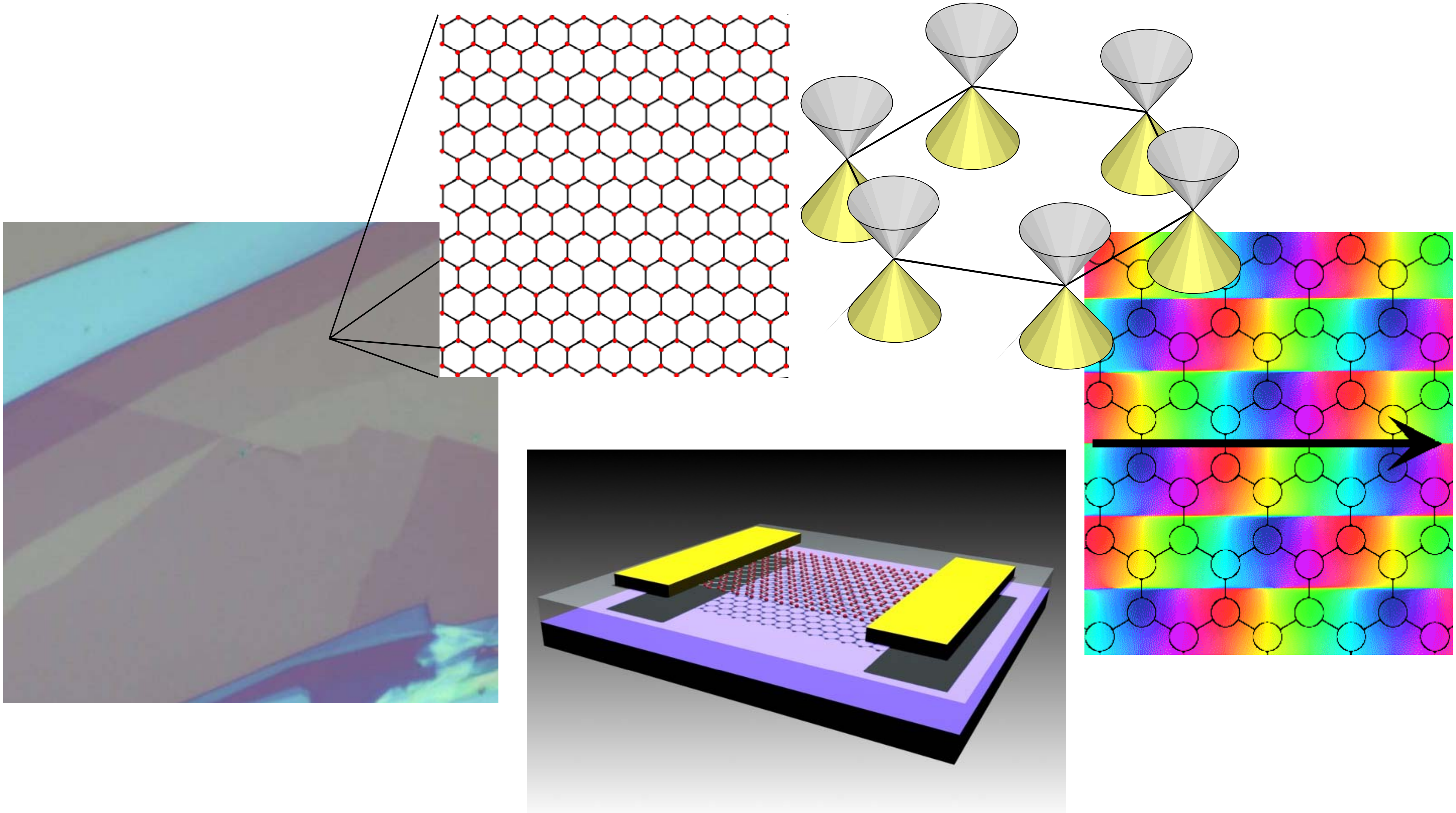
Graphene: Scratching the Surface



Michael S. Fuhrer
Director, Center for Nanophysics and Advanced Materials
University of Maryland, USA

I. Transport/scanned probe expts. on graphene in UHV

Correlated charged impurities [*PRL* **107**, 206601 (2011)]

Imaging charge disorder of bare SiO_2 [in preparation]

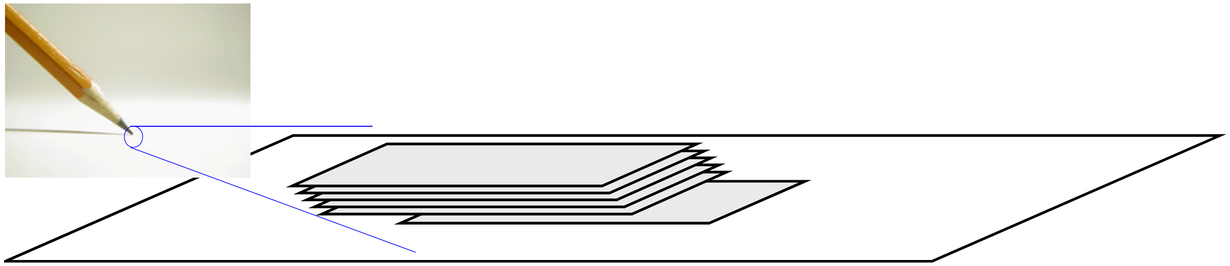
II. Graphene devices

Bandgapped bilayer graphene [*Nano Lett.* **10**, 4521 (2010)]

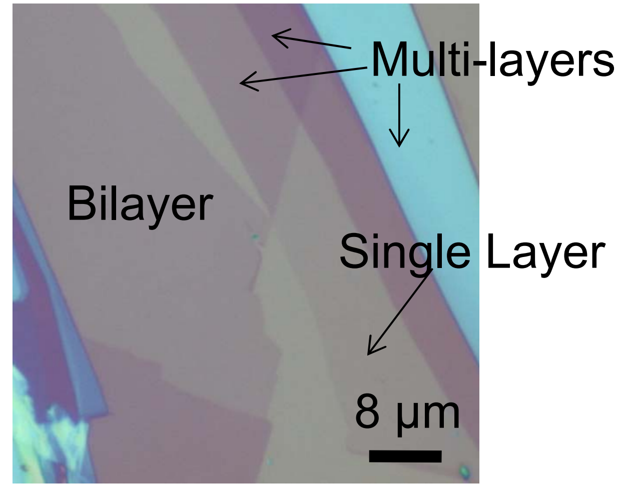
A bilayer graphene hot electron bolometer [ArXiv:1111.1202]

Graphene Devices – Fabrication

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

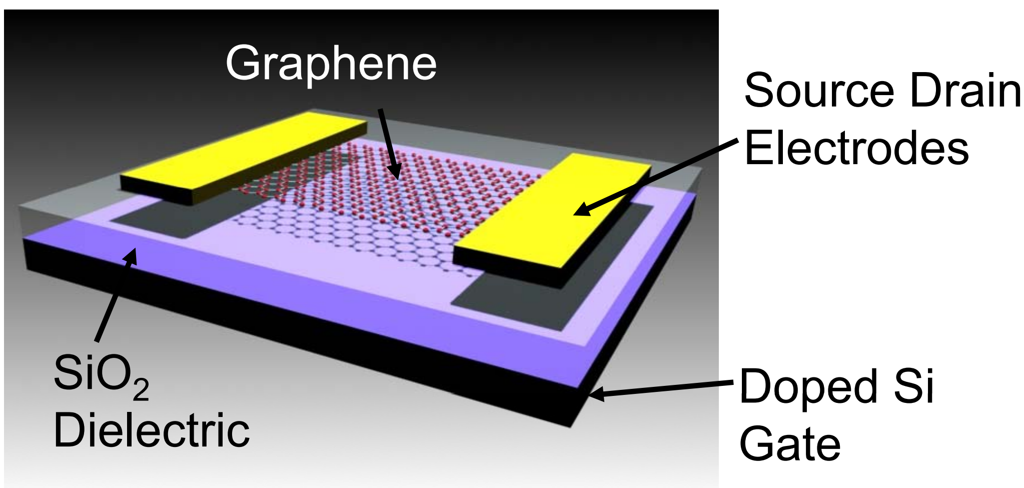


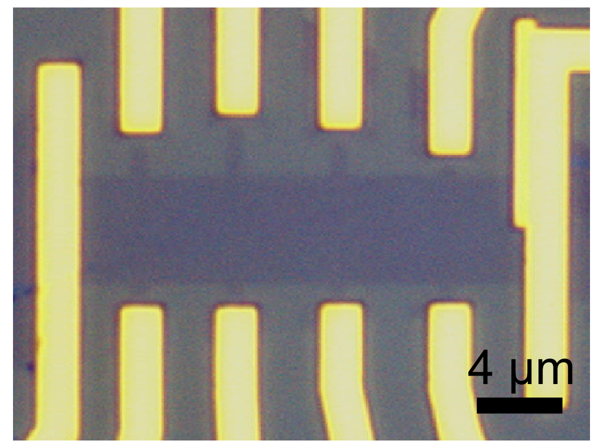
"Mechanical exfoliation"



As-exfoliated graphene

- Starting material is single-crystal Kish graphite
- Mechanically exfoliate on 300 nm SiO_2/Si chips

