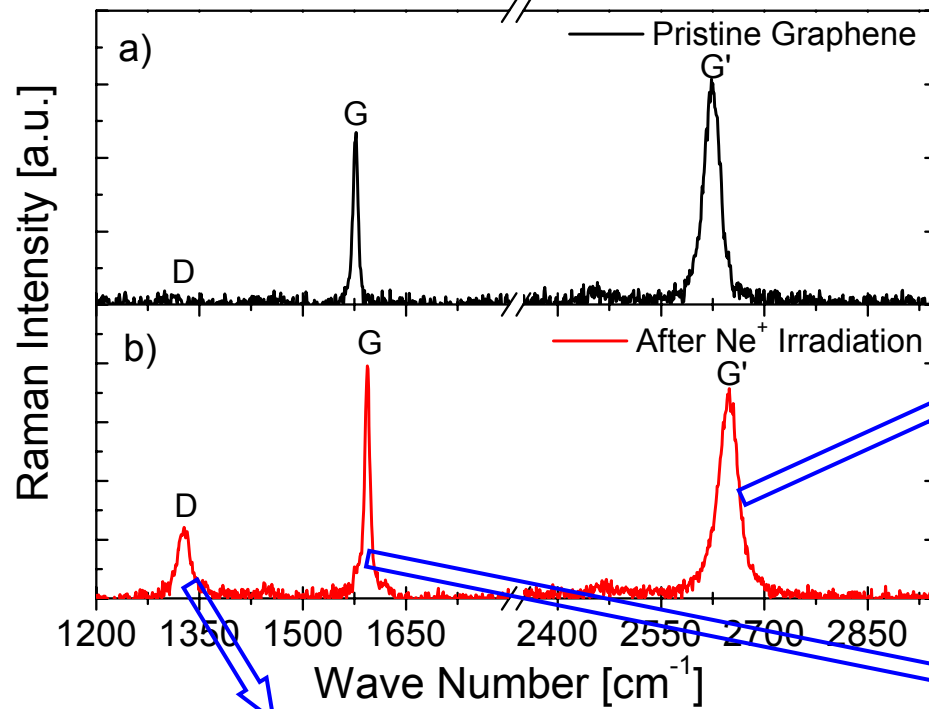
- Starting material is single-crystal Kish graphite
- Mechanically exfoliate on 300 nm SiO₂/Si chips



Single layer device after e-beam lithography

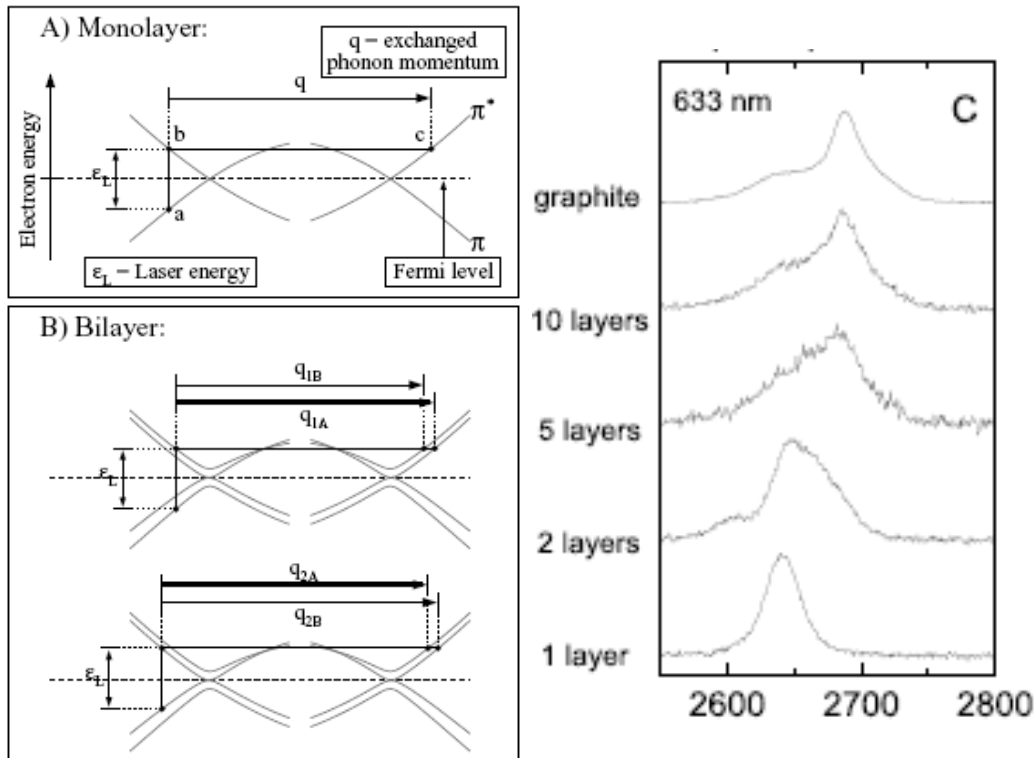
Method adapted from Novoselov, et al. *PNAS* **102** 10341 (2005)

Raman spectroscopy of graphene



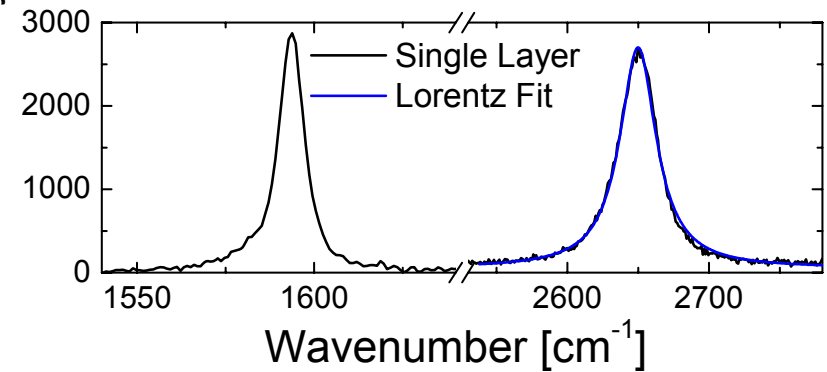
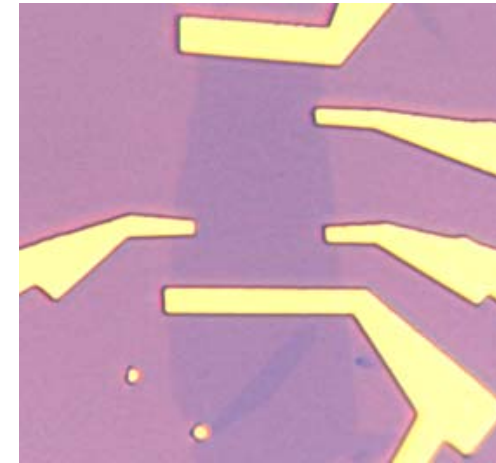
Graphene fingerprint in Micro-Raman

- Raman G' band is two-photon/two-phonon resonant excitation; sensitive to electronic structure of graphene



Ferrari, et al., *PRL* **97**, 187401 (2006)

Fuhrer group sample

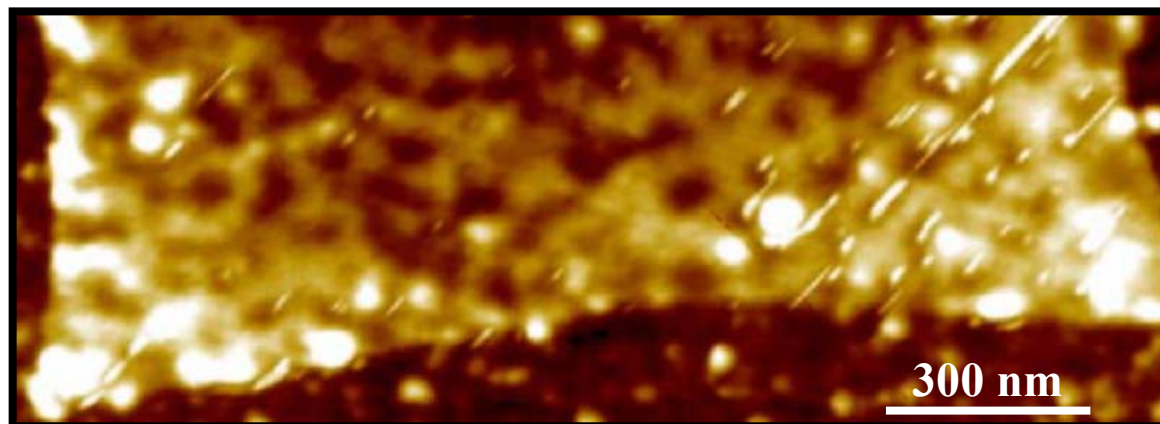
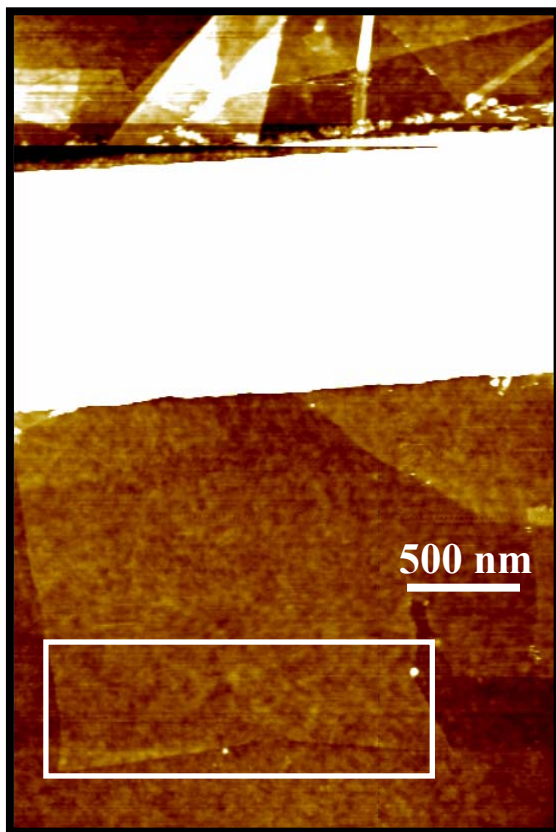


single Lorentzian G' peak indicates single-layer graphene

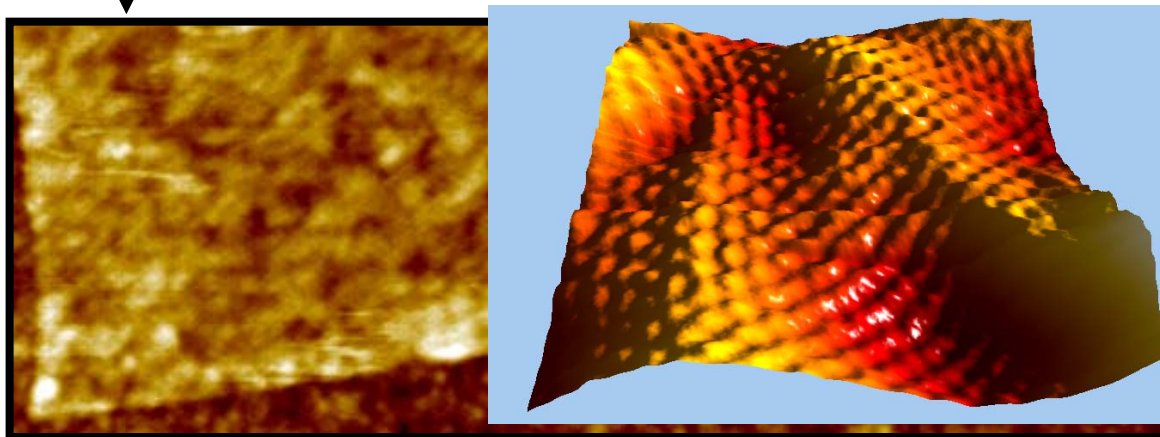
Removing Photoresist Residue from Graphene

Ishigami, et al., *Nano Letters* 7, 1643 (2007)

Residues from PMMA/MMA photoresist



Novel photoresist residue removal process
Anneal in flowing H₂ at 400°C



Complete removal of photoresist residues

Atomically clean STM images

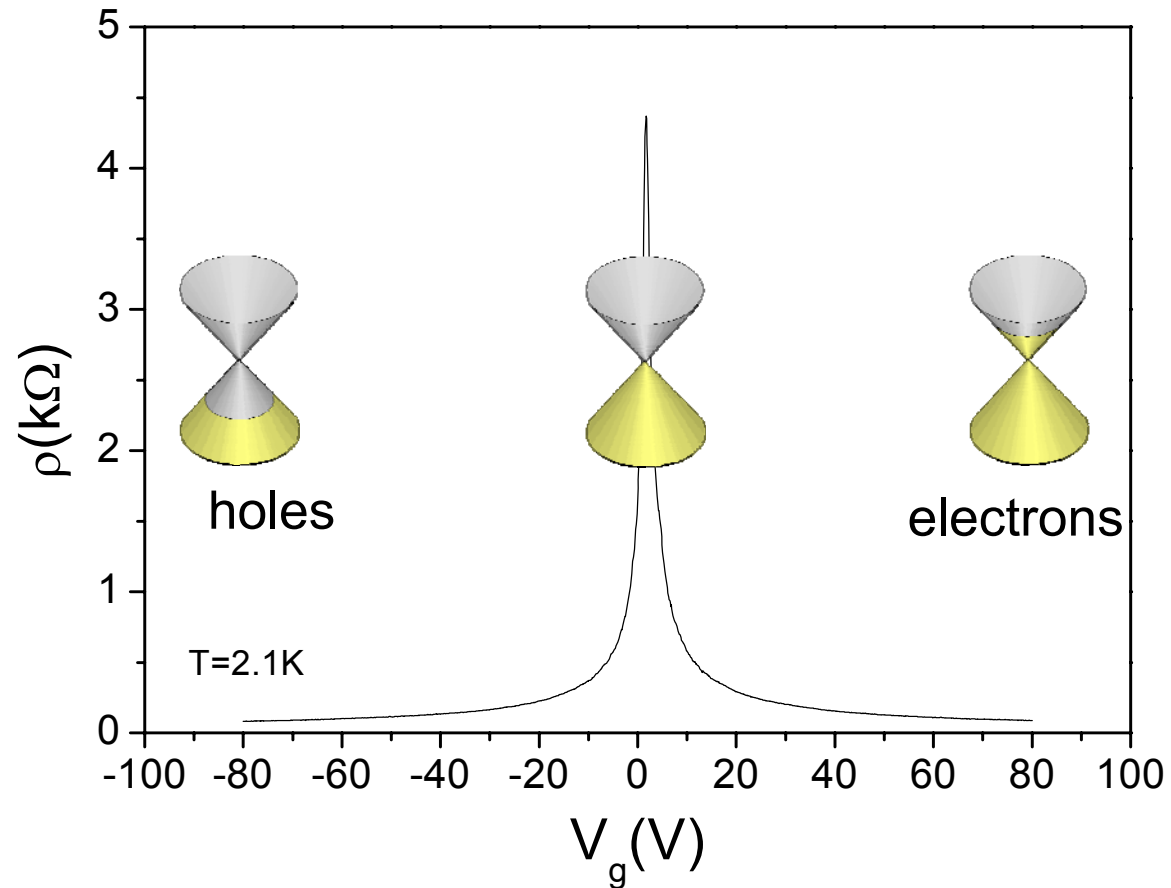
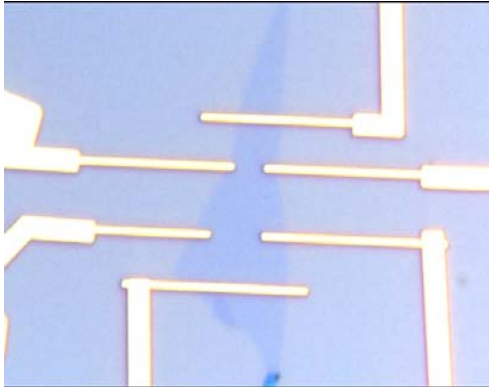
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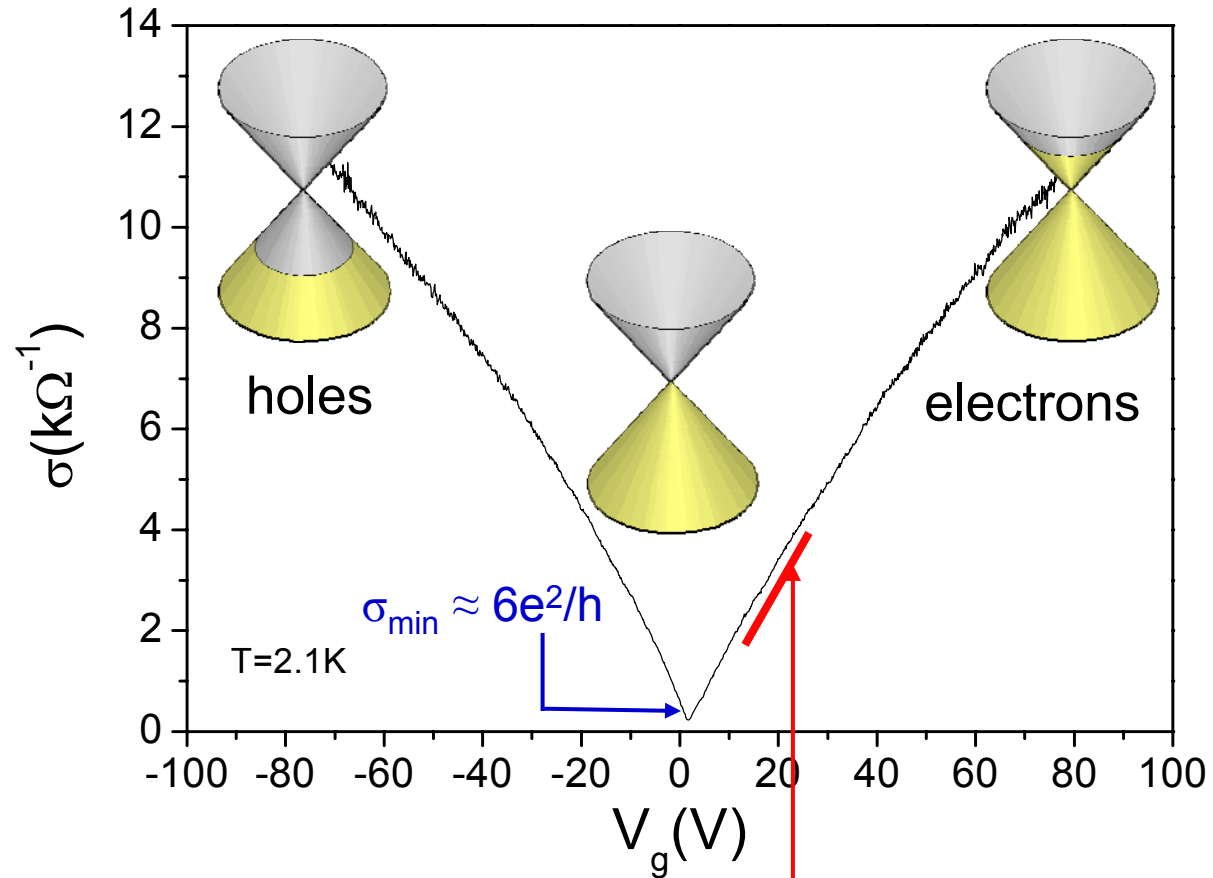
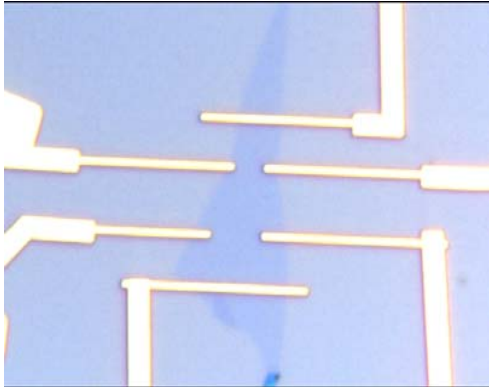
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Electrical Characterization of Graphene



- Ambipolar, symmetric conduction
- Finite minimum conductivity $\sim [4-10]e^2/h$
- Field-effect mobility up to 20,000 cm^2/Vs

Electrical Characterization of Graphene



- Ambipolar, symmetric conduction
- Finite minimum conductivity $\sim [4-10]e^2/h$
- Field-effect mobility up to 20,000 cm^2/Vs

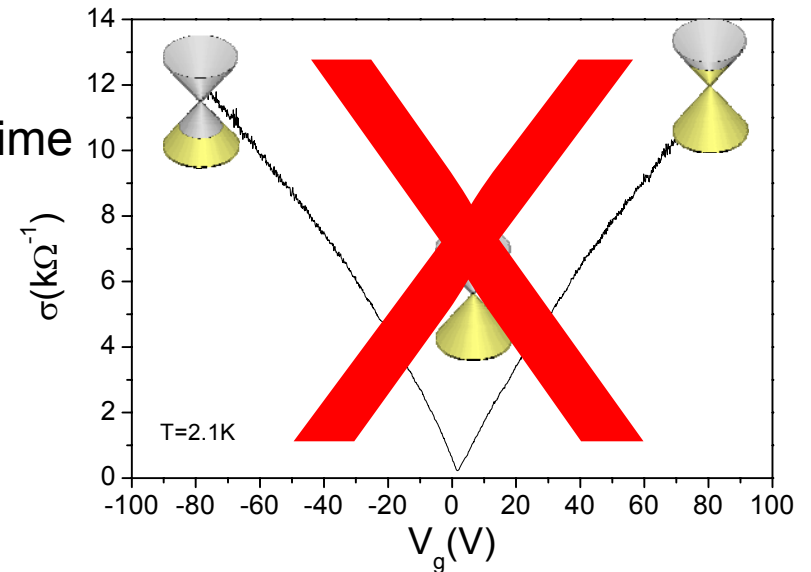
$$\mu_{FE} = \frac{1}{e} \frac{d\sigma}{dn} = \frac{1}{c_g} \frac{d\sigma}{dV_g}$$

Boltzmann Transport

$$\sigma = \frac{e^2 v_F^2}{2} D(E) \tau$$

$D(E)$ is density of states
 τ is momentum relaxation time
 v_F is Fermi velocity

Graphene: $D(E) = \frac{2E_F}{\pi \hbar^2 v_F^2}$



But: Fermi's Golden Rule:

$$\frac{1}{\tau} \propto \frac{2\pi}{\hbar} |\langle k|V|k' \rangle|^2 D(E)$$

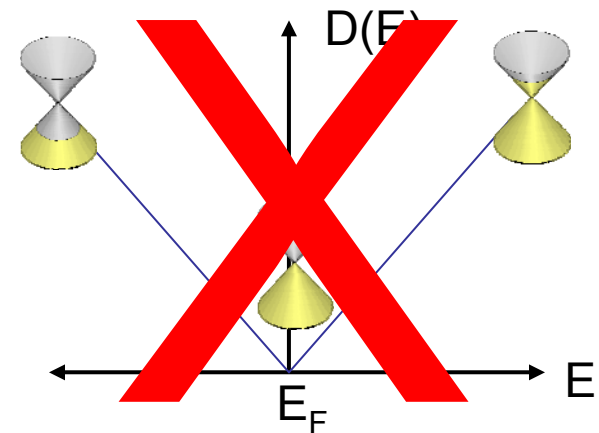
$$\therefore D(E)\tau \propto \text{constant!}$$



σ is independent of E_F !

True for point defects, phonons

see e.g. Pietronero (1980), T. Ando (1996)

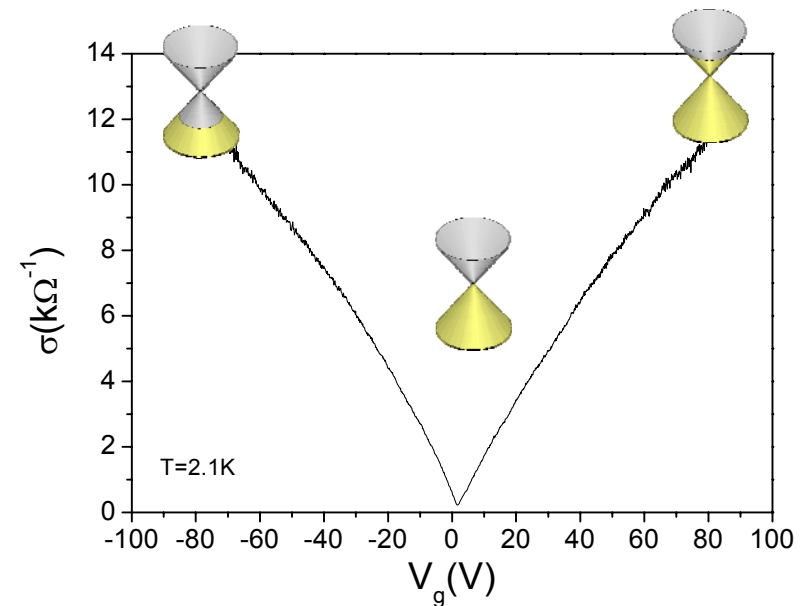


How to explain linear $\sigma(V_g)$?

$$\frac{1}{\tau} \propto \frac{2\pi}{\hbar} \left| \langle k | V | k' \rangle \right|^2 D(E)$$

Interaction must be q -dependent

$$q = |\mathbf{k} - \mathbf{k}'| \sim k_F$$



Coulomb interaction: $V_{Coulomb} = \frac{2\pi e^2}{\kappa q}$

N.B. In graphene, screened Coulomb interaction remains $\sim 1/k_F$



$$\sigma \sim E_F^{1/2} \sim n \sim V_g$$

See:

Ando, *J. Phys. Soc. Jpn.* **75**, 074716 (2006)

Nomura & MacDonald *PRL* **98**, 076602 (2007)

Cheianov & Fal'ko *PRL* **97**, 226801 (2006)

Hwang, Adam, & Das Sarma, *PRL* **98**, 186806 (2007)

Minimum Conductivity of Graphene

- At minimum conductivity point, graphene breaks into electron and hole “puddles”
- Minimum conductivity *decreases* with increasing impurity concentration

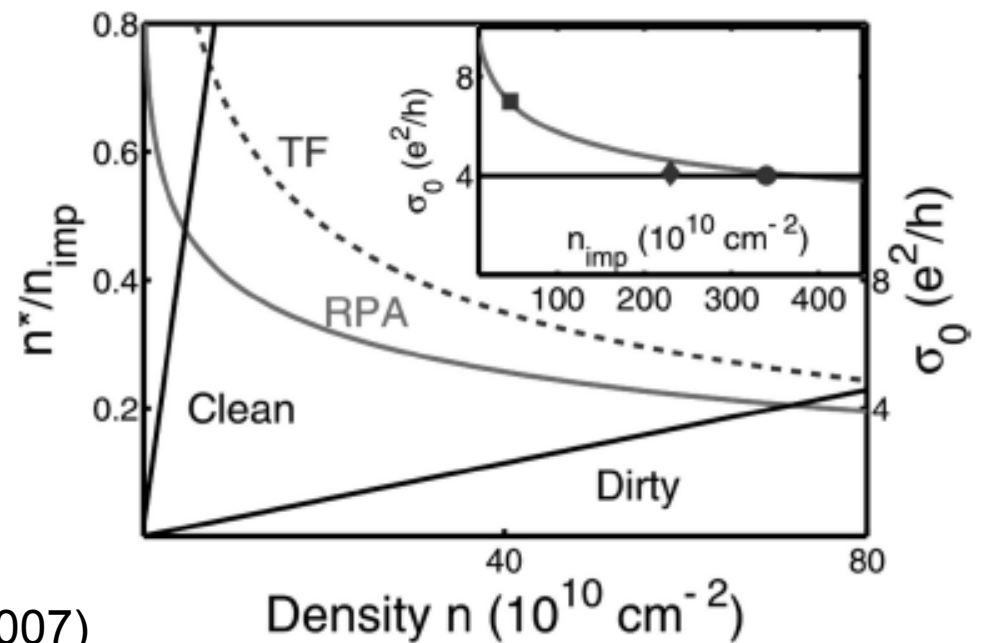
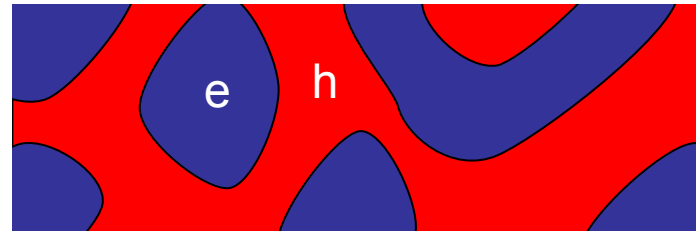
$$\sigma_{\min} = n^* e \mu$$

Residual density in puddles

$$n^* = n_{\text{imp}} [2r_s^2 C_0^{\text{RPA}}(r_s, 4k_F d)]$$

$$\mu = \frac{1}{n_{\text{imp}}} \left[20 \frac{e}{h} \right]$$

Weak function of n_{imp}, r_s, d



Adam, et al., *PNAS* **104**, 18392 (2007)

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- II. Fabrication and Characterization of Graphene on SiO₂
 - Micro-Raman spectroscopy
 - Cleaning graphene

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 - Lattice defects

Charged Impurity Scattering: Potassium Doping in UHV

J. H. Chen, et al. *Nature Physics* 4, 377 (2008)

- Clean graphene in UHV at $T = 20$ K
- Potassium evaporated on graphene from getter

Upon doping with K:

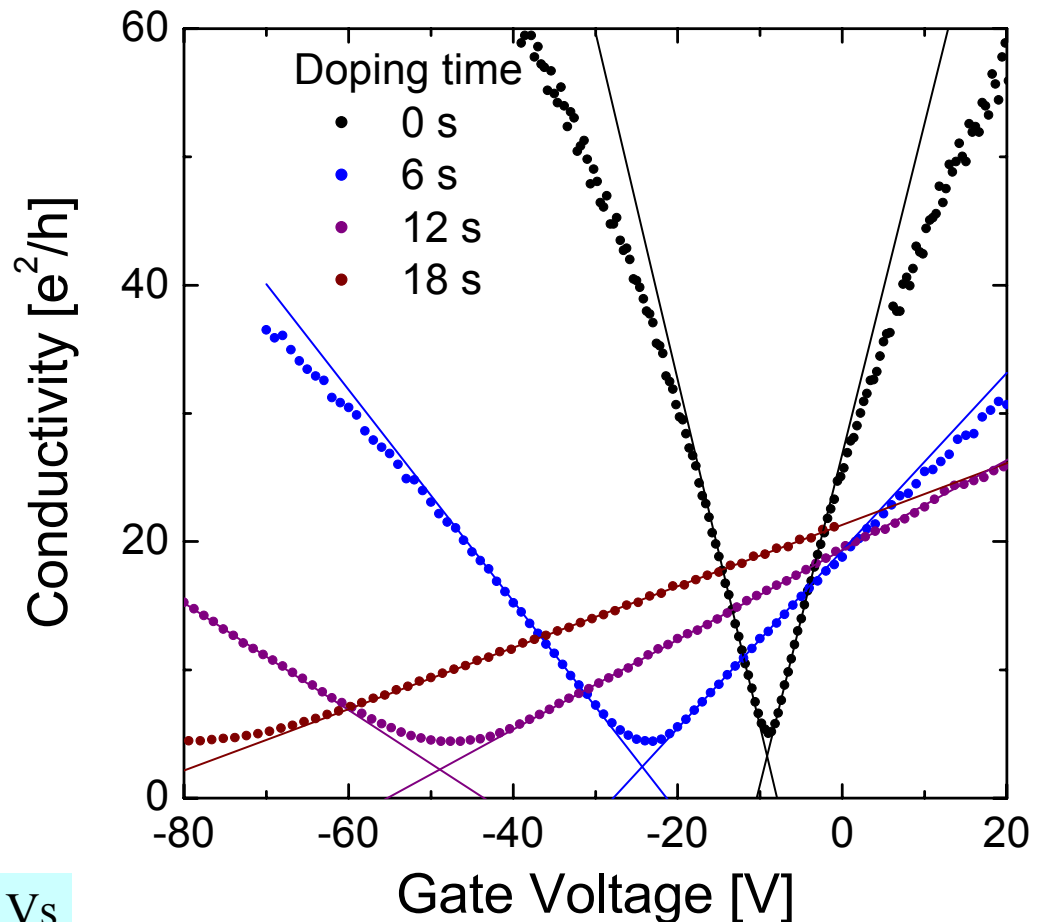
- 1) mobility decreases
- 2) $\sigma(V_g)$ more linear
- 3) σ_{\min} shifts to negative V_g
- 4) plateau around σ_{\min} broadens
- 5) σ_{\min} decreases (slightly)

All these feature predicted for Coulomb scattering in graphene

Adam, et al., PNAS 104, 18392 (2007)

Magnitude of scattering in quantitative agreement with theory:

$$\mu = \frac{5 \times 10^{11} \text{ V s}}{n_{\text{imp}}}$$



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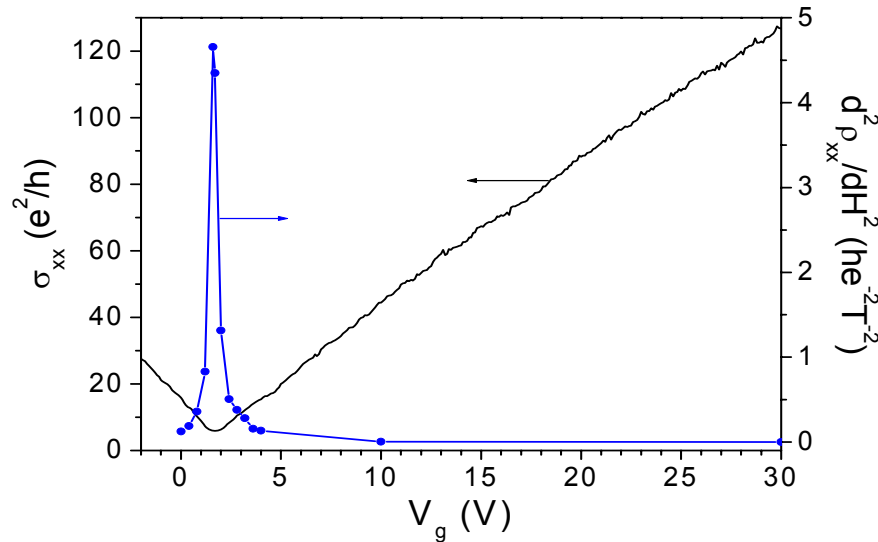
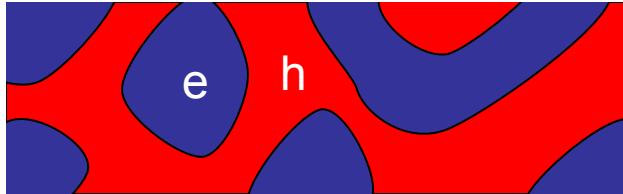
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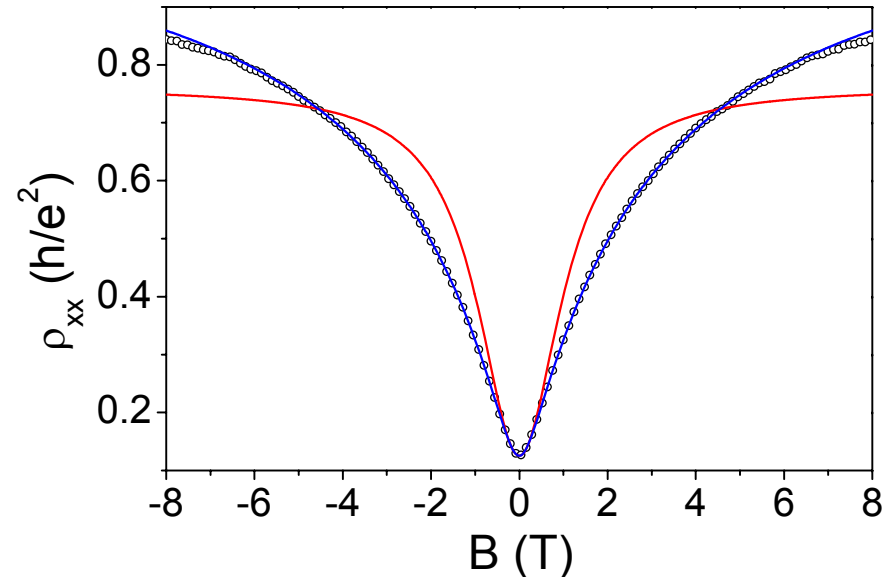
Magnetoresistance at Minimum Conductivity Point