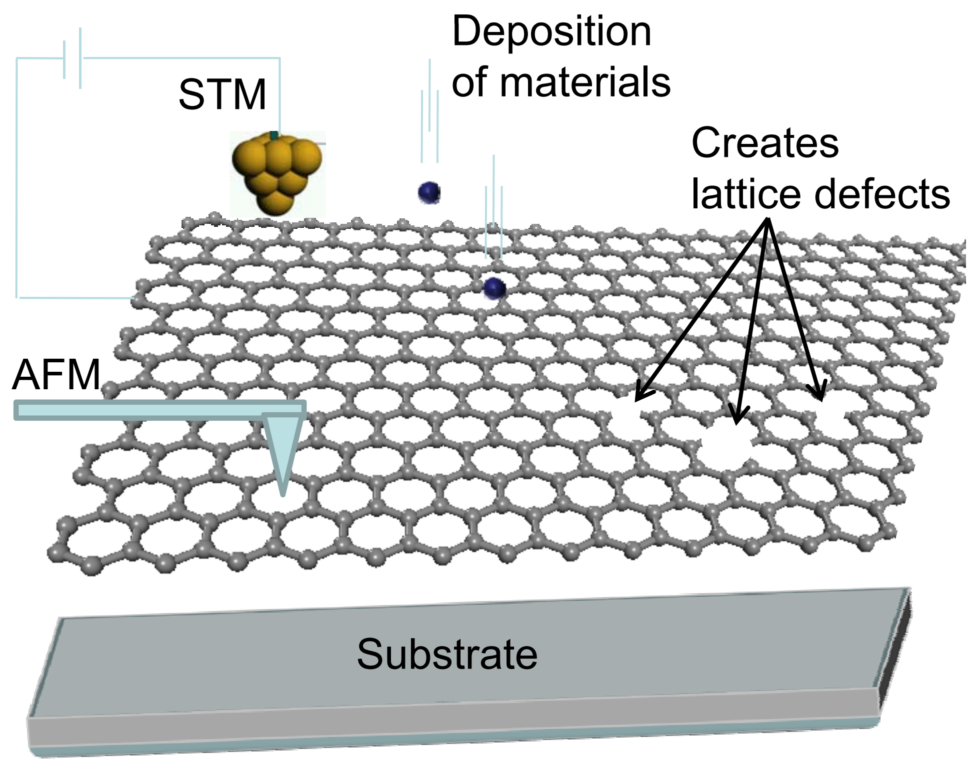




Single layer Hall-bar-shaped device after e-beam lithography and etching

Graphene: "Scratching the Surface"

Every carbon atom in graphene is a surface atom



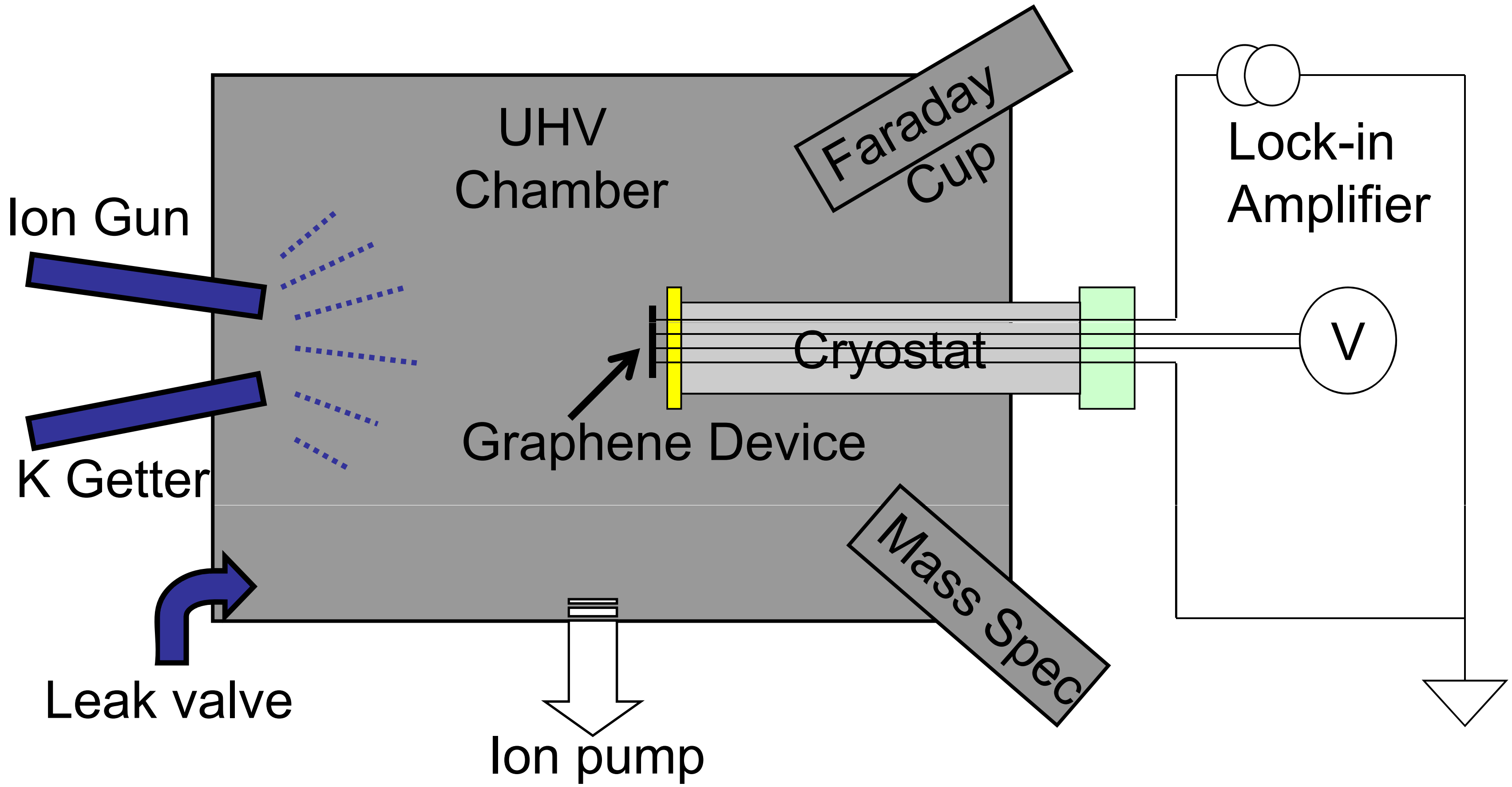
- Surface science tools well suited for characterization of Graphene
- Ultra High Vacuum eliminates unwanted adsorption

➔ Enough time for well-controlled experiments!

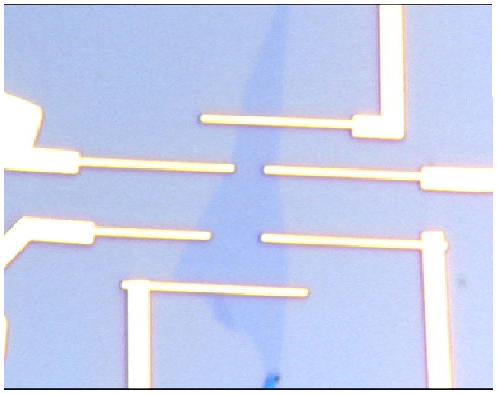
	Pressure	Minimum Coverage Time*
Atmospheric pressure	**760 torr**	**10^{-9} sec**
Ultra high vacuum	**10^{-9} to 10^{-11} torr**	**0.3 - 30 hrs**

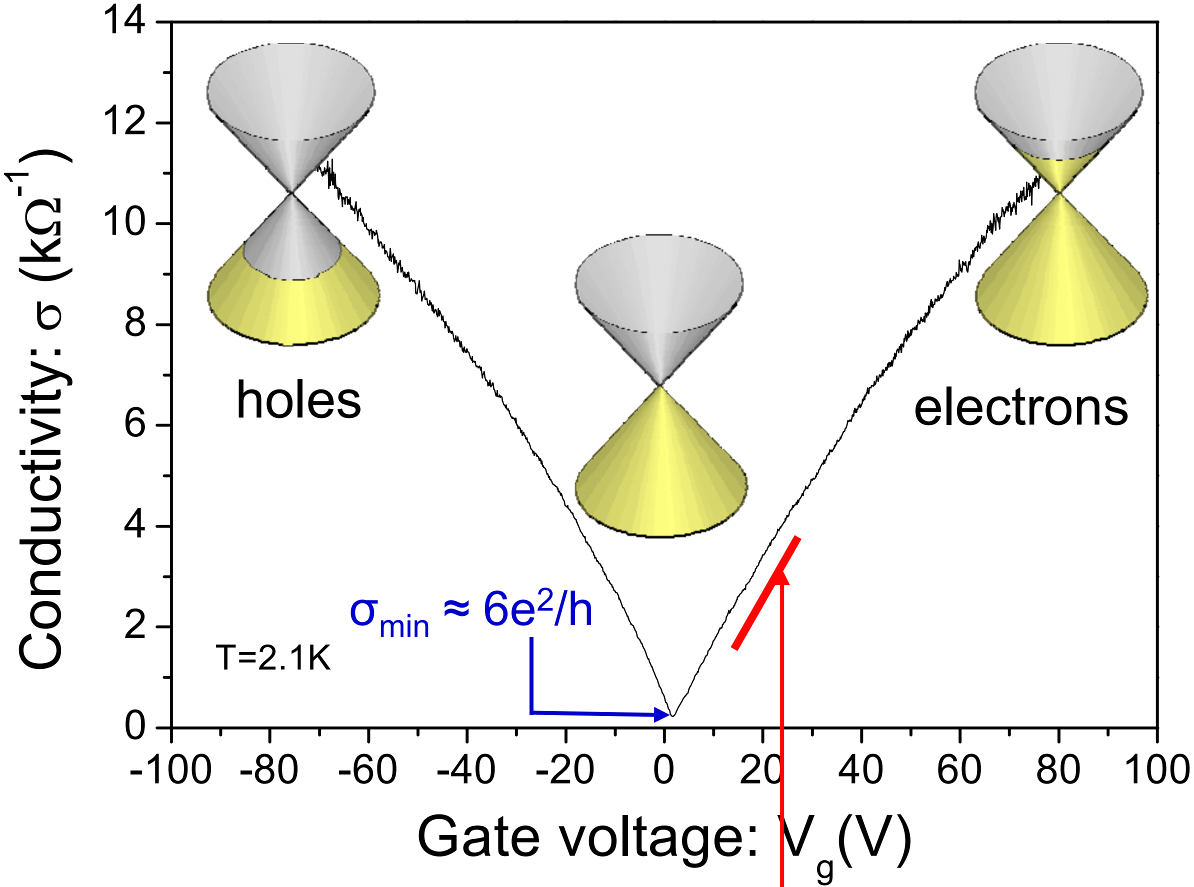
[*Minimum coverage time calculated using a sticking coefficient of 1]