S. Cho and M. S. Fuhrer, *PRB* **77**, 084102(R) (2008)

- At minimum conductivity point, graphene breaks into electron and hole “puddles”
Hwang, et al., *PRL* **98**, 186806 (2007); Adam, et al., *PNAS* **104**, 18392 (2007)



Large spike in magnetoresistance at Dirac point



- Expt. $T = 300$ K
- Two-fluid model $\alpha = 0.4$
- EMT model $\sigma_{xx,1} = 0.88$

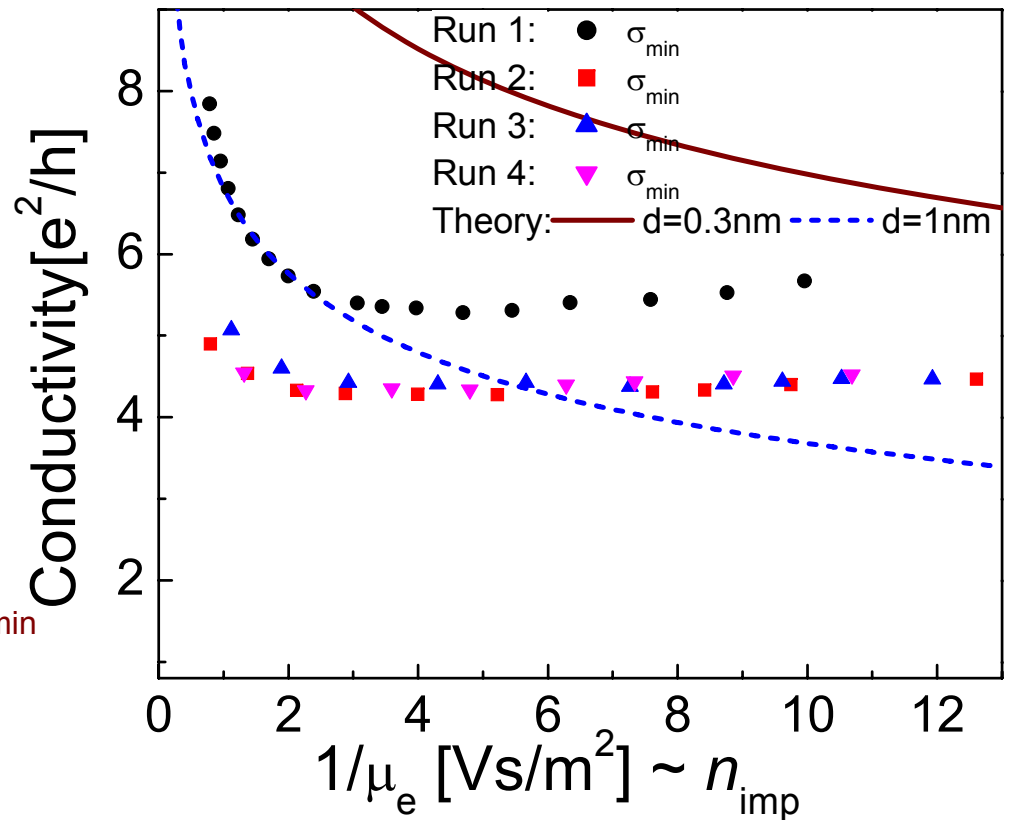
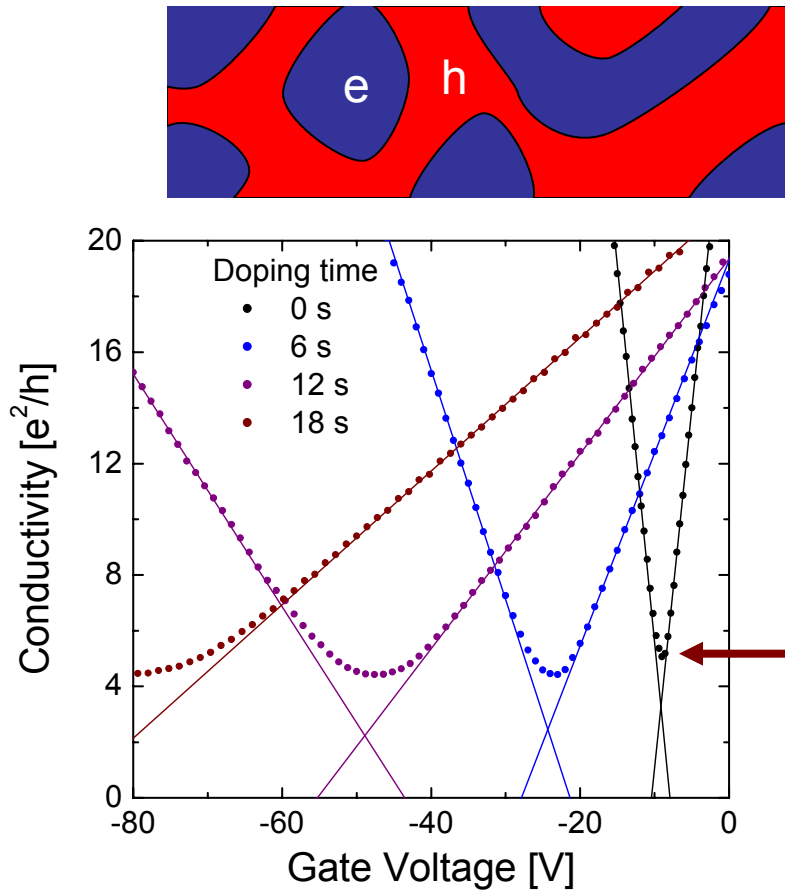
Functional form of $\rho(B)$:
effective medium theory for inhomogeneous e/h regions
[Guttal and Stroud, *PRB* **71** 201304 (2005)]

Charged Impurity Scattering: Minimum Conductivity

J. H. Chen, et al. *Nature Physics* 4, 377 (2008)

- At minimum conductivity point, graphene breaks into electron and hole “puddles”
- Minimum conductivity *decreases* with increasing impurity concentration

Adam, et al., *PNAS* 104, 18392 (2007)



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Electron-Phonon Scattering

J. H. Chen, et al. *Nature Nanotechnology* 3, 206 (2008)

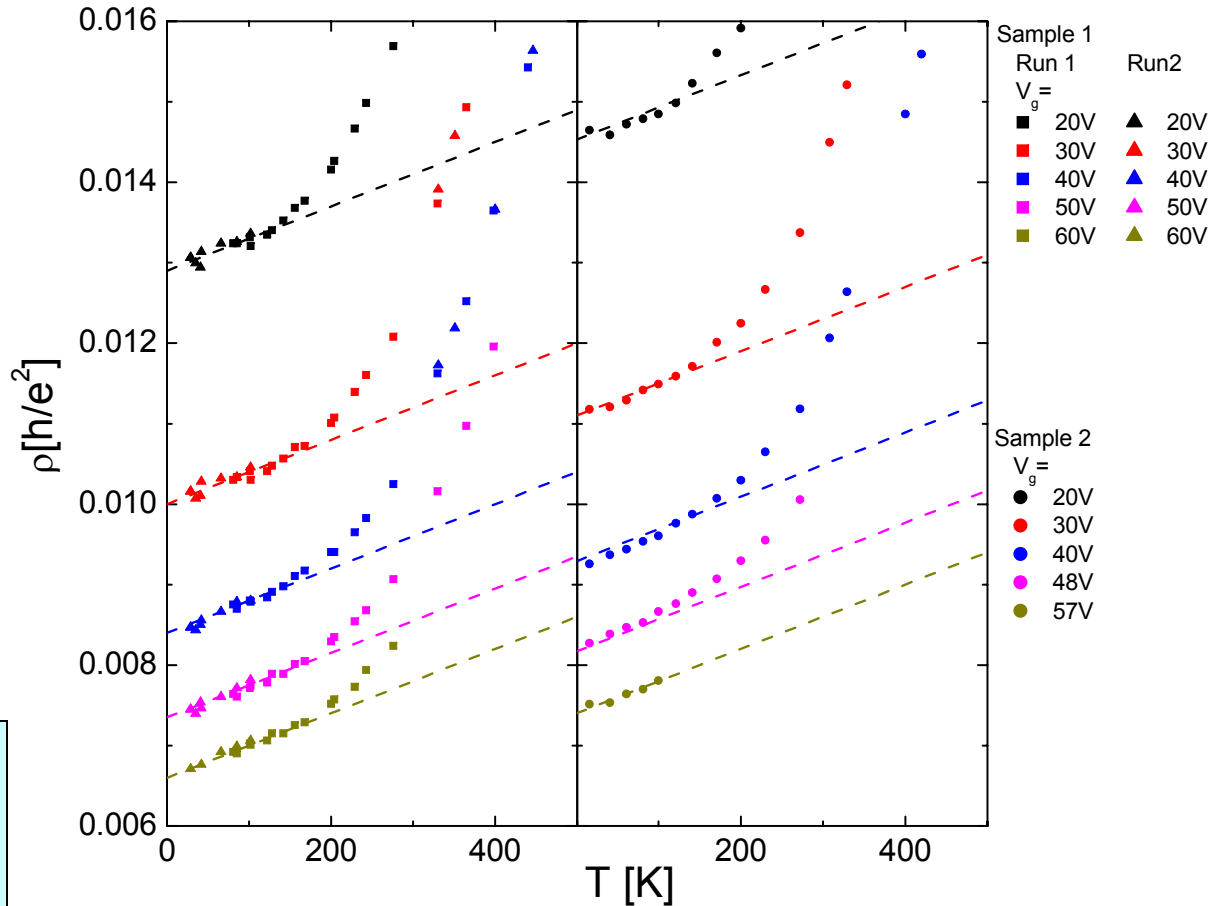
Linear T -dependence
at low T

Longitudinal acoustic
phonons in graphene

$$\rho = \rho_0 + AT$$

$$A = 0.1 \text{ } \Omega/\text{K}$$

A is independent of
charge carrier density, as
predicted



$$\rho_A(T) = \left(\frac{h}{e^2} \right) \frac{D_A^2 k_B T}{16 e^2 \hbar^2 \rho_s v_s^2 v_F^2}$$

$$\rightarrow D_A = 18 \pm 1 \text{ eV}$$

(good agreement w/CNT, graphite)

Electron-Phonon Scattering

J. H. Chen, et al. *Nature Nanotechnology* 3, 206 (2008)

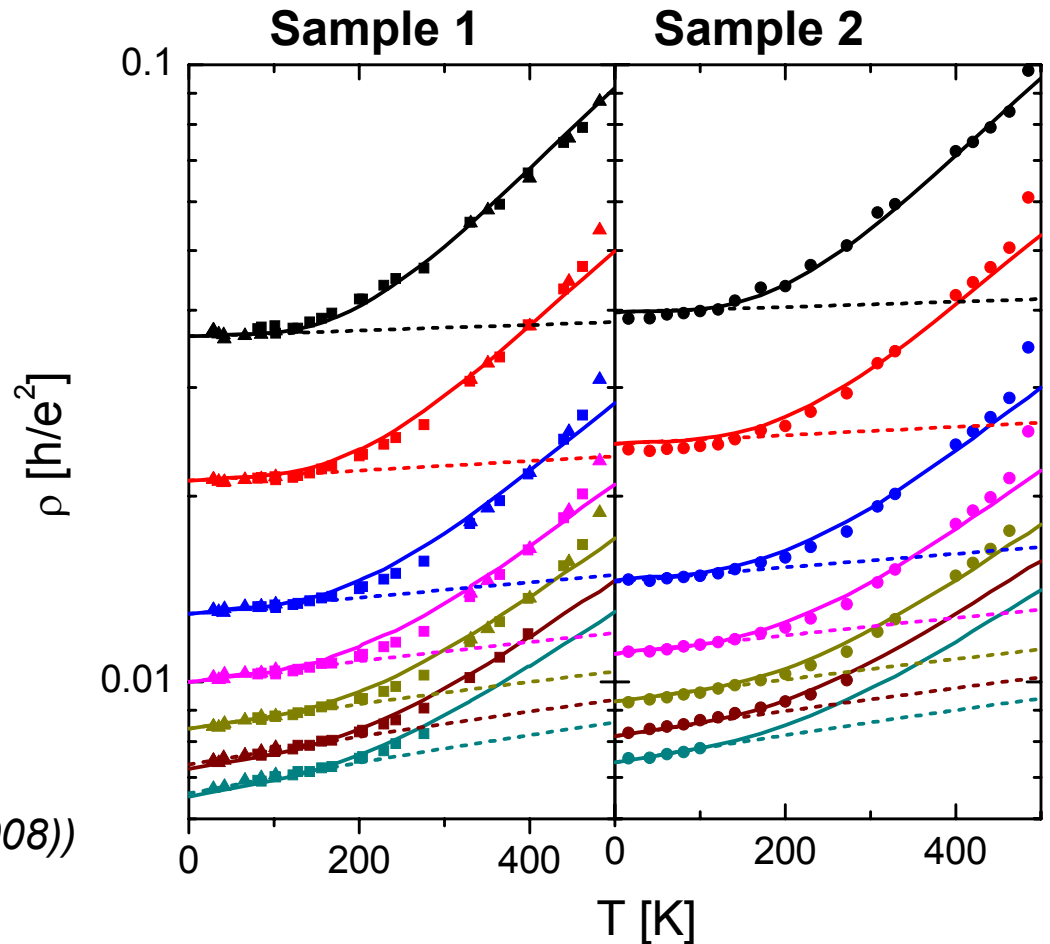
Activated T -dependence at high T

Consistent with:

$$\rho(n, T) = \rho_0 + AT + Bn^{-\alpha} \left(\frac{1}{e^{(59\text{meV})/k_B T} - 1} + \frac{6.5}{e^{(155\text{meV})/k_B T} - 1} \right)$$

LA phonons in graphene

polar optical surface phonons in SiO₂
(see Fratini and Guinea, *PRB* 77, 195415 (2008))



(3 global fit parameters for all curves)

Potential due to polar optical phonons is long-ranged; leads to density-dependent resistivity

Mobility Limits in Graphene

J. H. Chen, et al. *Nature Nanotechnology* 3, 206 (2008)

Room Temperature Limits:

Currently:

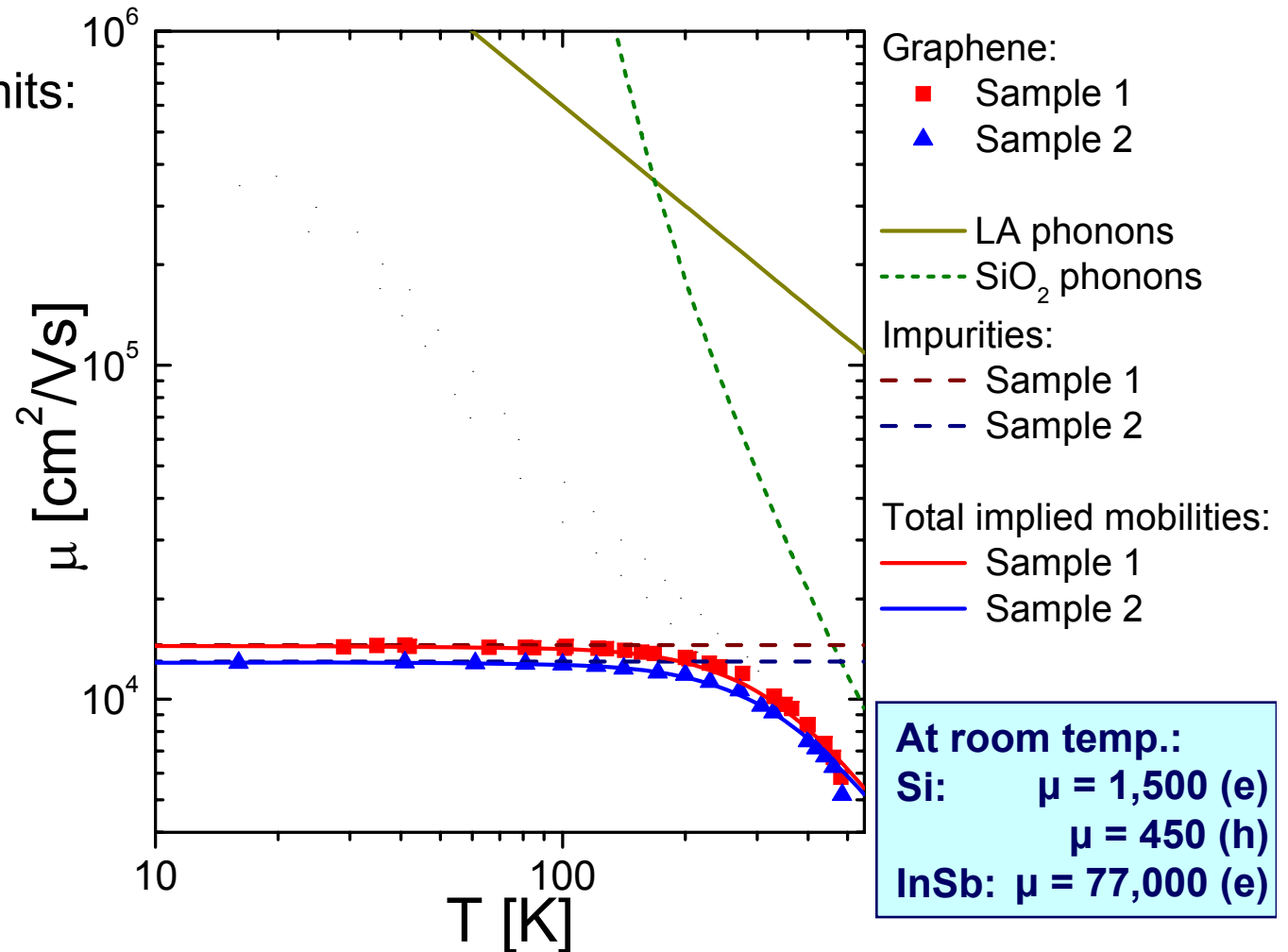
$\mu_{RT} \sim 10,000 \text{ cm}^2/\text{Vs}$
(charged impurities)

Substrate-limited:

SiO₂ surface phonons:
 $\mu_{RT} \sim 40,000 \text{ cm}^2/\text{Vs}$

Intrinsic:

acoustic phonons:
 $\mu_{RT} \sim 200,000 \text{ cm}^2/\text{Vs}$
@ $n = 10^{12} \text{ cm}^{-2}$



Room temperature mobility of **200,000 cm²/Vs** possible!

Ballistic transport over >2 microns

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r_s in graphene?

$$r_s = \frac{a}{a_b} = \frac{m^* e^2}{4\pi^{3/2} \epsilon \hbar^2 n^{1/2}}$$

Problem: what is mass?
(no characteristic length a_b for massless particles)

There still exists a unitless quantity:

$$r_s = \frac{U}{K}$$

U = potential energy of two electrons at distance a

K = kinetic energy of electron with wavelength $\lambda = 2\pi a$

Massive particles:

$$r_s = \frac{U}{K} = \frac{a}{a_b}$$

$$U = \frac{e^2}{4\pi\epsilon a}$$

$$K = \hbar v_F k = \frac{\hbar v_F}{a}$$



$$r_s = \frac{e^2}{4\pi\epsilon\hbar v_F}$$

Independent
of density!

Graphene's Fine Structure Constant?

$$\alpha = \frac{e^2}{4\pi\epsilon_0\hbar c} \approx \frac{1}{137.036}$$

α is fine structure constant
“coupling constant” – describes
relative strength of Coulomb
interaction

For graphene, define:

$$\alpha \equiv r_s = \frac{e^2}{4\pi\epsilon\hbar v_F}$$

Fine structure constant, with

$$c \rightarrow v_F$$

$$\epsilon_0 \rightarrow \epsilon$$

describes strength of Coulomb interaction

For:

$$v_F = 10^8 \text{ m/s} = c/300$$

$$\epsilon = 2.5\epsilon_0$$

$$\alpha \approx 1$$

Graphene is:

weakly interacting for condensed matter,
strongly interacting for relativistic Fermions

Interesting opportunities:

Atomic collapse of hypercritical nuclei: $Z_c = 1/\alpha = 137$ (difficult to achieve in nuclear physics),

Possible in graphene where $Z_c \approx 1$

Tuning the “Fine Structure Constant”

C. Jang, et al. *Physical Review Letters* **101**, 146805 (2008)

Conventional 2D electron system:

$$r_s = \frac{m^* e^2}{4\pi^{3/2} \epsilon \hbar^2 n^{1/2}}$$

Tune r_s thru density n

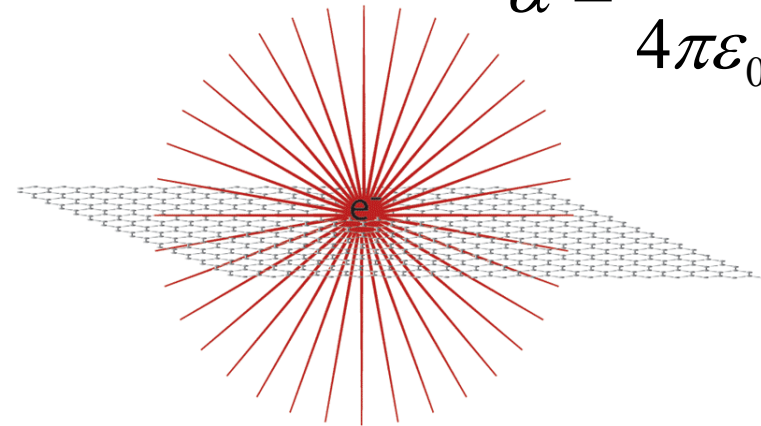
Graphene:

$$\alpha = \frac{e^2}{4\pi\epsilon\hbar v_F} = \frac{e^2}{4\pi\kappa\epsilon_0\hbar v_F}$$

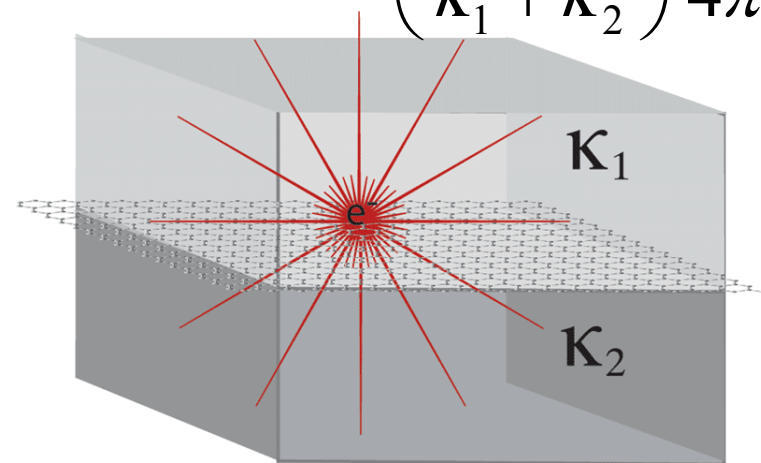
Graphene’s “Fine Structure Constant”
 α independent of n

But, can tune κ !

$$\alpha = \frac{e^2}{4\pi\epsilon_0\hbar v_F}$$



$$\alpha = \left(\frac{2}{\kappa_1 + \kappa_2} \right) \frac{e^2}{4\pi\epsilon_0\hbar v_F}$$



Two Effects of Dielectric Screening

C. Jang, et al. *Physical Review Letters* **101**, 146805 (2008)

Reducing α :

- Reduces interaction of carriers with charged impurities
 - Dominant effect for charged-impurity scattering
- Reduces screening by carriers
 - Dominant effect for short-range scattering

Within RPA:

$$\sigma_L = \frac{2e^2}{h} \frac{n}{n_{imp}} \frac{1}{F_L(\alpha)}; \quad F_L(\alpha) = \pi\alpha^2 + \dots$$

Coulomb scattering reduced:
Mobility μ_L increases

$$\sigma_S = \frac{\sigma_0}{F_S(\alpha)}; \quad F_S(\alpha) = \frac{\pi}{2} - \frac{32\alpha}{3} + \dots$$

Short-range scattering increased:
Conductivity σ_S decreases

$$\sigma_{min} = n^* e \mu_L; \quad n^* \sim \alpha^2; \quad \mu_L \sim \frac{1}{\alpha^2}$$

e - h puddle density decreased,
mobility increased:

Min. conductivity σ_{min} constant

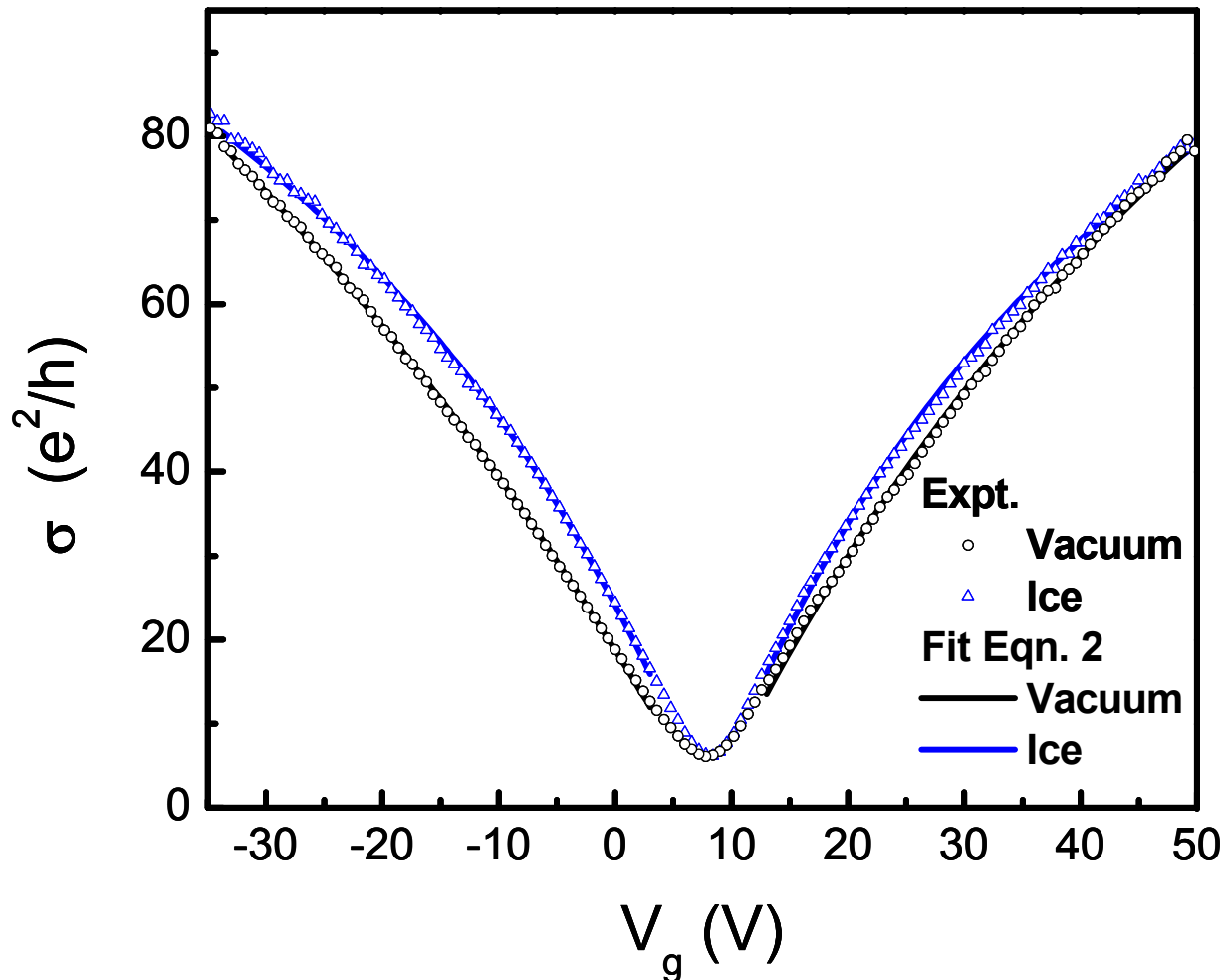
Effects of Dielectric Screening

C. Jang, et al. *Physical Review Letters* 101, 146805 (2008)

Add ice to clean graphene in UHV:

$$\alpha (\text{SiO}_2/\text{vacuum}) = 0.81 \quad \alpha (\text{SiO}_2/\text{ice}) = 0.56$$

Fit:



$$\sigma^{-1} = (ne\mu_L)^{-1} + \sigma_S^{-1}$$

Coulomb (long-range) scattering

Short-range scattering

(slight asymmetry in Coulomb scattering; take symmetric component of each)

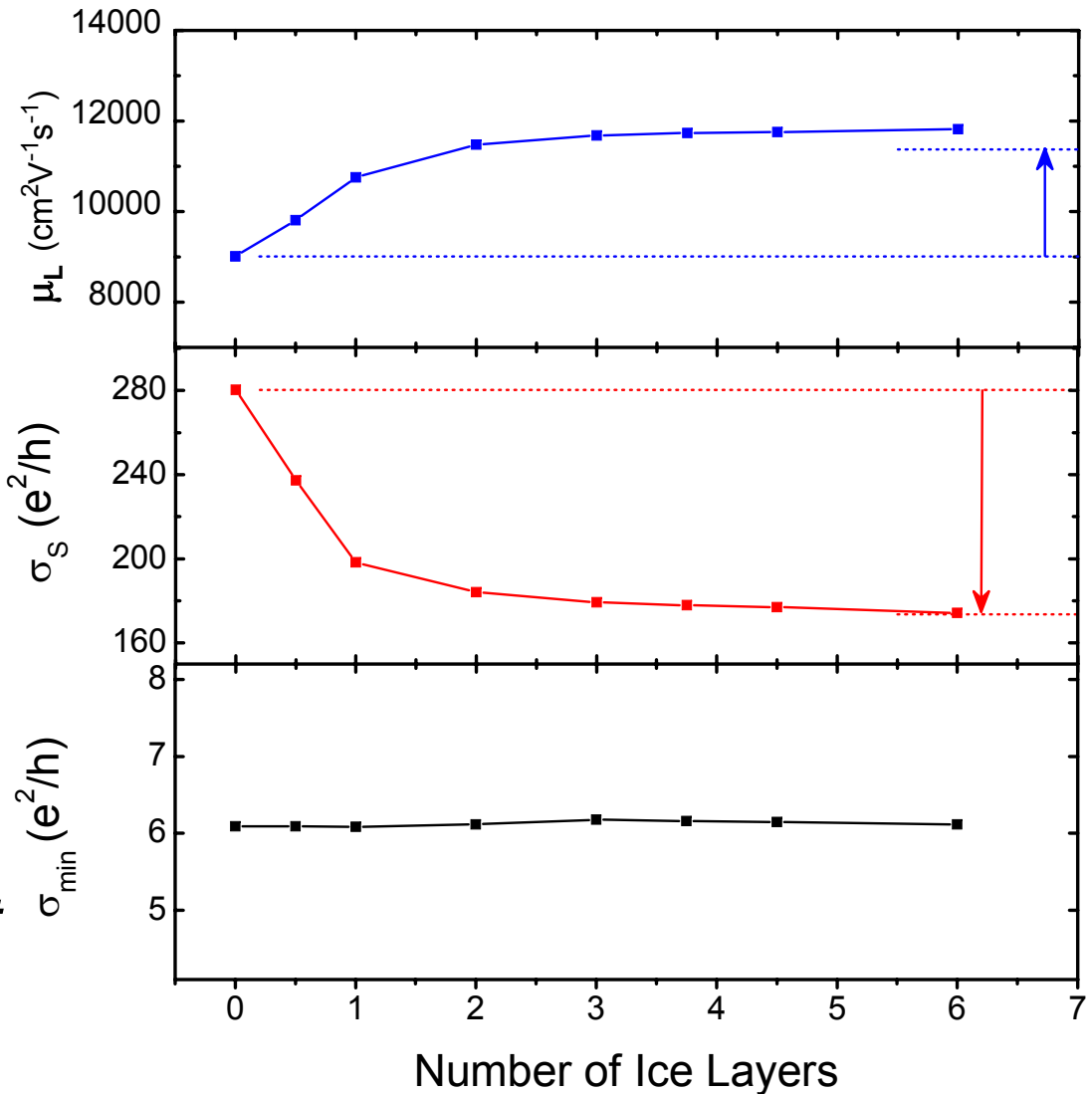
Dielectric Screening: Theory and Expt.

C. Jang, et al. *Physical Review Letters* **101**, 146805 (2008)

Coulomb scattering reduced:
Mobility μ_L increases

Short-range scattering increased:
Conductivity σ_S decreases

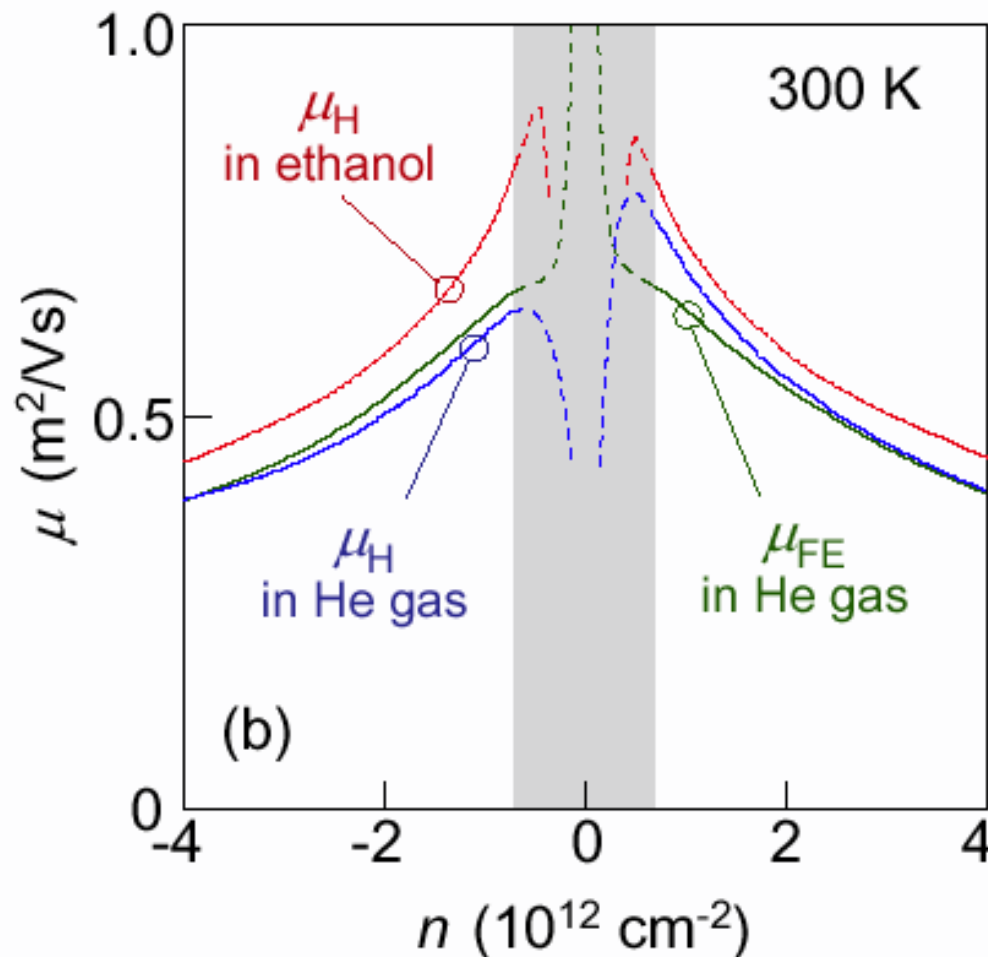
$e-h$ puddle density decreased,
mobility increased:
Min. conductivity σ_{\min} constant