Electronic transport in UHV: Experimental setup



Electrical Characterization of Graphene





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

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

Graphene's Conductivity

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

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

Fermi's Golden Rule:

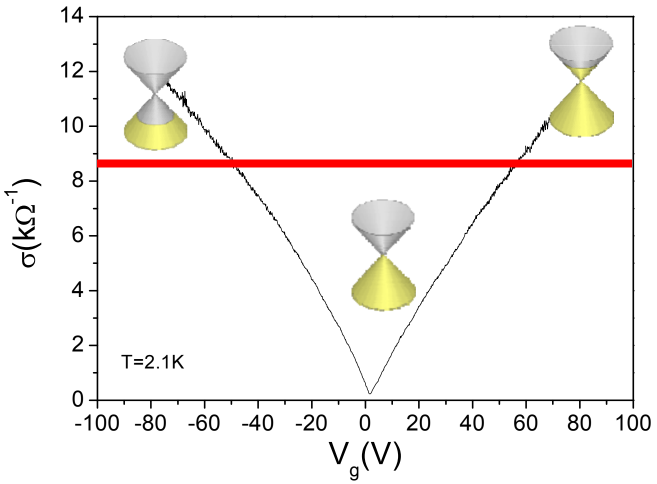
$$\frac{1}{\tau} \propto \frac{2\pi}{\hbar} \left|\langle k|V|k'\rangle\right|^2 D(E)$$

$$\sigma \propto \left|\langle k|V|k'\rangle\right|^{-2}$$

"white-noise disorder": V(q) = constant

σ is independent of E_F!

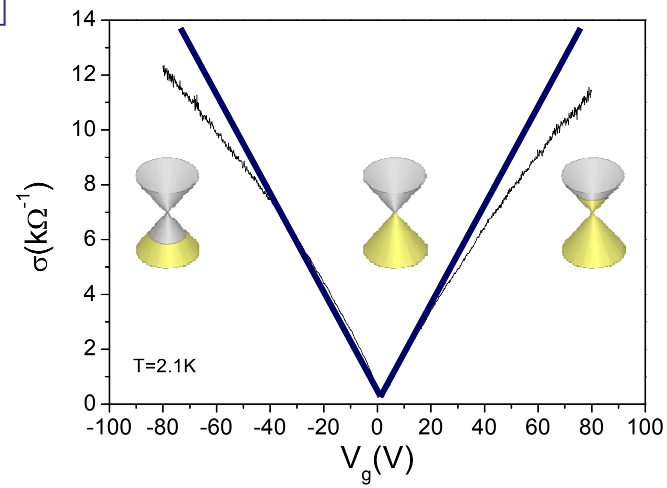
$q = |\mathbf{k} - \mathbf{k}'| \sim k_F$



True for point defects, phonons
see e.g. Pietronero (1980), T. Ando (1996)

Charged impurities: $V(q) = \frac{2\pi e^2}{\kappa q}$

$\sigma \sim k_F^2 \sim n \sim V_g$



Ando (2006), Nomura & MacDonald (2007), Cheianov & Fal'ko (2006), Hwang, Adam, & Das Sarma (2007)

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Charged Impurity Scattering: Potassium Doping in UHV

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

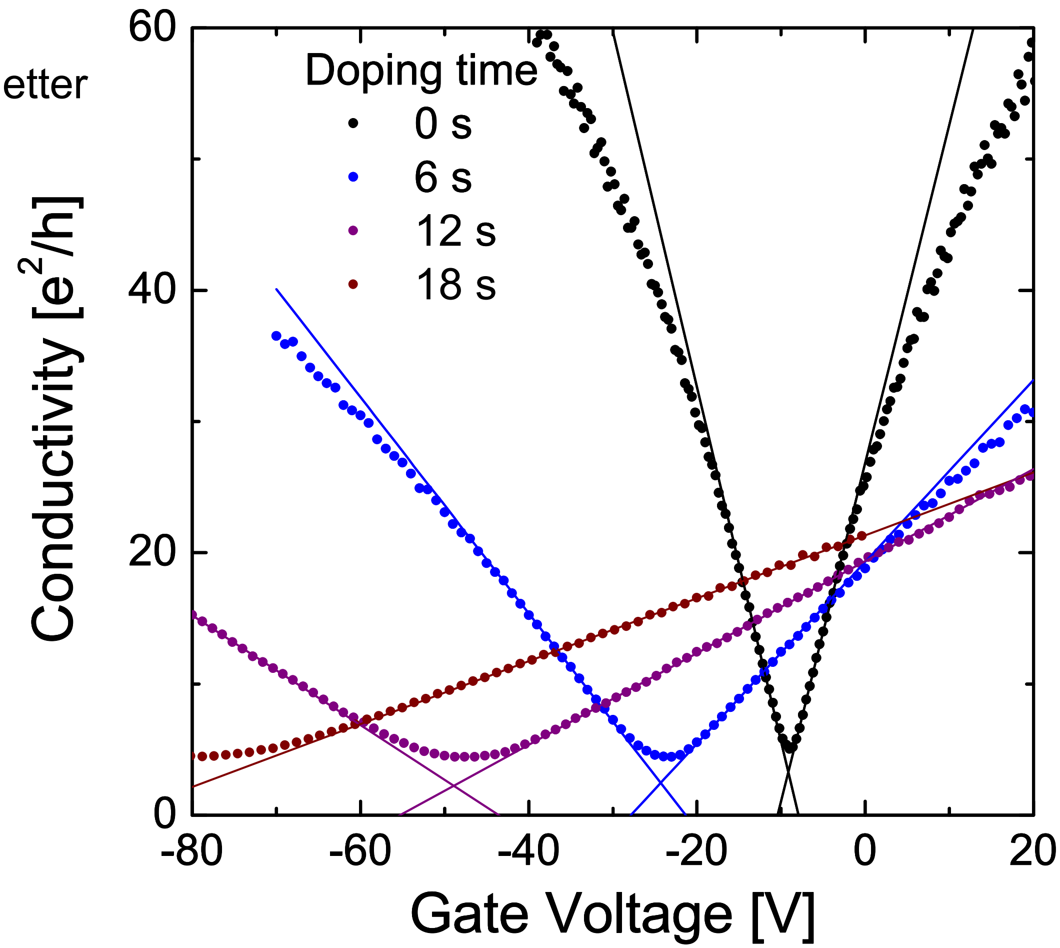
- Clean graphene in UHV at T = 20 K
- Potassium evaporated on graphene from getter

Upon doping with K:

1) mobility decreases
2) σ(Vg) 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:

$$\sigma = 20\frac{e^2}{h}\left(\frac{n}{n_{imp}}\right)$$

or

$$\mu = \frac{5\times10^{15}\,\mathrm{V^{-1}s^{-1}}}{n_{imp}}$$

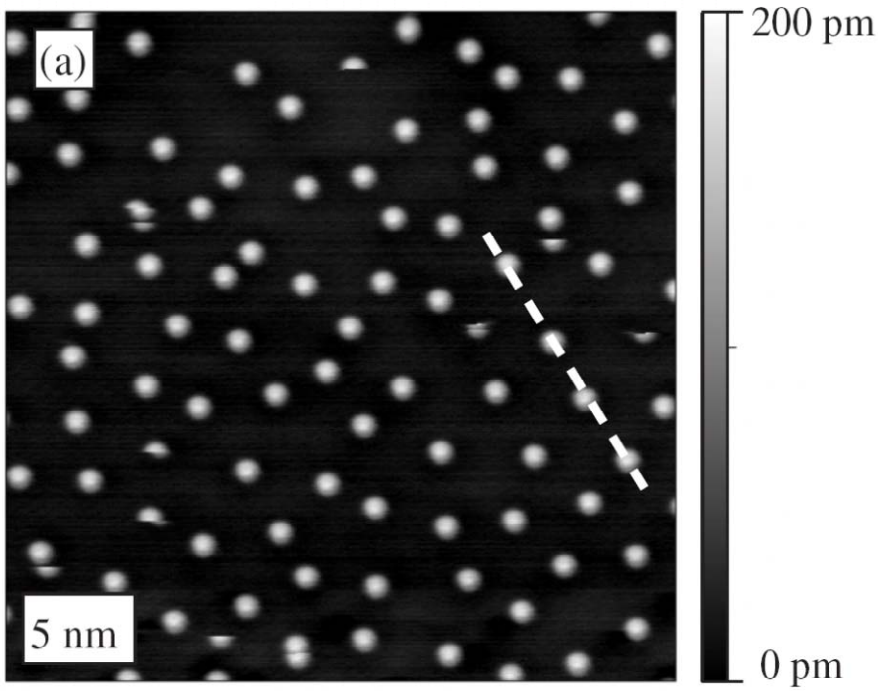
K on Graphite

Correlations in K on graphite:

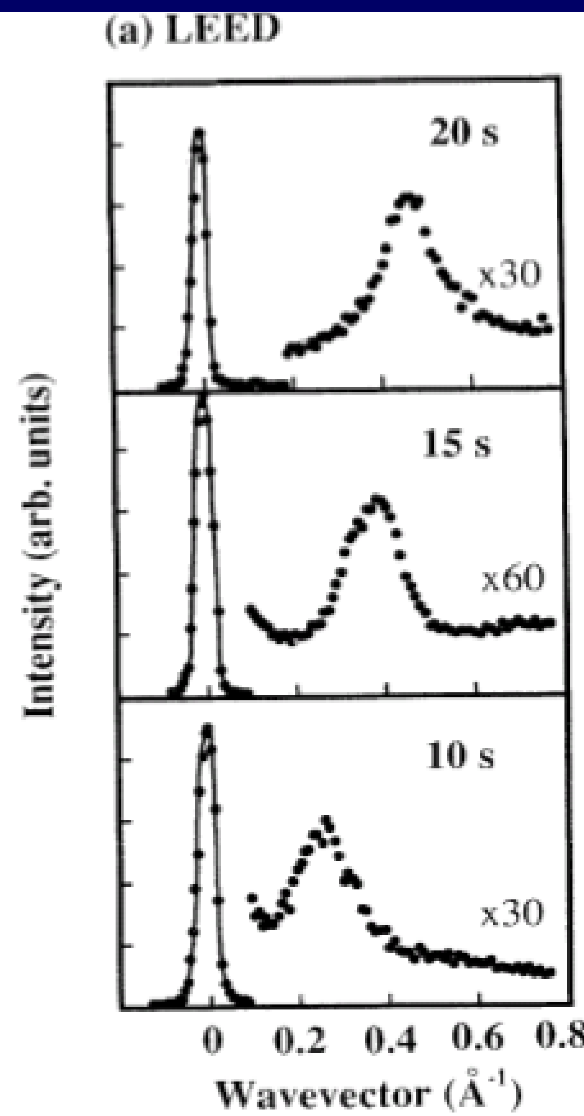
-"dilute" phase is disordered lattice or liquid