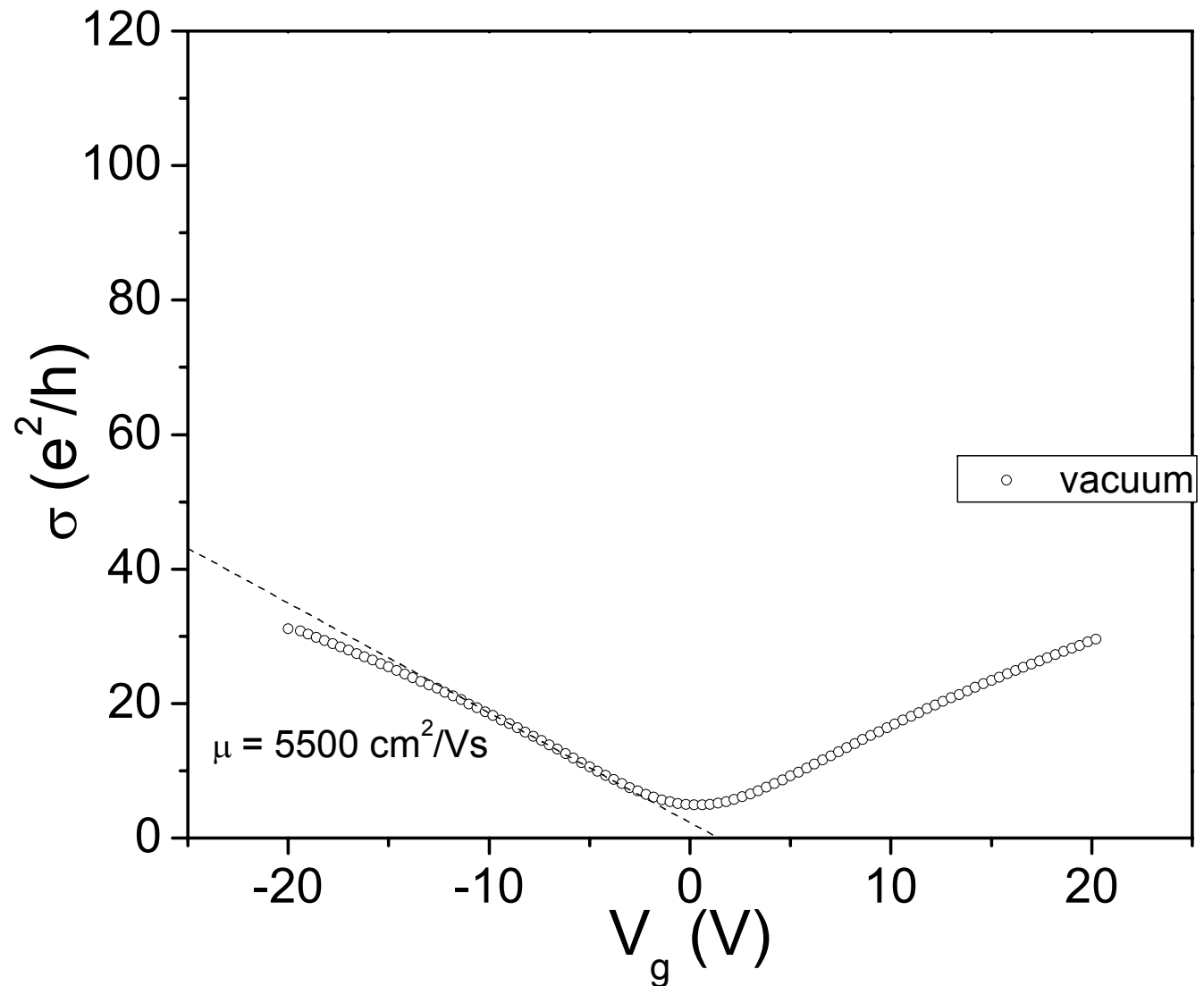
Graphene in high-K liquids – a mystery?

T. M. Mohiuddin et al. Arxiv:0809.1162 (Manchester group)

- Mobility increases <50% in ethanol ($\kappa = 25$) and liquid water ($\kappa = 80$)
- Concluded that charged impurities NOT dominant scatterers in graphene

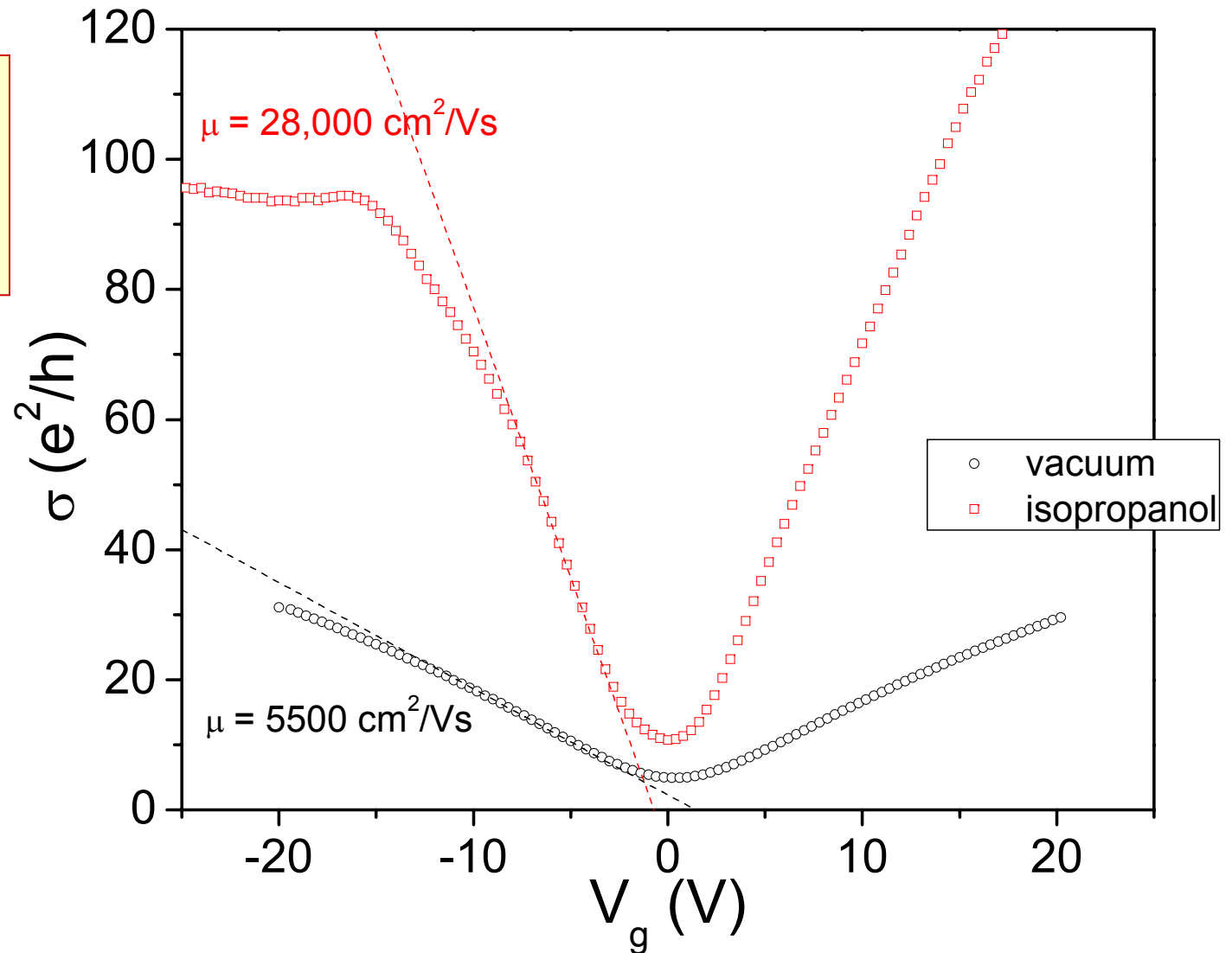


Graphene in high-K liquids – our group



Graphene in high-K liquids – our group

Isopropanol
 $\kappa = 19.9$
 $\alpha = 0.167$
Mobility up 510%

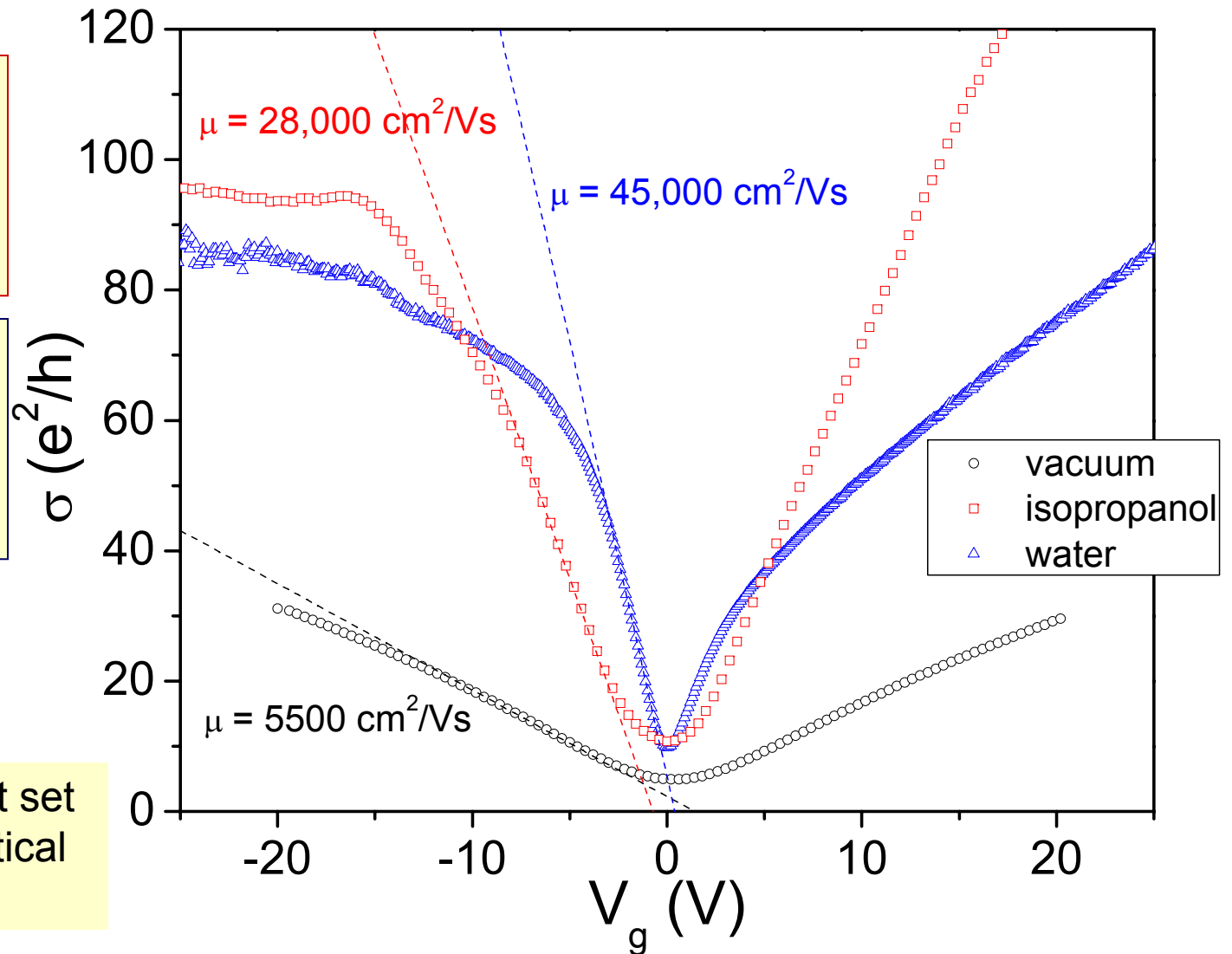


Graphene in high-K liquids – our group

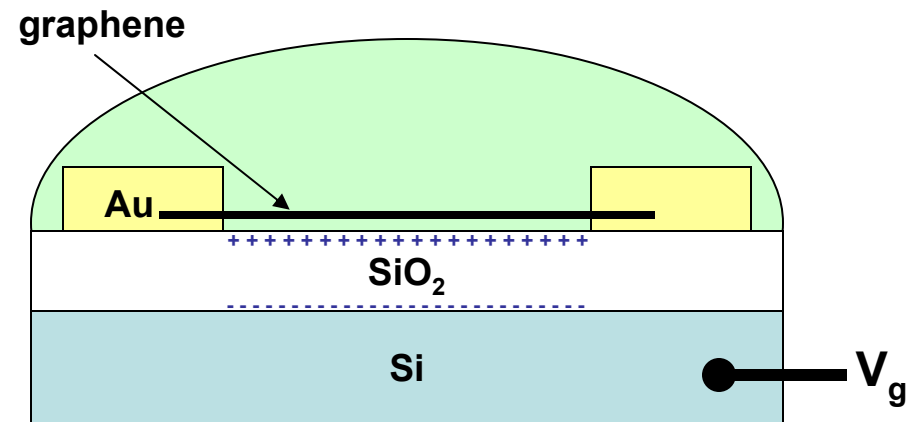
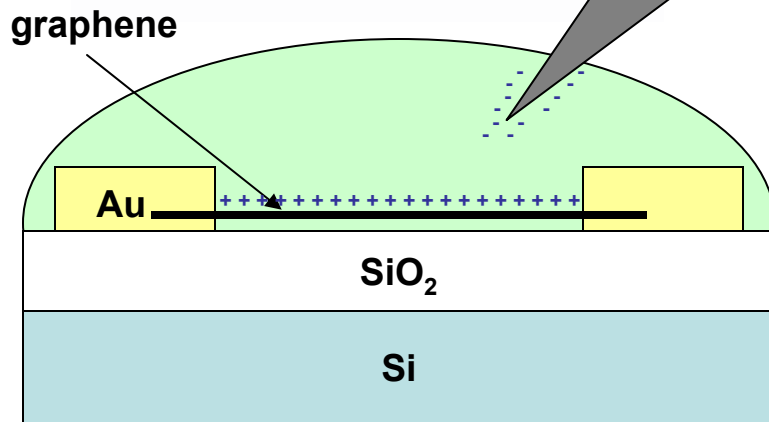
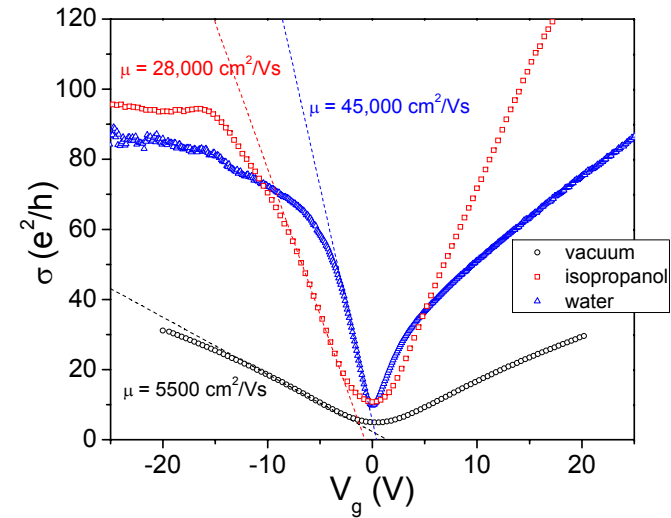
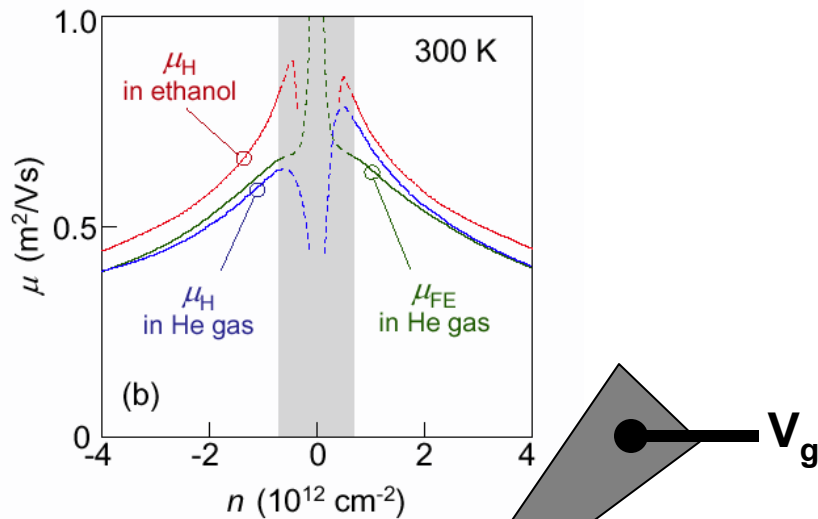
Isopropanol
 $\kappa = 19.9$
 $\alpha = 0.167$
Mobility up 510%

DI Water
 $\kappa = 80$
 $\alpha = 0.047$
Mobility up 820%

Possibly reaching limit set
by substrate polar optical
phonons



What is the difference?



Mohiuddin et al. :

- Used electrolyte as gate
- Gate charges are ions \rightarrow scatterers!

Our work :

- Used SiO_2 back-gate
- No add'l scatterers

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Graphene Corrugation - Scattering

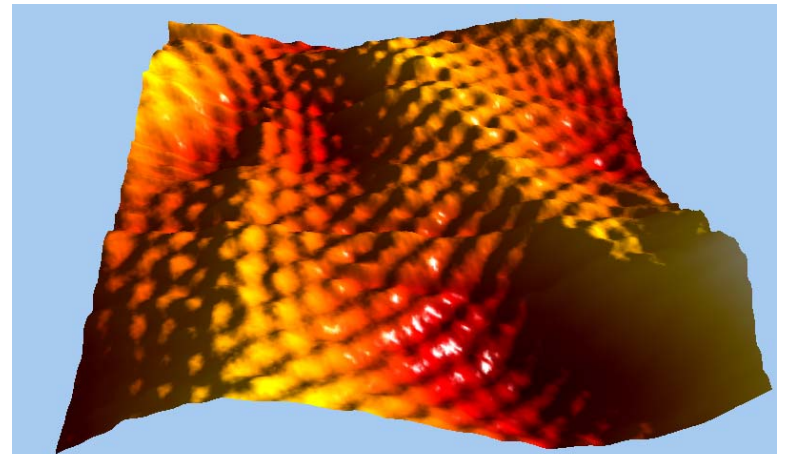
$$\sigma = \frac{2e^2}{h} E_F \tau \quad \frac{1}{\tau} \propto \frac{2\pi}{\hbar} \left| \langle k | V | k' \rangle \right|^2 D(E) \quad \begin{array}{l} q\text{-dependent interaction} \\ \rightarrow \text{carrier-density dependent } \sigma(n) \end{array}$$

1) Coulomb interaction: $q = |\mathbf{k} - \mathbf{k}'| \sim k_F \implies \sigma \sim n$

2) Corrugated graphene[†]: $\underbrace{\langle [h(r) - h(0)]^2 \rangle}_{\text{height-height correlation function}} \propto r^{2H} \implies \sigma \sim n^{2H-1}$

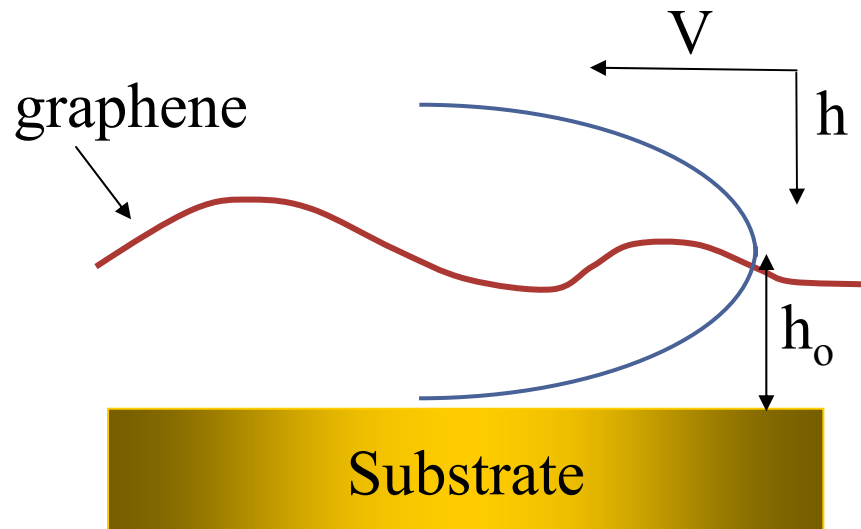
[†]Katsnelson & Geim, *Phil. Trans. R. Soc. A*
366, 195-204 (2008)

What is exponent 2H?



Graphene Corrugation

Model 1:
 Intrinsic graphene bending constrained via interface confining potential

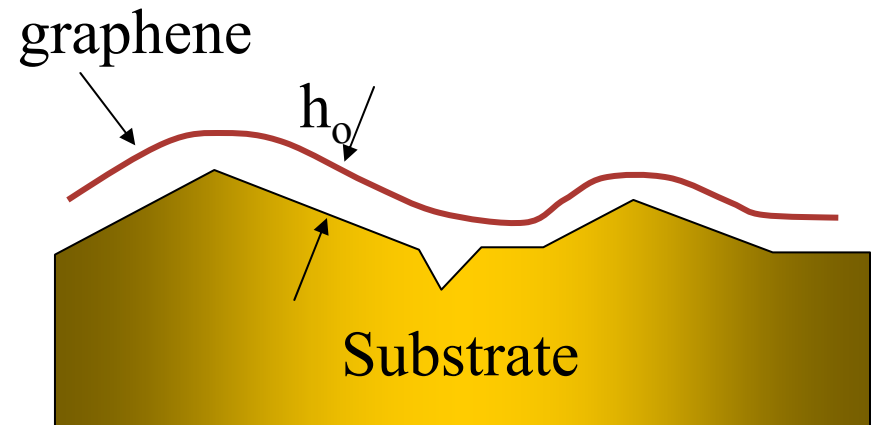


$$F = \frac{1}{2} \kappa [\nabla^2 h(x, y)]^2 + \frac{1}{2} V h^2(x, y)$$

$$\langle (h(r) - h(0))^2 \rangle \sim r^2$$

$\sigma(n) \sim n$
 (mimics Coulomb scattering)

Model 2:
 Corrugations determined by strong direct interaction governed by height variations of the substrate



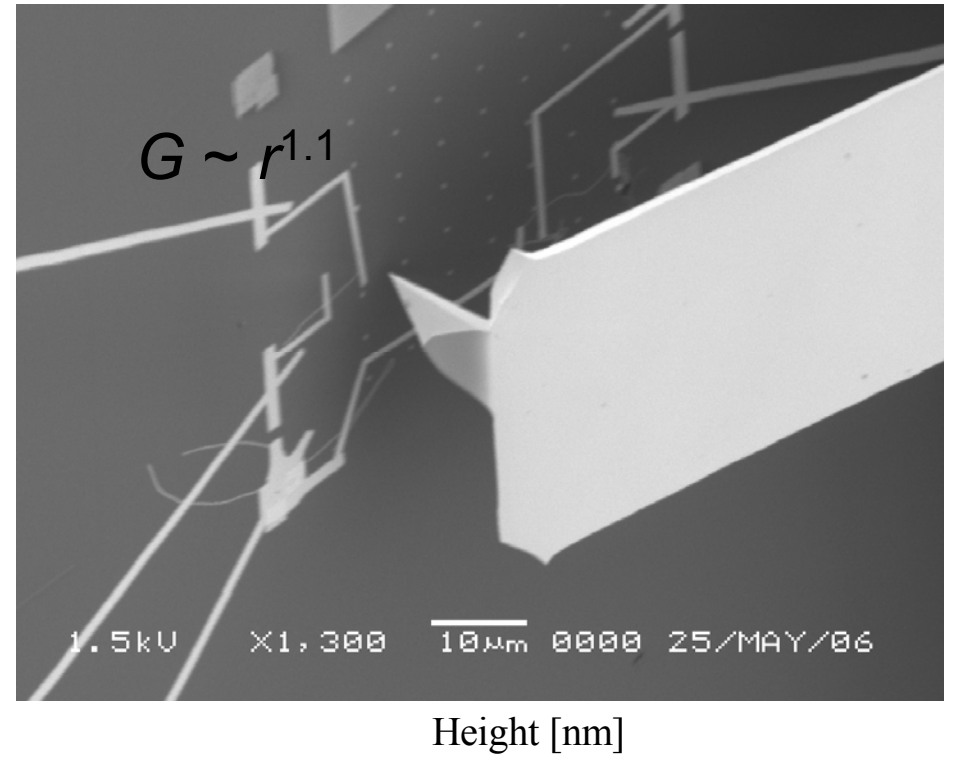
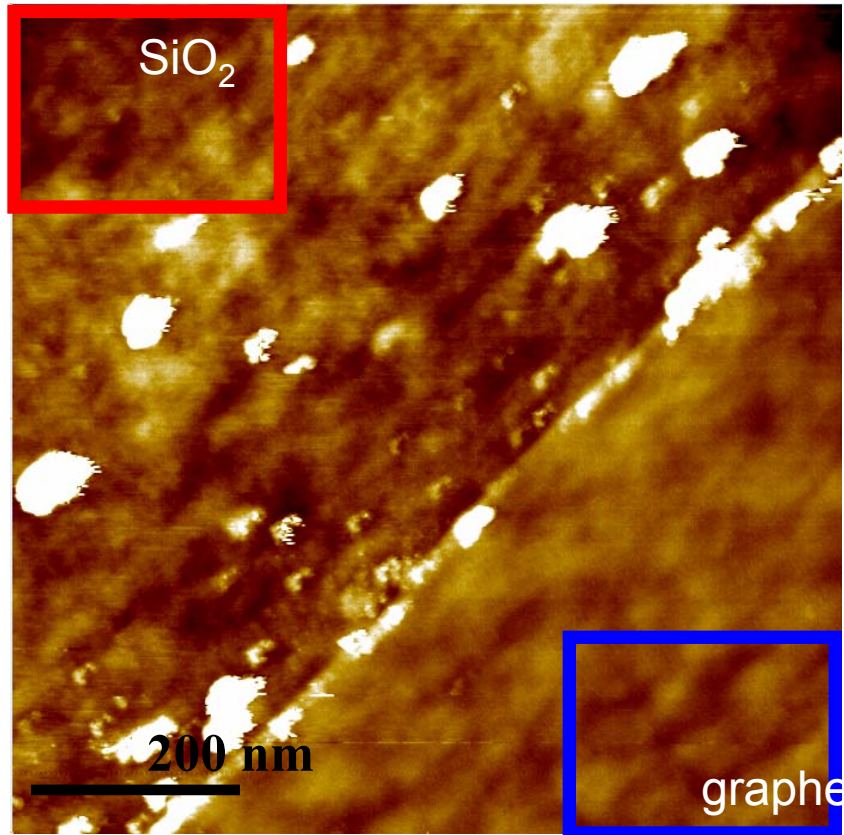
Height-height correlations will match those of the substrate.

Typical non-equilibrium surfaces show:
 $\langle (h(r) - h(0))^2 \rangle \sim r^{2H}$
 with $2H \approx 1$.

$\sigma(n) \sim \text{constant}$
 (mimics short range scattering)

Graphene Corrugations on SiO₂

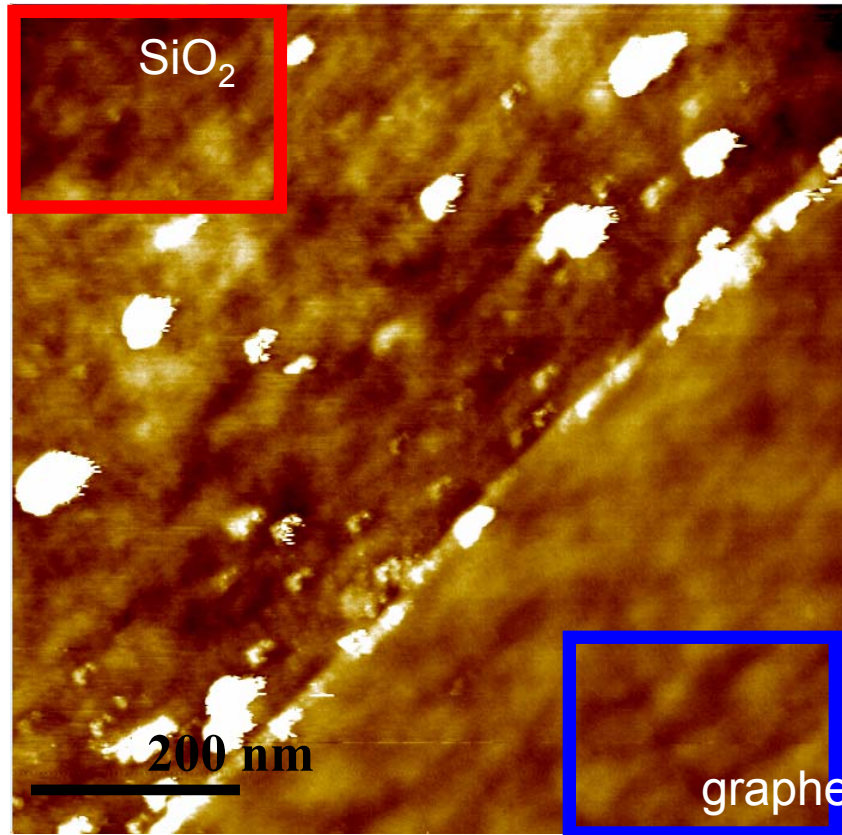
Non-contact AFM image in UHV



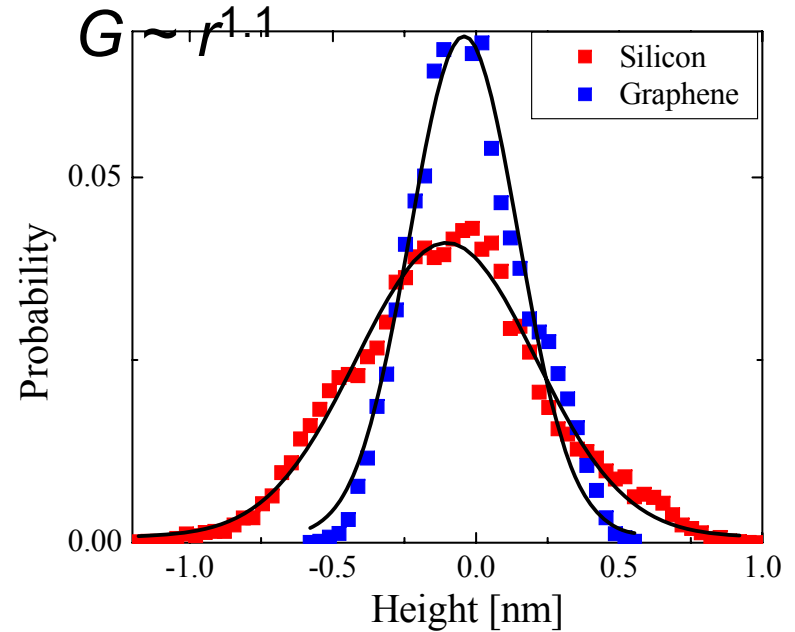
M. Ishigami et al., *Nano Letters* 7, 1643 (2007)

Graphene Corrugations on SiO₂

Non-contact AFM image in UHV



Oxide-graphene boundary

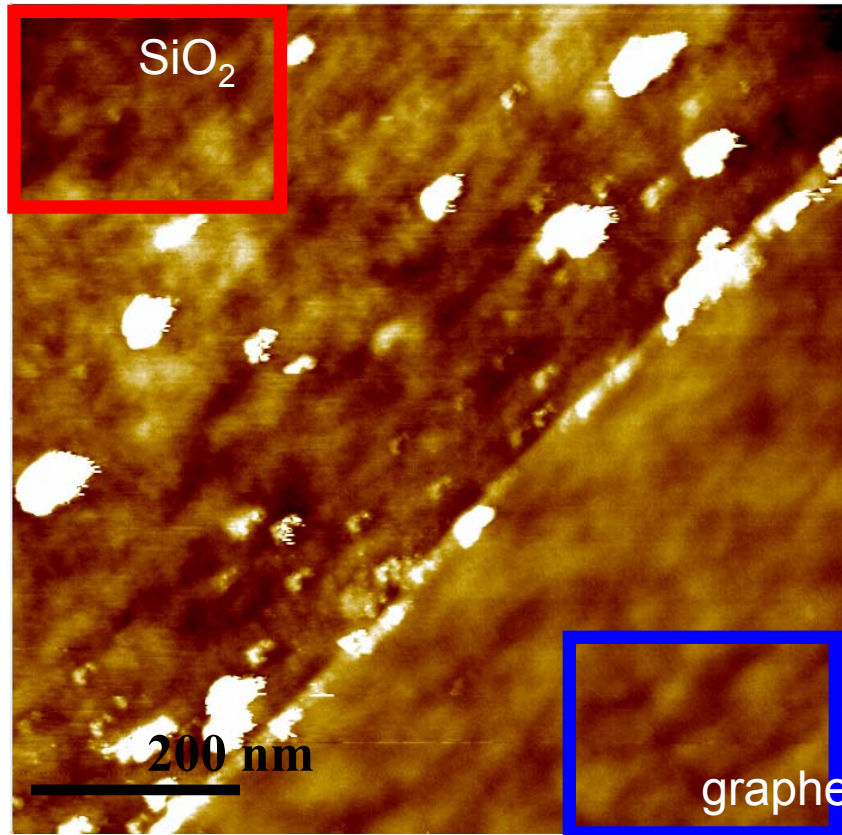


- $\sigma_{\text{oxide}} = 3.1 \text{ \AA}$ and $\sigma_{\text{graphene}} = 1.9 \text{ \AA}$
- Graphene 60% smoother than SiO₂

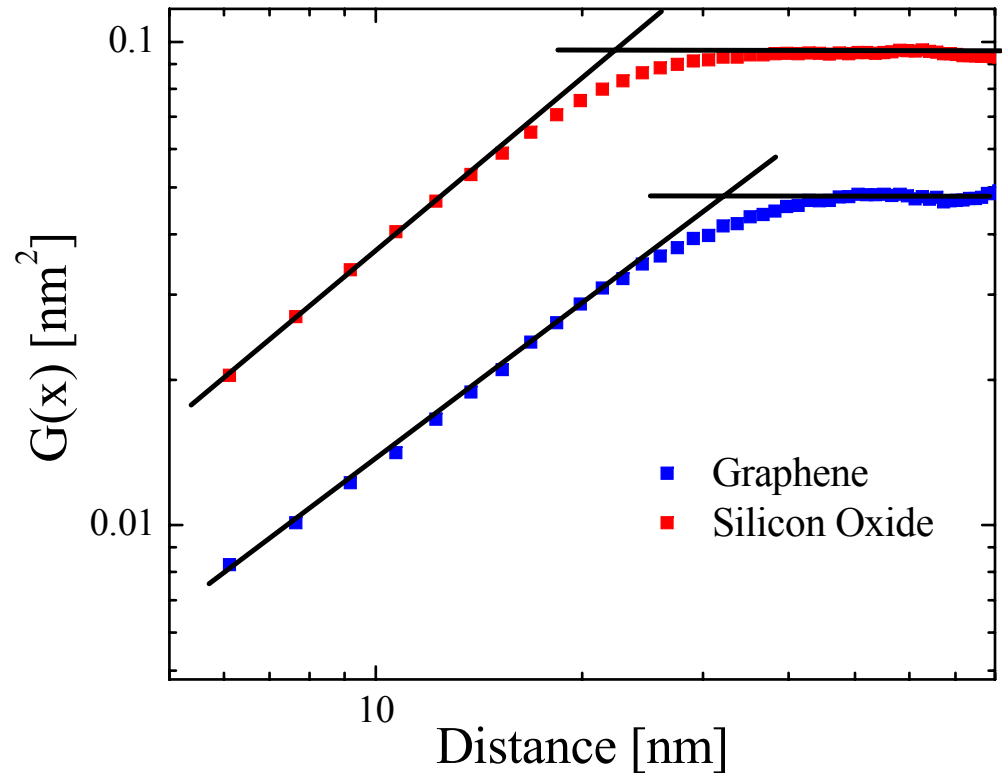
M. Ishigami et al., *Nano Letters* 7, 1643 (2007)