- repulsive interactions between K

- coexistence with dense 2x2 phase (C_8K) when dilute phase is more dense than ~7x7



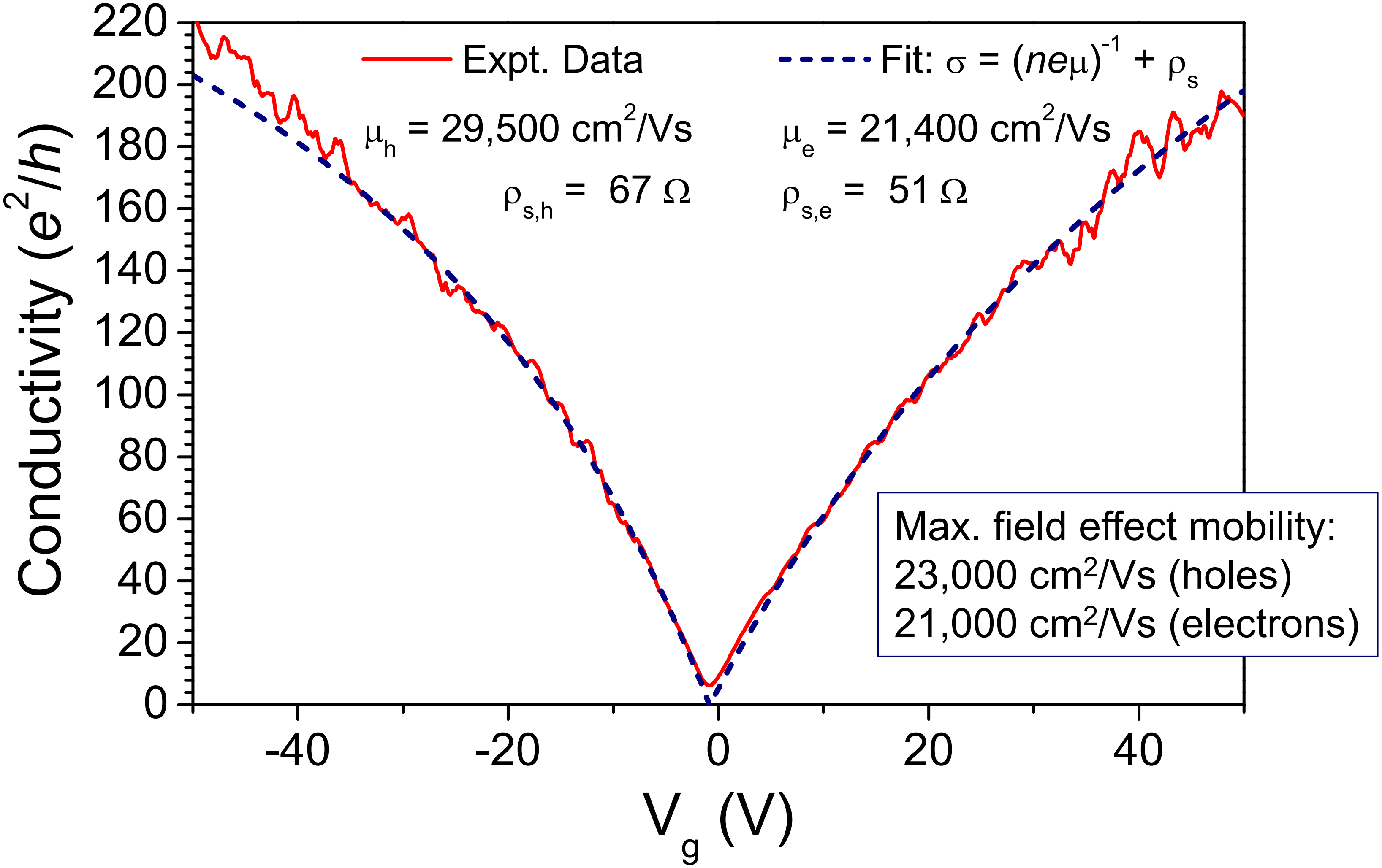
STM of K on graphite @ *T* = 11 K:
Renard et al., *PRL* **106**, 156101 (2011)



LEED of K on graphite @ *T* = 90K
Li, Hock and Palmer, *PRL* **67**, 1562 (1991)

Pristine sample (before potassium)

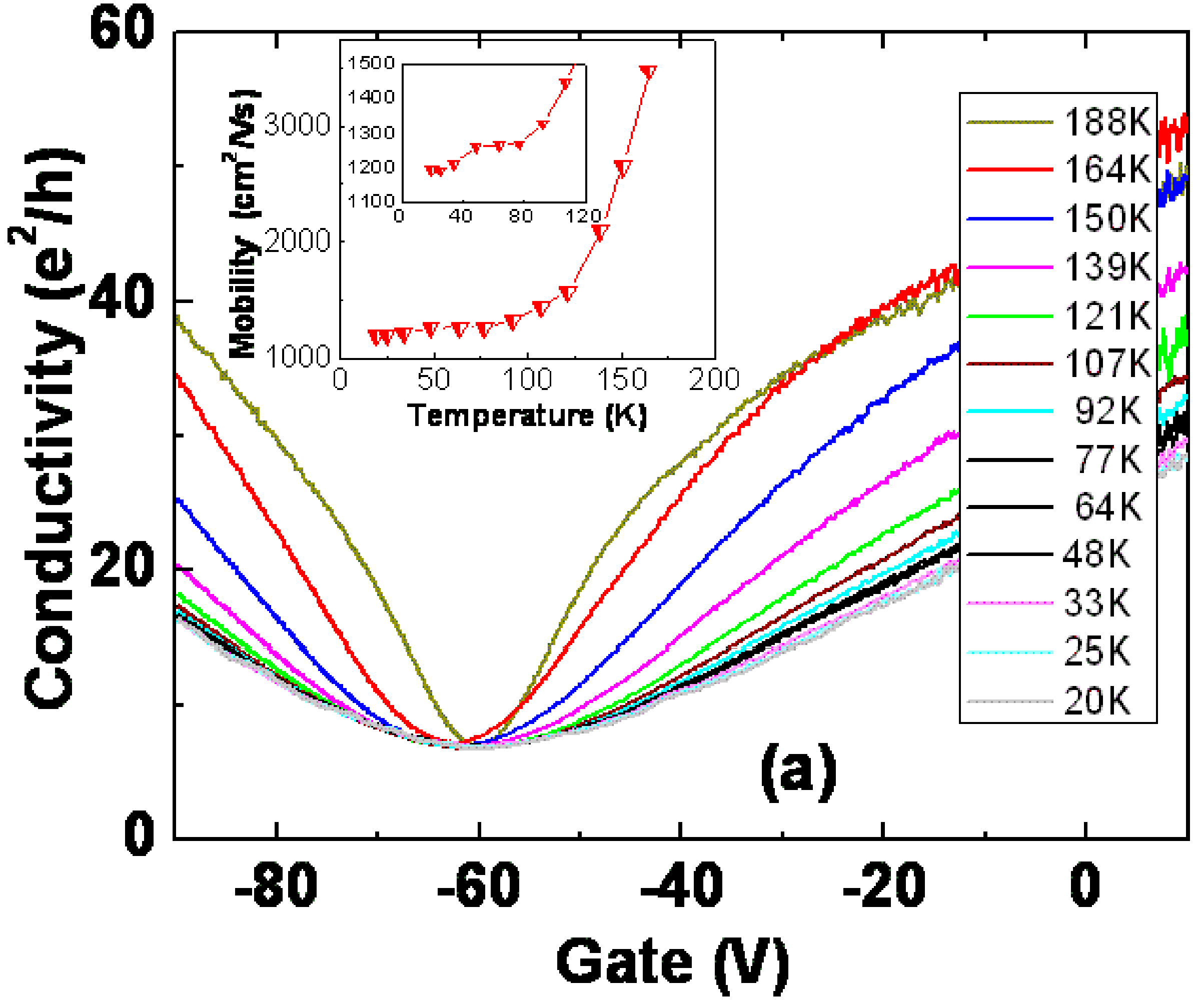
J. Yan and M.S. Fuhrer, *PRL* **107**, 206601 (2011)



Assume initial disorder μ_e, $\rho_{s,e}$ do not change; uncorrelated with K atoms

Annealing Potassium

J. Yan and M.S. Fuhrer, *PRL* **107**, 206601 (2011)



- On heating: mobility increase >4x, doping persists to ~170 K
- Very little change in minimum conductivity

Theory of Correlated Disorder in Graphene

Li, Hwang, Das Sarma, *PRL* **107**, 156601 (2011)

Assume spatial pair distribution function for impurities of form:

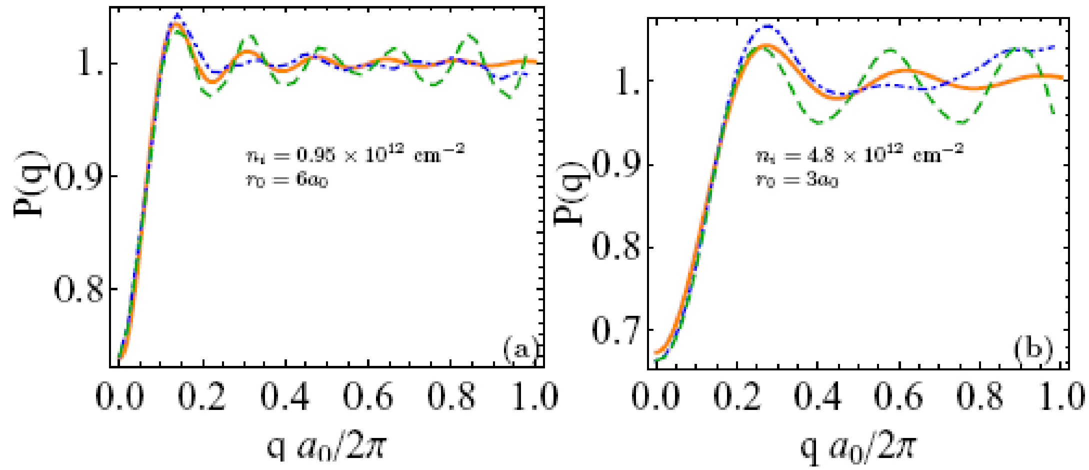
$$g(\mathrm{r}) = \begin{cases} 0 & |\mathrm{r}| < r_0 \\ 1 & |\mathrm{r}| > r_0 \end{cases}$$

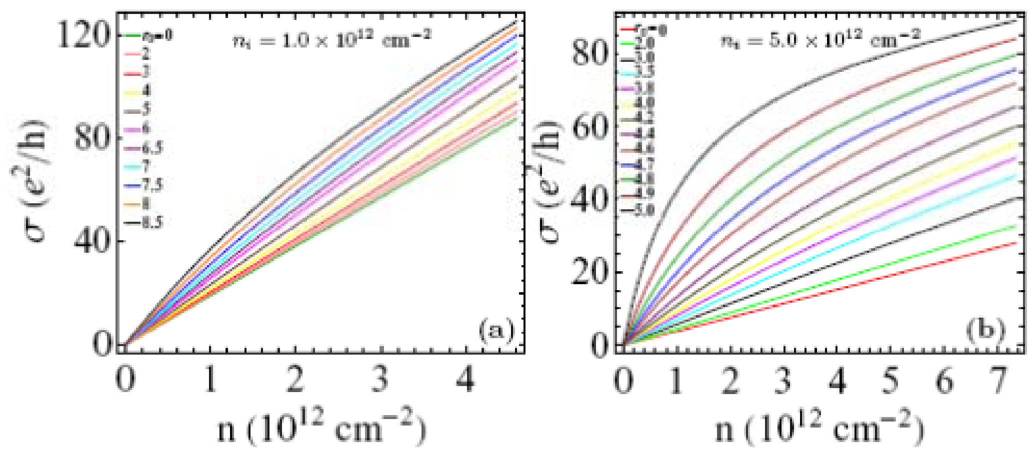
Calculate structure factor $P(\boldsymbol{q})$
Approximated analytically by:

$$P(q) = 1 - 2\pi n_{imp} \frac{r_0}{q} J_1(q r_0)$$

Disorder potential:

$$\left|\frac{V(q)}{\varepsilon(q)}\right|^2 \Rightarrow \left|\frac{V(q)}{\varepsilon(q)}\right|^2 P(q)$$





Fitting to Theory

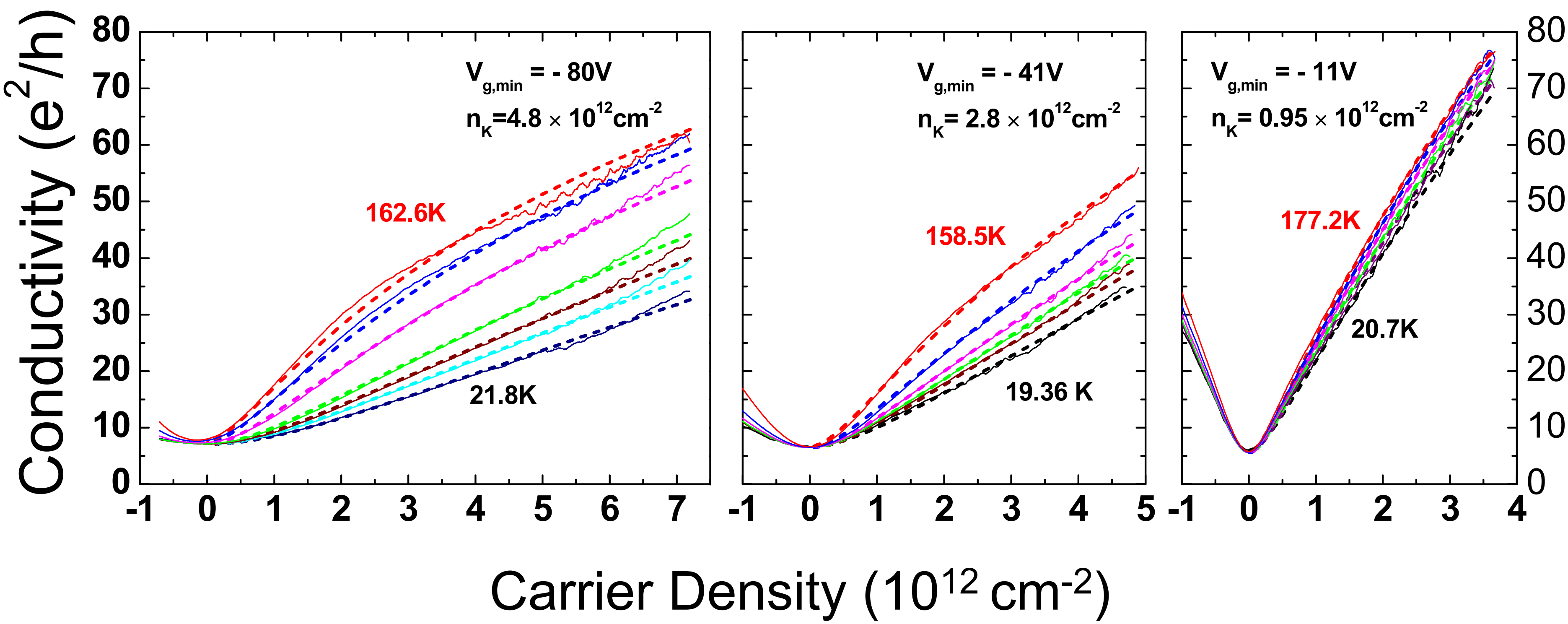
Expt: *PRL* **107**, 206601 (2011) Theory: *PRL* **107**, 156601 (2011)

Assume:

- r_0 = 0.5 nm at lowest *T* (K can be no denser than C_8K phase)
- Assume initial disorder before adding K is fixed
- Small acoustic phonon contribution of 0.1 Ω/K

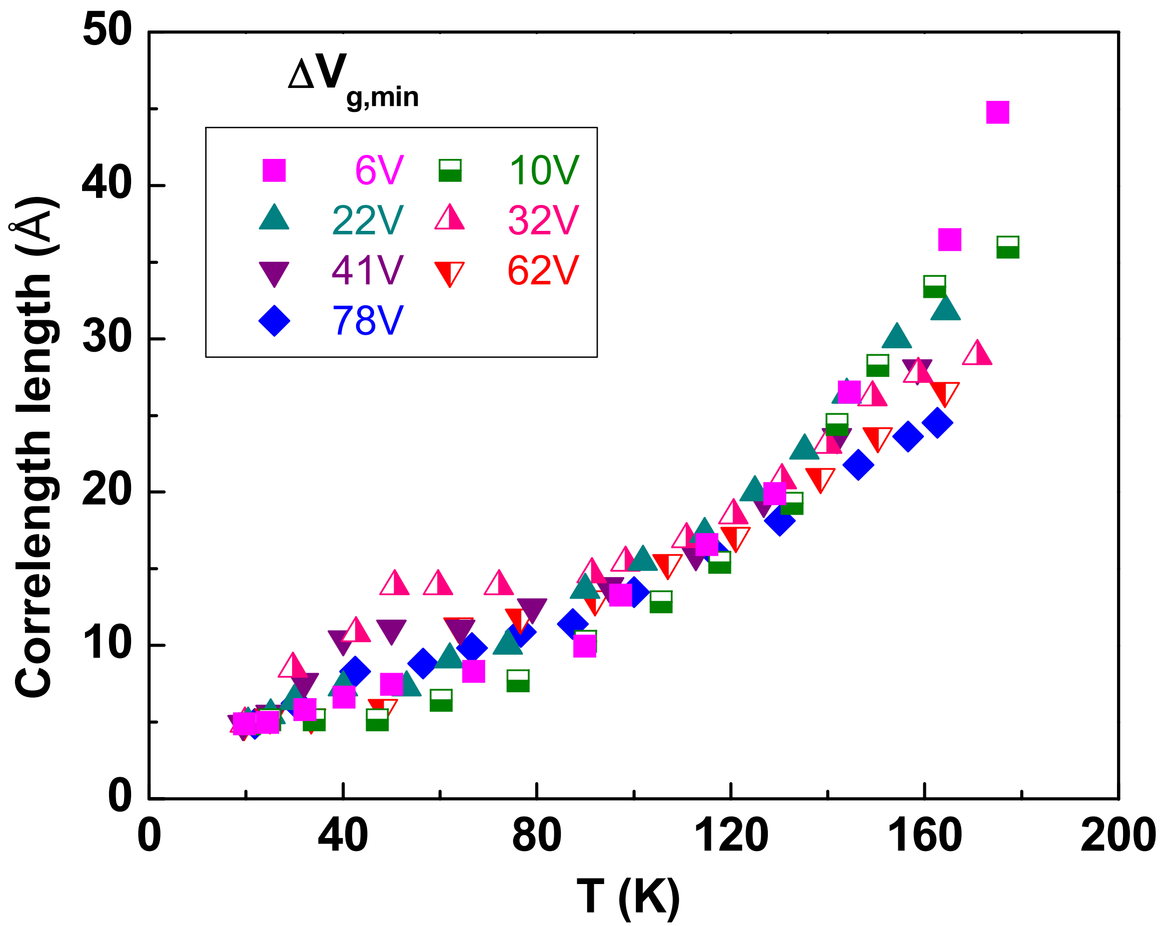
Minimum conductivity treated by: $\sigma_{tot}(n) = \left(\sigma(n)^2 + \sigma_{\min}^2\right)^{1/2}$

n_K = 4.8 x 10^{12} cm^{-2} n_K = 2.8 x 10^{12} cm^{-2} n_K = 0.95 x 10^{12} cm^{-2}



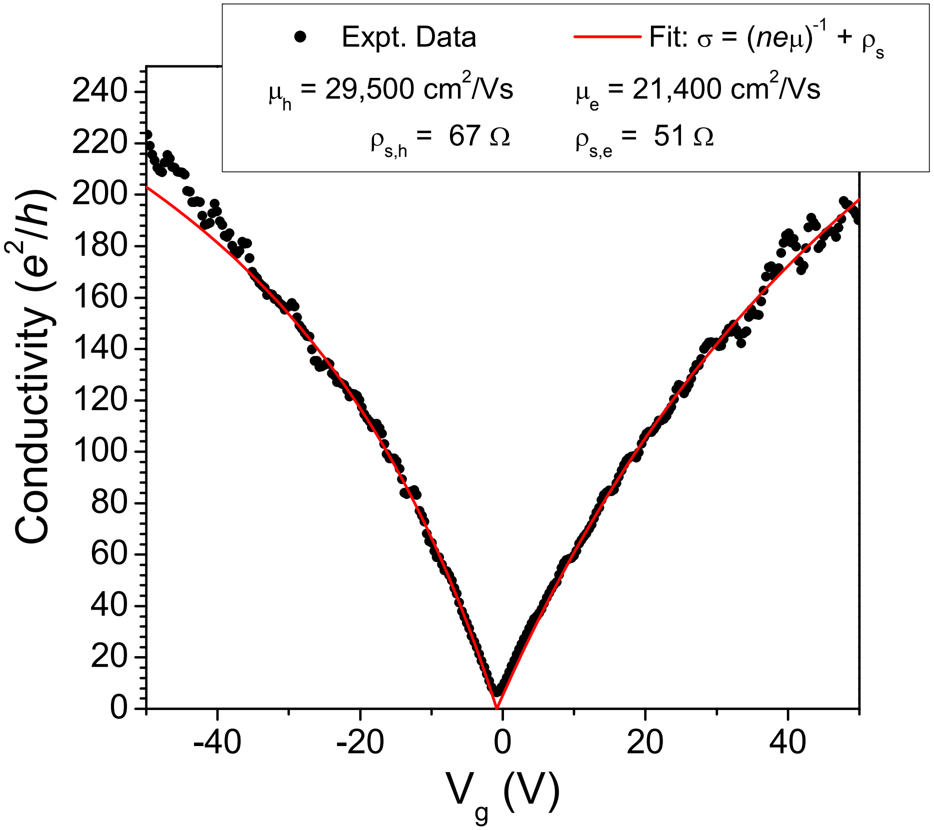
Correlation Length

J. Yan and M.S. Fuhrer, *PRL* **107**, 206601 (2011)

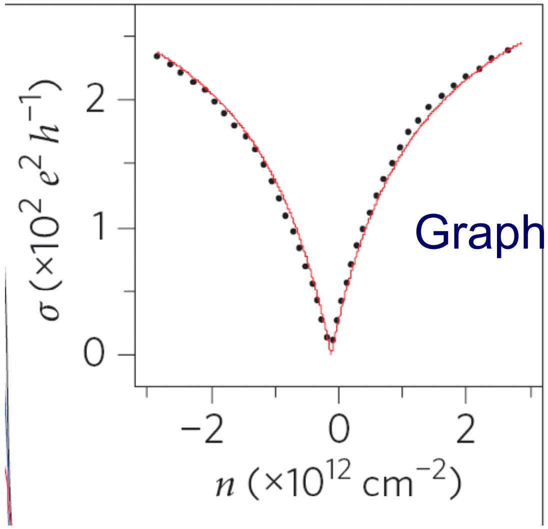


- Single add'l fit parameter: correlation length $r_0(T)$
- Potassium remains highly disordered; $r_0 < r_{imp}$.

Pristine sample revisited



See also Geim group (graphene/SiO_2):
PRL **100**, 016602 (2008)
Kim group (graphene/SiO_2):
PRL **99**, 246803 (2007)
Kim group (graphene/h-BN):
Nature Nano. **5**, 722 (2010)



Graphene/h-BN

Fit to: $$\sigma^{-1} = \frac{1}{ne\mu_L} + \rho_s$$

Assumptions: μ_L corresponds to long-range (charged impurity) disorder, ρ_s is short-ranged disorder (point defects).

Problem: source of ρ_s is a mystery!

Graphene's Conductivity

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

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

Fermi's Golden Rule:

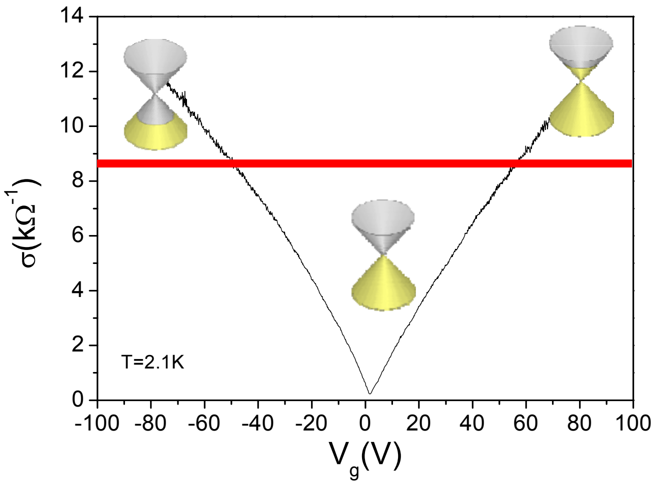
$$\frac{1}{\tau} \propto \frac{2\pi}{\hbar} \left| \langle k | V | k' \rangle \right|^2 D(E)$$

$$\Rightarrow \quad \sigma \propto \left| \langle k | V | k' \rangle \right|^{-2}$$

"white-noise disorder": *V(q) = constant*

σ is independent of E_F!

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



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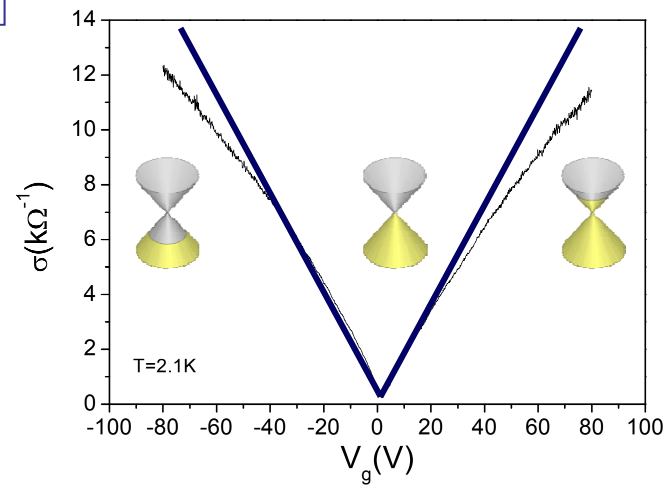


Ando (2006), Nomura & MacDonald (2007), Cheianov & Fal'ko (2006), Hwang, Adam, & Das Sarma (2007)

Correlated Disorder

Li, Hwang, Das Sarma, *PRL* **107**, 156601 (2011)

Modify scattering potential with structure factor:

$$\left|\tilde{V}(q)\right|^2 \Rightarrow \left|\tilde{V}(q)\right|^2 S(q)$$

$$S(q) = 1 - 2\pi n_K \frac{r_c}{q} J_1\left(qr_c\right)$$

Fermi's Golden Rule:

$$\frac{\hbar}{\tau} = \left(\frac{n_K}{4\pi}\right) \int dq \left[\tilde{V}(q)\right]^2 S(q) \left[1 - \cos^2\theta\right]$$

Expand for $k_F r_c \leq 1$: $$S(q) = 1 - \pi n_K r_c^2 \left(1 - \frac{q^2 r_c^2}{8}\right)$$

Then:

$$\sigma(n) = \left(\frac{1}{ne\mu_L} + \rho_S\right)^{-1}$$

$$\mu_L = \frac{\mu_{L,0}}{1-\beta}$$

$$\rho_s = \left(\frac{h}{e^2}\right)\left[r_s^2 G_2\left(r_s\right)\beta^2\right] = 290\Omega \times \beta^2$$

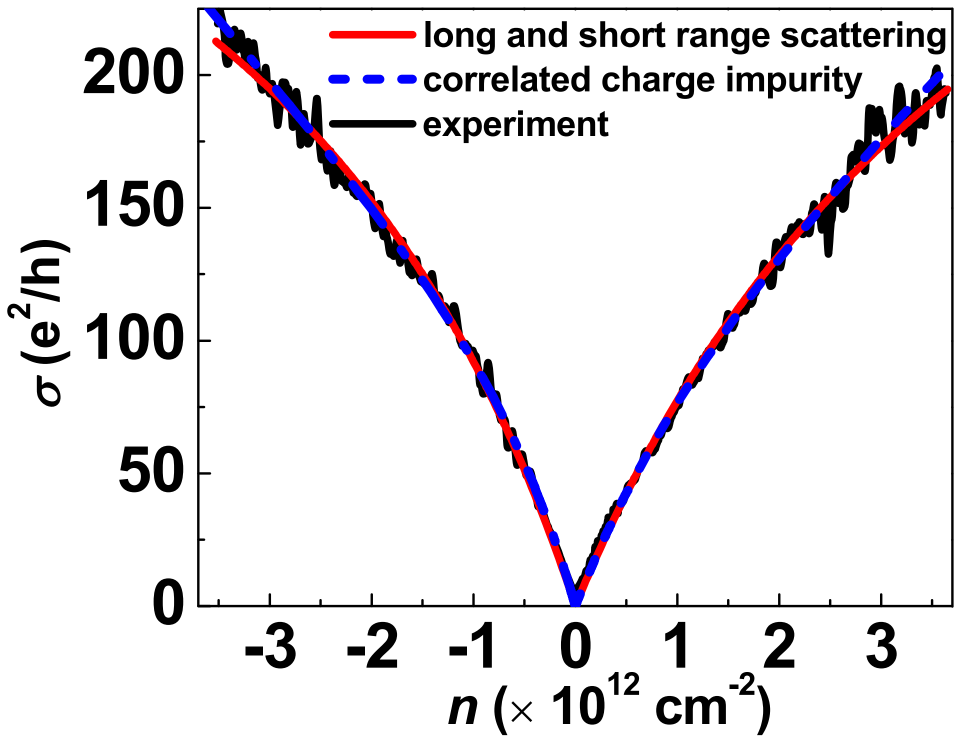
$$\beta = \pi n_{imp} r_c^2 < 1$$

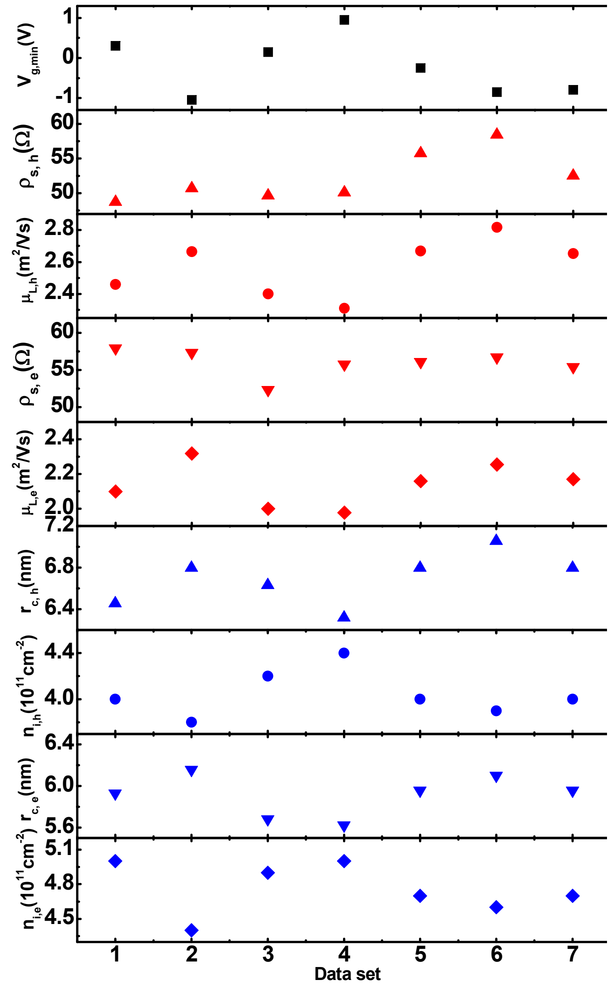
$\beta < 1$ expresses degree of correlations

"ρ_s" term may be due to correlations in charged impurities; no need to invoke another disorder type!

Pristine sample revisited

J. Yan and M.S. Fuhrer, *PRL* **107**, 206601 (2011)





n_{imp} = 4.6 x 10^{11} cm^{-2} and r_c = 6.1 nm for electrons

n_{imp} = 3.9 x 10^{11} cm^{-2} and r_c = 7.0 nm for holes

Note: $\beta \sim 1/6$

Assuming that the SiO_2 mobile surface charges correspond to a nondegenerate plasma frozen at a temperature T_0, the correlation length $r_c = \kappa k_B T_0 / n_{imp} e^2 \sim 6$ nm predicts $T_0 \sim 170$ K

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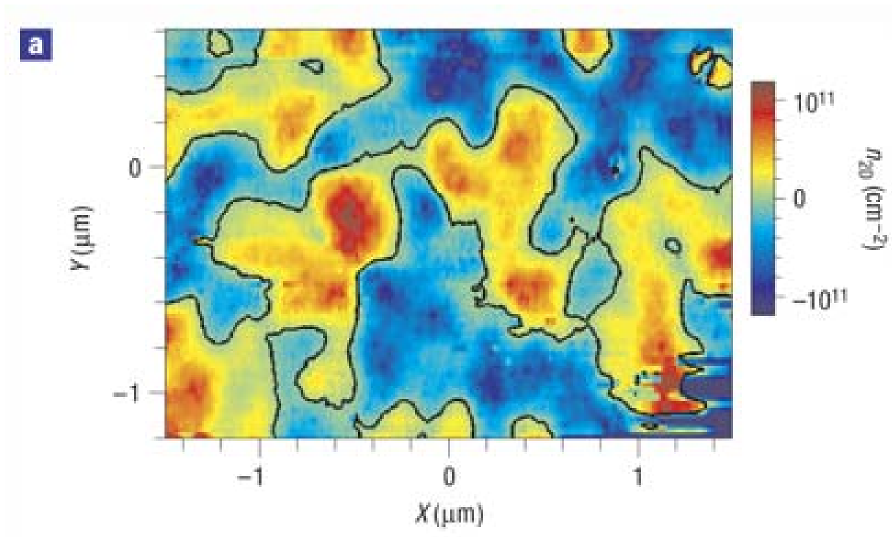
First attempt to measure charge disorder on SiO_2: scanning SET

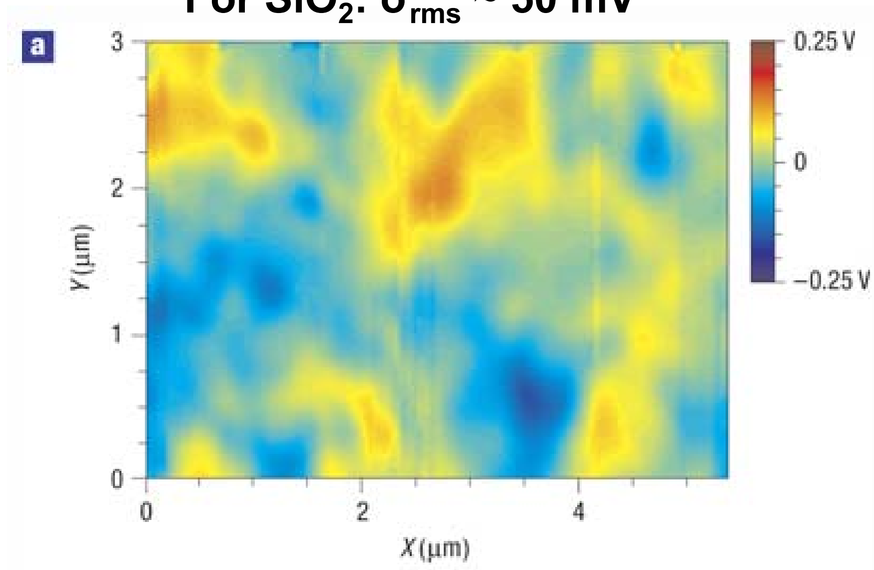
Martin *et al.* *Nature Physics* **4**, 144 (2008)   (Yacoby group, Harvard)

- **Potential mapping of graphene and SiO_2 using a single electron transistor (SET)**
- **High charge sensitivity: fraction of an electron**
- **Limited spatial resolution ~150nm**



Graphene
$\Delta n = \pm\ 3.9 \times 10^{10}\ cm^{-2}$
Intrinsic: $\Delta n = 2.3 \times 10^{11}\ cm^{-2}$

For SiO_2: σ_{rms} ~ 50 mV

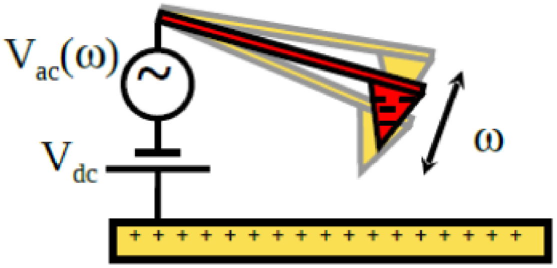


SiO_2 fluctuations:
$\Delta n = \pm\ 2 \times 10^{9}\ cm^{-2}$
***d* = 150 nm from SiO_2**

- **Concluded that fluctuations over SiO_2 could not cause observed fluctuations in graphene**

Kelvin Probe Microscopy

- **Topography and surface potential data are collected simultaneously**
- **V_{dc} nullifies the contact potential difference (CPD) between tip and sample**
- **Then $(V_{dc} + \Delta\varphi) = 0$; $V_{dc} = -\Delta\varphi$**

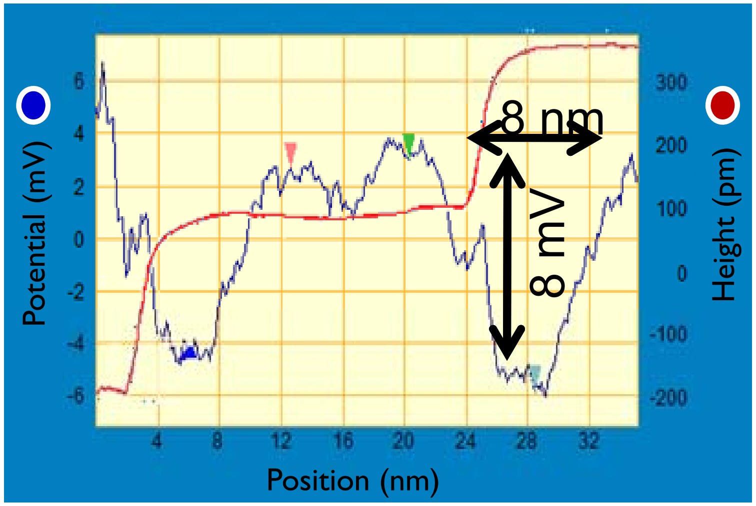


$$A \sim F_z(\omega) \sim (V_{dc} + \Delta\Phi) V_{ac}(\omega)$$

Advantages:

- **Spatial resolution ~tip size (~ 20 nm)**
- **~ mV surface potential resolution**

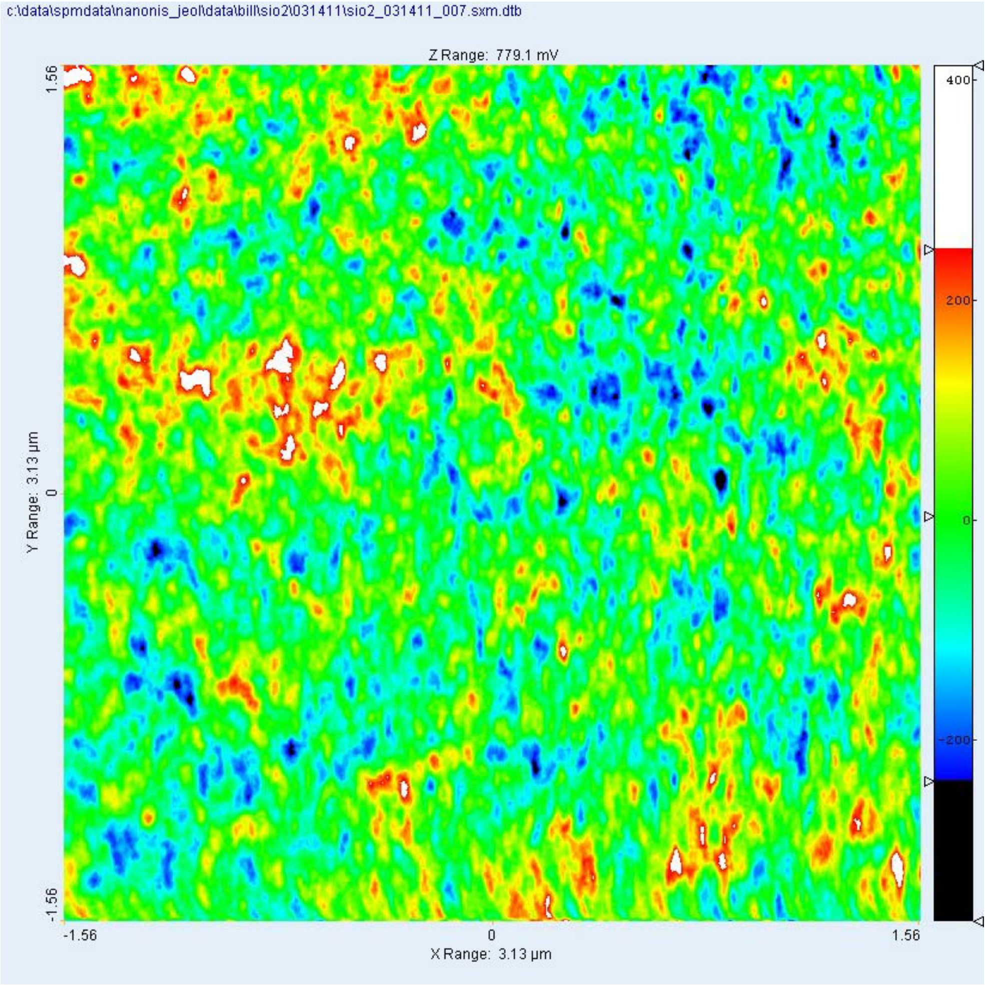
- **Experiment conducted in UHV**
- **Tip is driven at V_{ac} applied at 450 Hz**
- **Topographical data is collected at resonance, ~300 kHz**



Example: Smoluchowski smoothing at Ag(111) step (step dipole) – K. Burson, W.G. Cullen

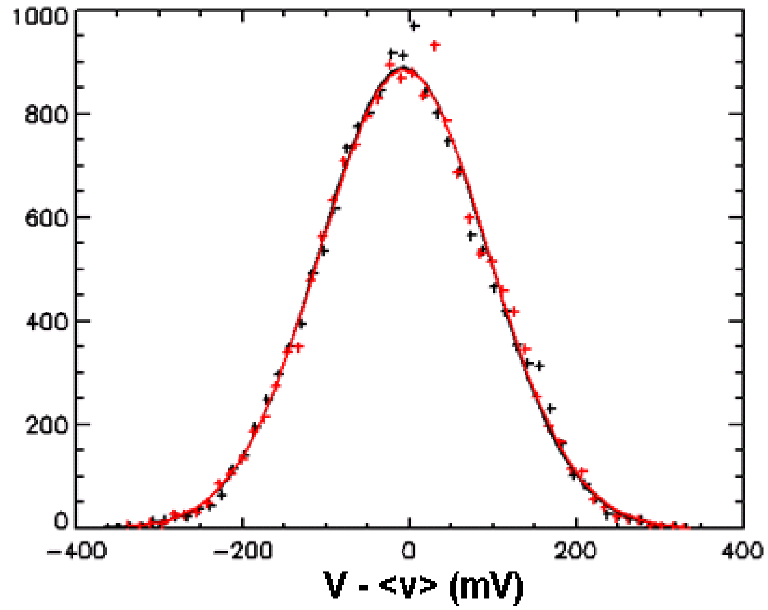
SiO_2 surface potential

Potential map – image size 3.128 μm x 3.128 μm



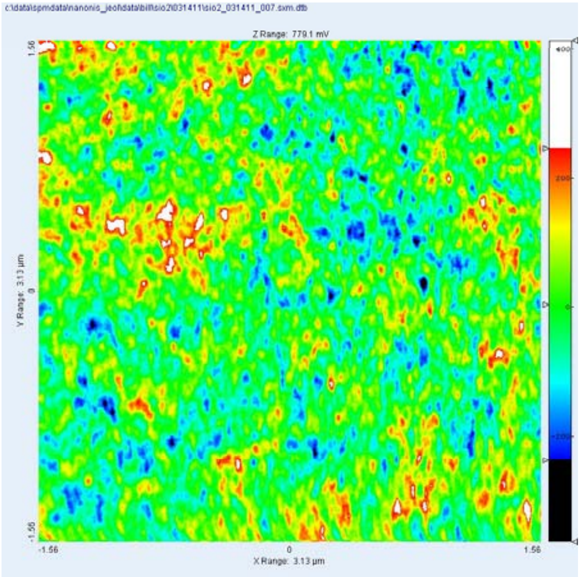
Fit Gaussian to histogram:

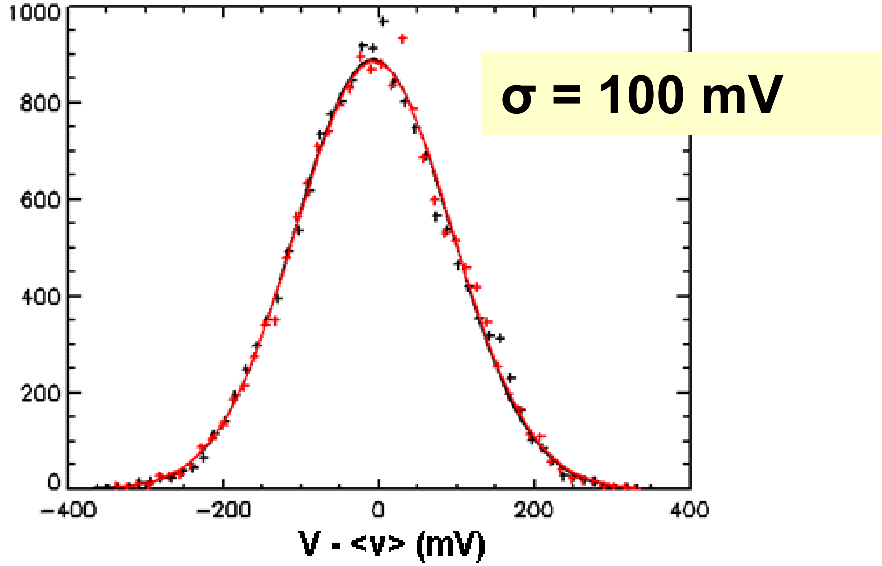
σ_{rms} = 100.3 mV



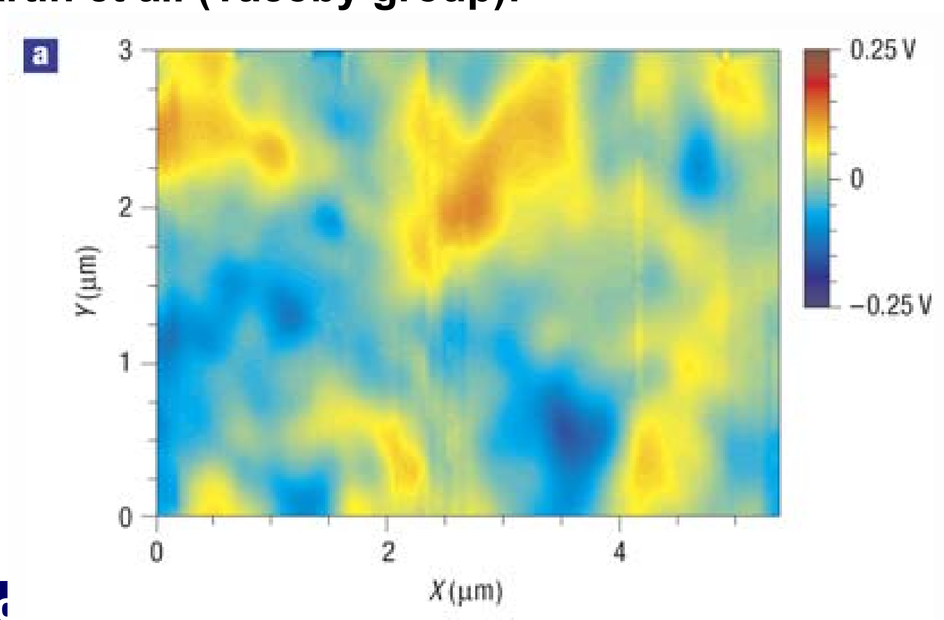
SiO_2 surface potential