Graphene Corrugations on SiO₂

Non-contact AFM image in UHV



Oxide-graphene boundary



Height-height correlations function

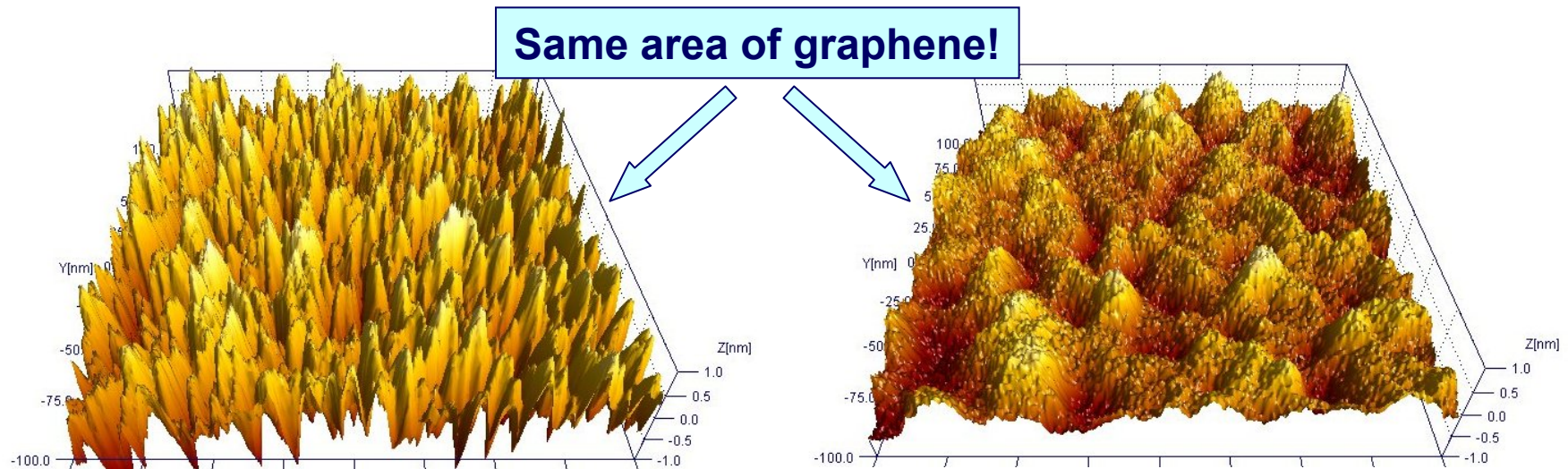
$$\langle (h(r) - h(0))^2 \rangle \sim r^{2H}$$

with $2H \approx 1$

$\sigma(n) \sim \text{constant}$
(mimics short range scattering)

M. Ishigami et al., *Nano Letters* 7, 1643 (2007)

STM vs. NC-AFM topography



STM: 1V, ~50 pA

Similar to Morgenstern group (preprint)

NC-AFM: 4.6 Hz Δf

Reproduces our earlier work
Ishigami, et al. Nano Letters 7, 1643 (2007)

- Both images acquired from same area, on 1-layer graphene device.
- Why does STM measure topography so differently?
- **STM more strongly interacting – electro-mechanical effect**

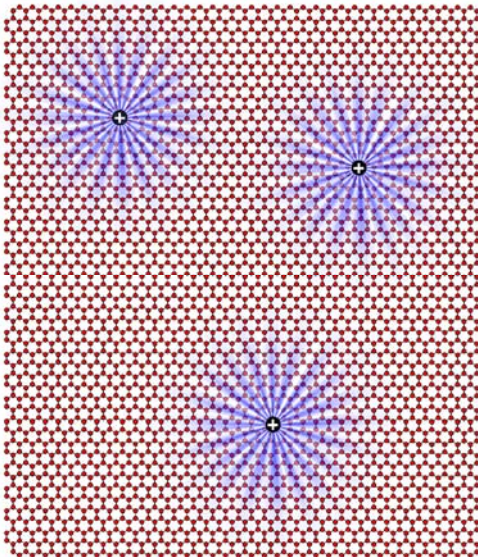
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Lattice defect scattering in graphene

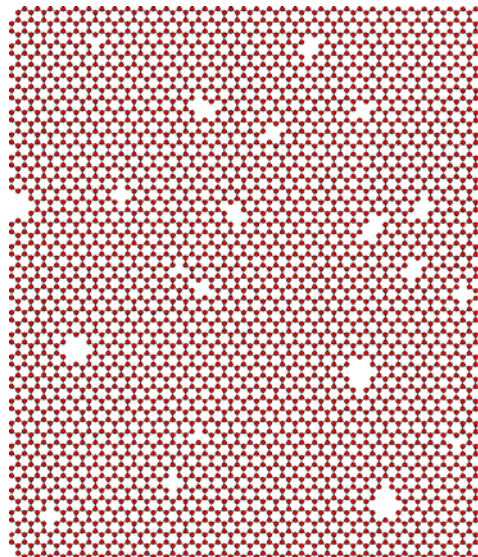
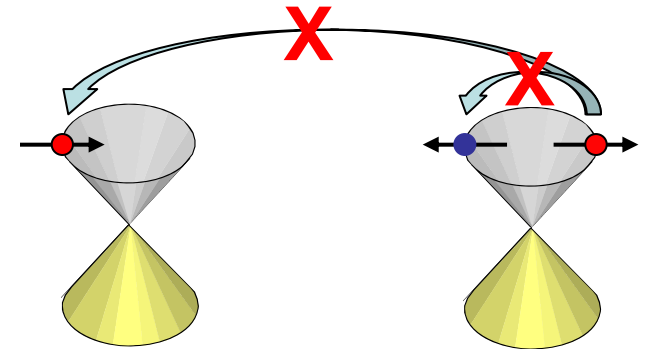


Charged-impurity scattering:

Linear $\sigma(V_g)$

Intravalley scattering
No backscattering
weak anti-localization

Metallic

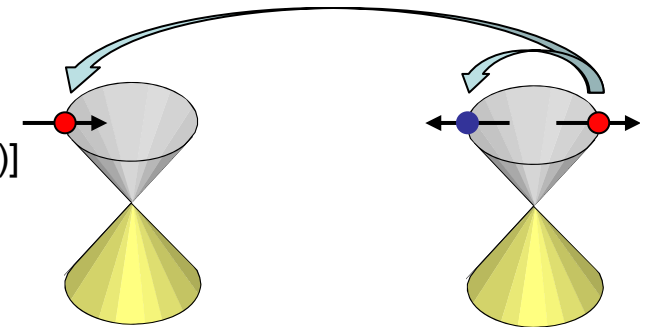


Defect scattering:

Constant $\sigma(V_g)$? [Shon, & Ando, (1998)]
Linear $\sigma(V_g)$? [Hentschel (2007);
Stauber (2007)]

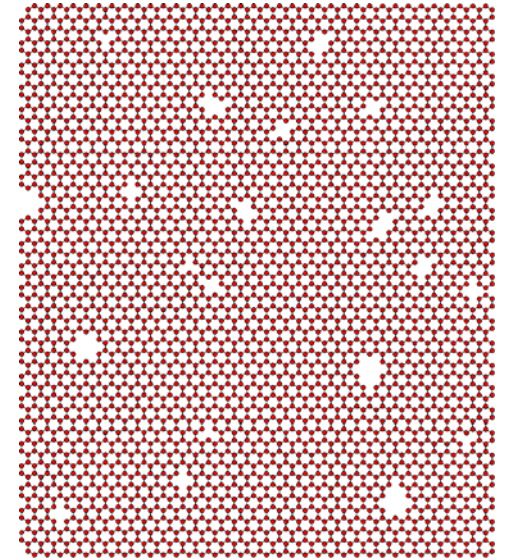
Expect *intravalley* and *intervalley* scattering
Backscattering allowed
weak anti-localization or *weak localization*?

Metallic or *insulating*?



Inducing lattice defects in graphene

- Sample is cleaned in H₂/Ar at 300 °C
[Ishigami, *Nano Letters* 7, 1643 (2007)]
- Sample baked in UHV at 220 °C overnight
- Ne⁺ or He⁺ ion irradiation at 500 eV via sputter gun
- Dose given by current collected by Faraday cup
- Sample annealed at 220 °C overnight between ion irradiation runs; mobility partially recovers on annealing



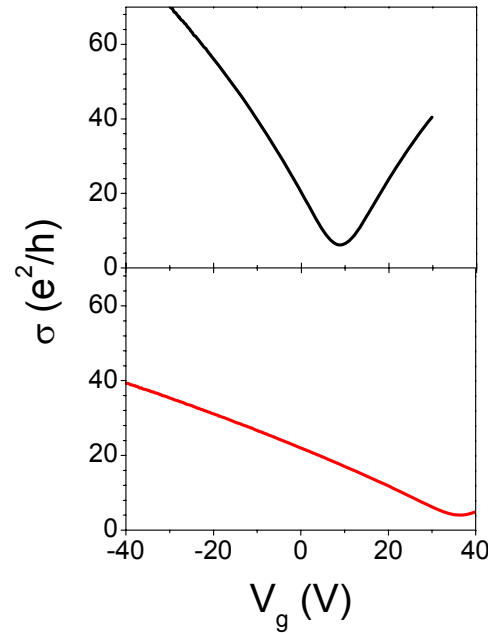
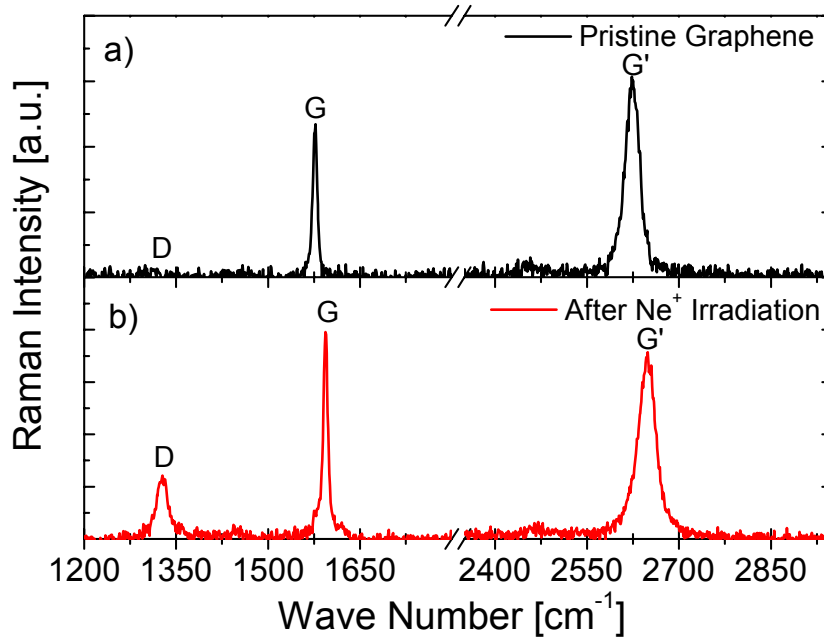
Expect:

- **One ion → one defect consisting of multi-atom vacancy**

See e.g. G. M. Shedd and P. E. Russell, *JVSTA* 9, 1261 (1991)

J. R. Hahn, et al., *PRB* 53, R1725 (1996)

Raman D peak - intervalley scattering



Mobility

$$\mu = 8600 \text{ cm}^2/\text{Vs}$$

$$\mu = 1300 \text{ cm}^2/\text{Vs}$$

Graphitic particles of grain size L_a :
$$L_a = \left[2.4 \times 10^{-10} \text{ nm}^3 \right] \lambda^4 \left(\frac{I_D}{I_G} \right)^{-1}$$

Cancado, et al.,
APL 88, 163106 (2006).

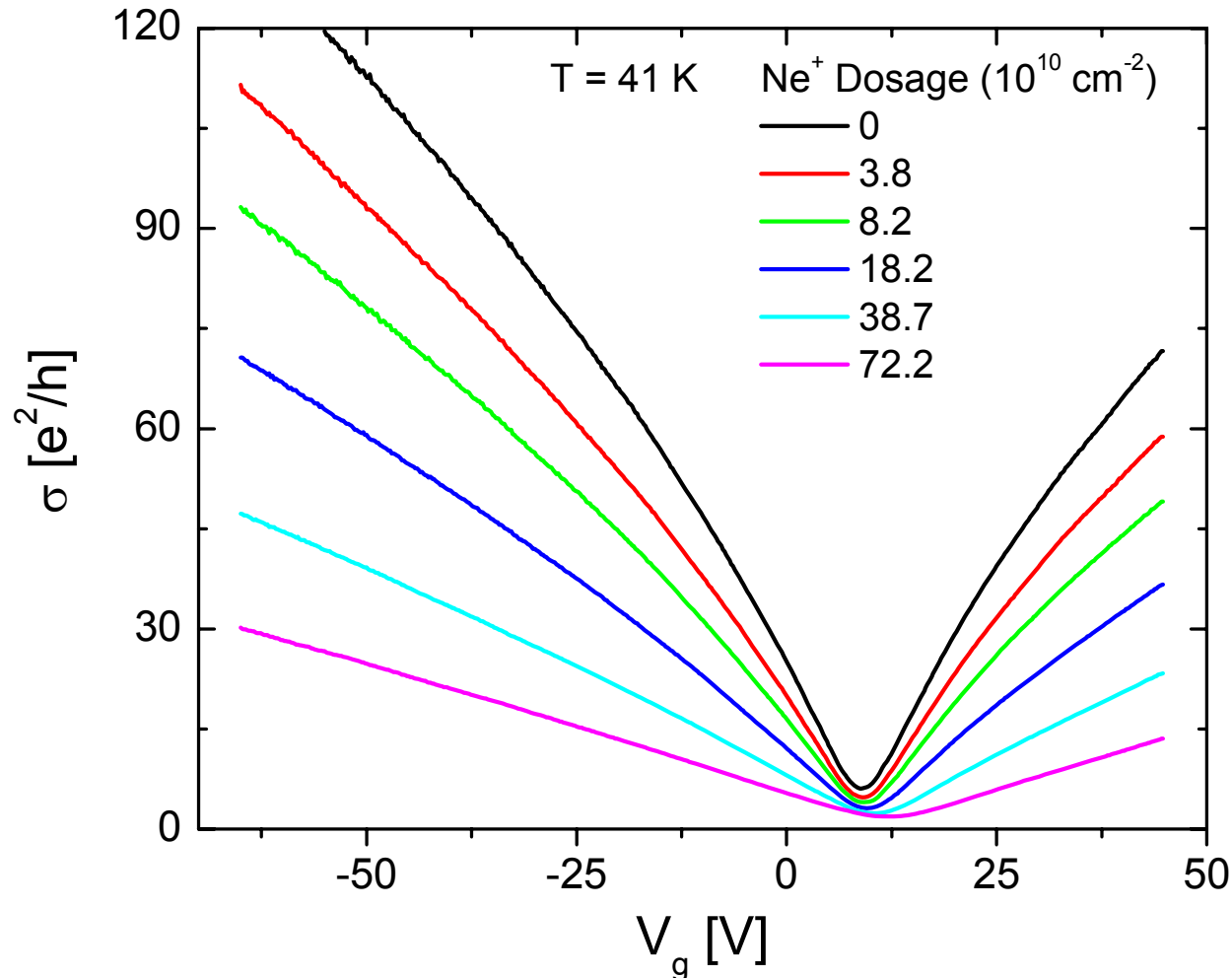
Point defects: identify L_a with defect scattering length.

Our samples: $L_a = 70 \text{ nm}$

$$\mu = 1300 \text{ cm}^2/\text{Vs}; n \approx 10^{13} \text{ cm}^{-2} \text{ (in ambient)} \rightarrow l_{\text{mfp}} \approx 50 \text{ nm}$$

Defect scattering lengths from Raman and transport agree

Defects in graphene



Fit each curve to: $\sigma^{-1} = (ne\mu)^{-1} + \rho_S$

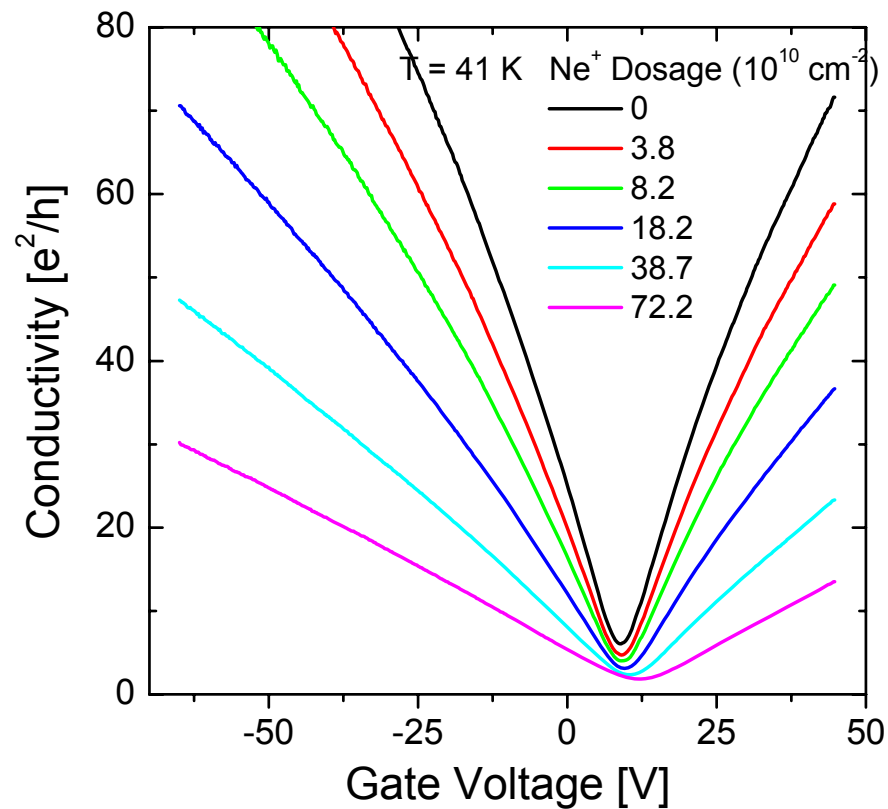
“Long-range scattering”
Constant mobility



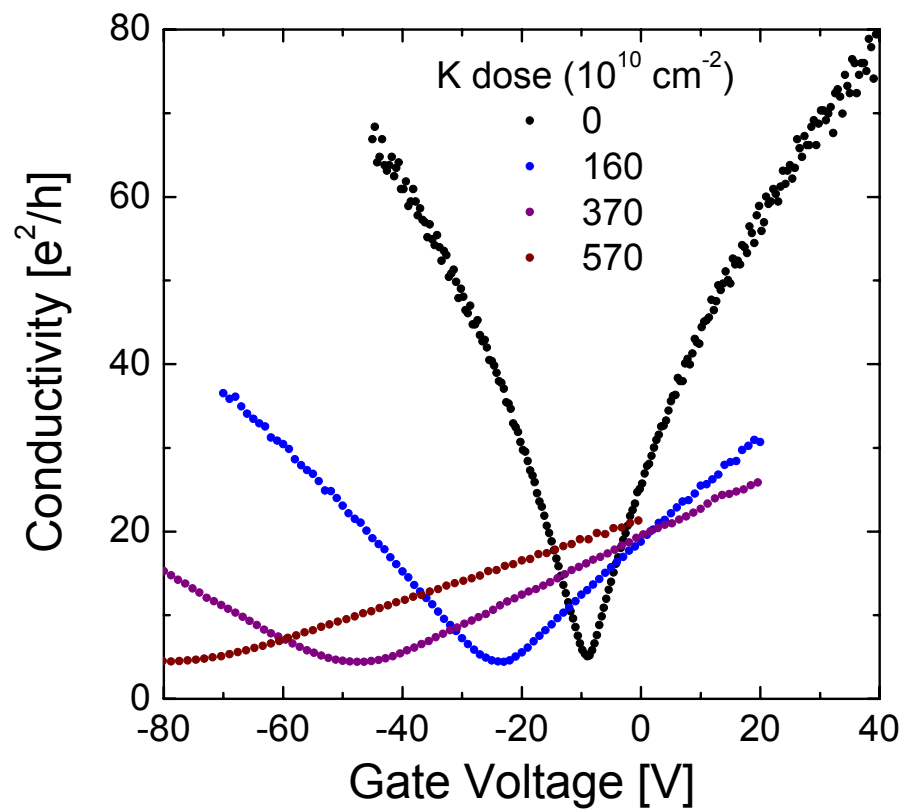
“Short-range scattering”
Constant resistivity

Defects in graphene

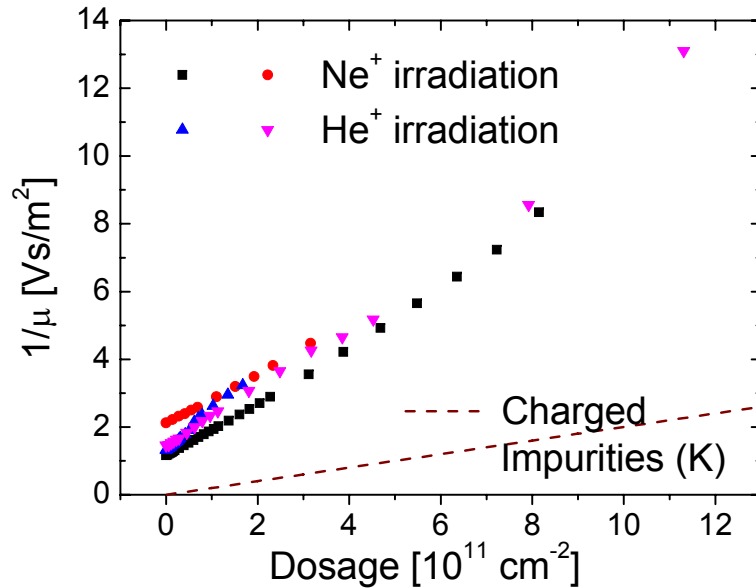
Ne⁺ irradiation (lattice defects)



K doping (charged impurities)

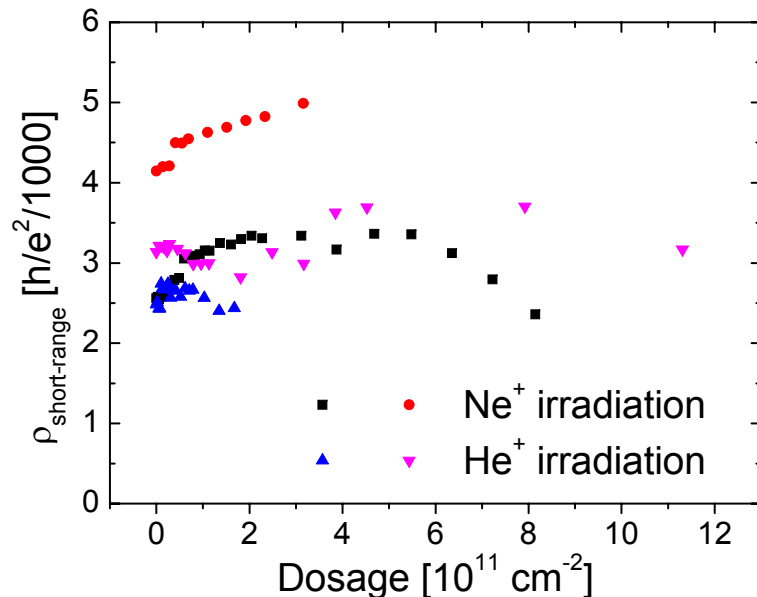


Defects in graphene



Defects:

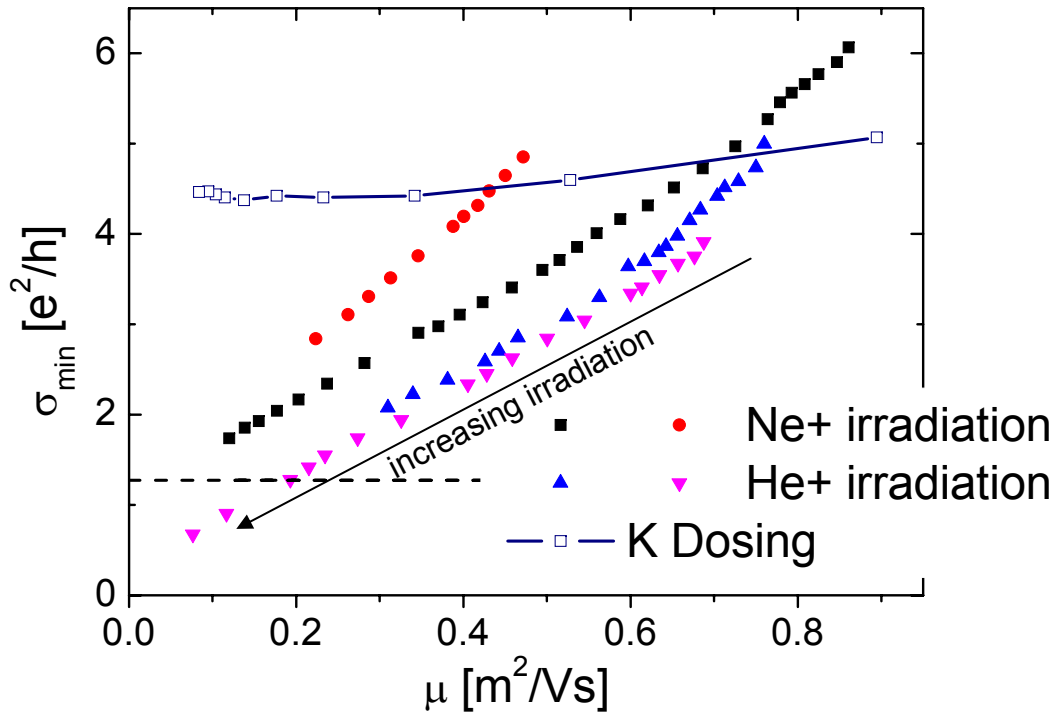
- Defects change the *linear* term in $\sigma(V_g)$
Like charged impurities!
- Linear $\sigma(V_g)$ scattering 4x stronger than for same concentration of charged impurities



Defects:

- Carrier-density-independent ρ_s scattering does *not* change
- ρ_s corresponds to $l_{\text{mfp}} \sim 2$ microns
 $\rightarrow \rho_s$ *cannot* be the scattering seen in Raman D band

Defects in graphene – Minimum conductivity



Minimum conductivity: $\sigma_{\min} = n^* e \mu$

n^* is carrier density in “puddles”

n^* is function of charged impurity density

Charged impurities:

n_{imp} increases: μ decreases, n^* increases
 $\rightarrow \sigma_{\min}$ changes very weakly

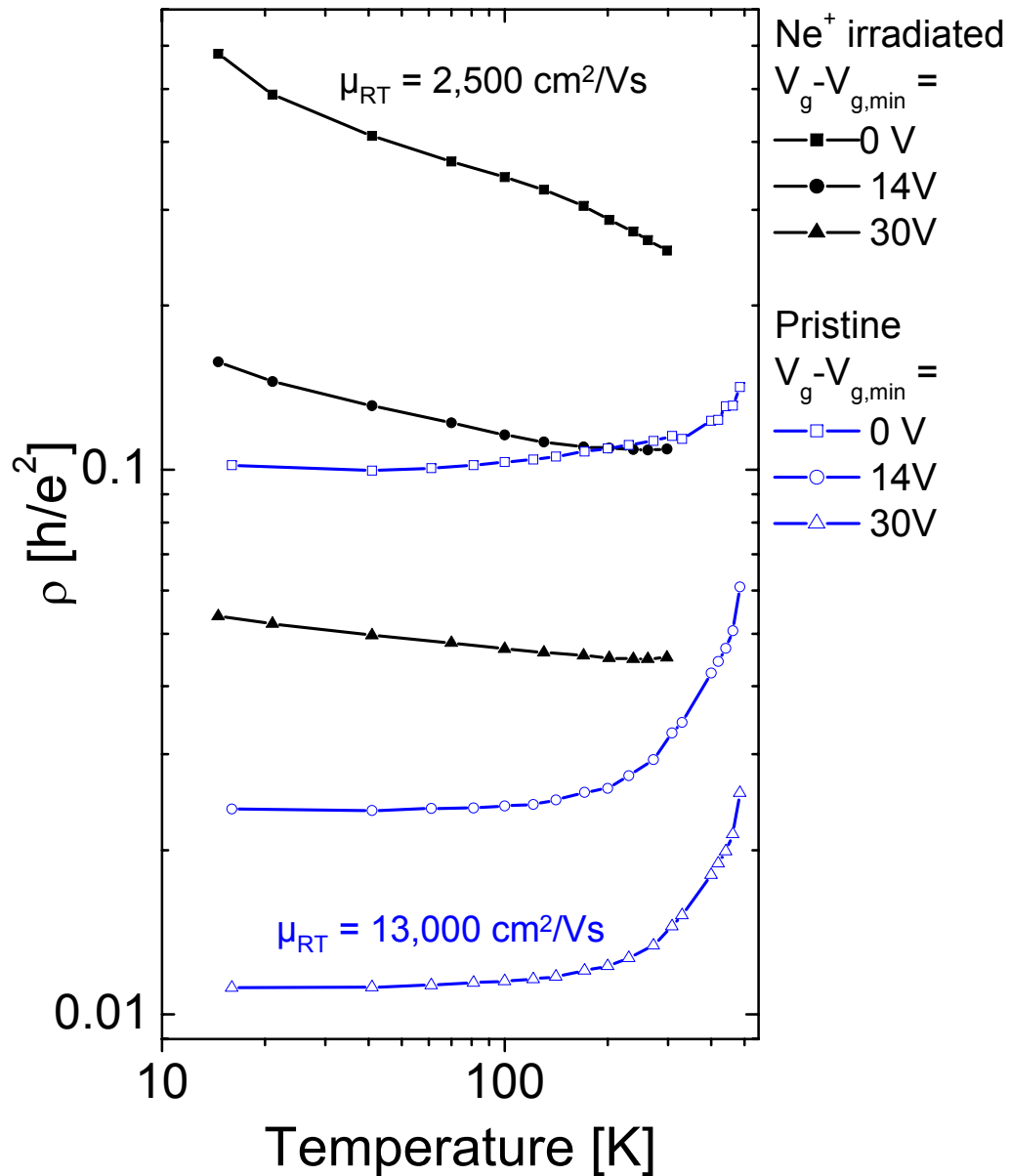
$$\sigma_{\min} = n^* e \mu \quad \begin{array}{l} n^* \uparrow \\ \mu \downarrow \end{array}$$

Defects:

n_{defect} increases: μ decreases, n^* constant
 $\rightarrow \sigma_{\min}$ proportional to μ

$$\sigma_{\min} = n^* e \mu \quad \begin{array}{l} n^* \text{ const.} \\ \mu \downarrow \end{array}$$

Defects in graphene – Metal or Insulator?



Theory:

Graphene with only *intravalley* scattering is metallic (weak anti-localization)

Graphene with *intervalley* scattering is insulator (weak localization)

[Bardarson, et al. *PRL* **99**, 106801 (2007)]

Experiment:

Graphene with charged impurities shows metallic $\rho(T)$ at low T

[Novoselov, *Nature* **438**, 197 (2005)]

[Chen, *Nature Nano* **3**, 206 (2008)]

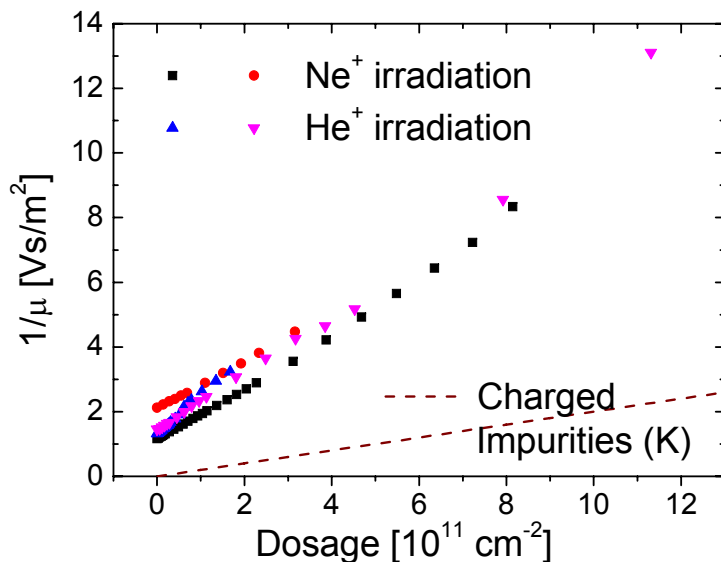
Graphene with defects shows diverging $\rho(T)$ at low T even for modest mobilities ($\sim 2,000 \text{ cm}^2/\text{Vs}$)!

Midgap states - Theory

[Hentschel & Guinea, *PRB* **76**, 115407 (2007); Stauber, Peres, & Guinea, *PRB* **76**, 205423 (2007)]

- Defect potential modeled as circular well of radius R , depth ϵ_0 , intervalley scattering Δ .
- Spectrum inside the potential well is gapped by Δ ; has bound midgap states.
- Conductivity is:

$$\sigma_d = ne\mu_d = \frac{2e^2}{h} \frac{n}{n_d} \ln^2(k_F R)$$



Experimentally, $\mu_d \approx [1.2 \times 10^{15} \text{ V}^{-1}\text{s}^{-1}]/n_d$

For $n = 2 \times 10^{12} \text{ cm}^{-2} \rightarrow R \sim 8 \text{ \AA}$

Reasonable value for 500 eV irradiation
(multiple-atom vacancies)

$\ln^2(k_F R)$ dependence *not* observed, but k_F only varies by factor of ~ 3 outside puddle regime ($n > n^*$)

Conclusions

- **Mobility of graphene on SiO₂ limited by charged impurities**
 - Charged impurities give linear $\sigma(V_g)$
 - Minimum conductivity determined by density in $e-h$ puddles
 - Addition of dielectric layer increases mobility
- **Room temperature intrinsic mobility $\sim 200,000$ cm²/Vs**
 - Remote interfacial phonon scattering from SiO₂ limits to $\sim 40,000$ cm²/Vs
- **Corrugations**
 - Graphene corrugations follow SiO₂ substrate roughness
- **Graphene with lattice defects**
 - Linear $\sigma(V_g)$ with 4x lower mobility compared to charged impurities
 - Consistent with midgap states, $R = 2-3$ Å
 - Depressed $\sigma_{\min} \sim \mu$; can be less than $4e^2/\pi h$
 - Intervalley scattering gives insulating $\rho(T)$; Raman D band

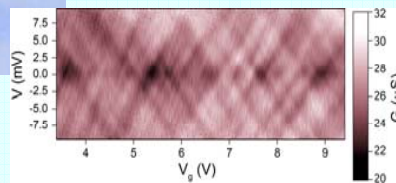
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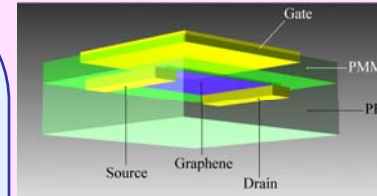
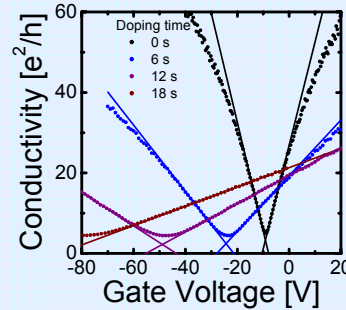


MR in graphene

*graphene
Fabry-Pérot*

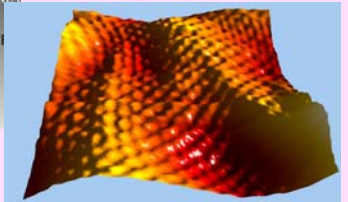


*UHV
doping,
dielectric expts.*



transfer-printing

STM



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Theory

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Funding:

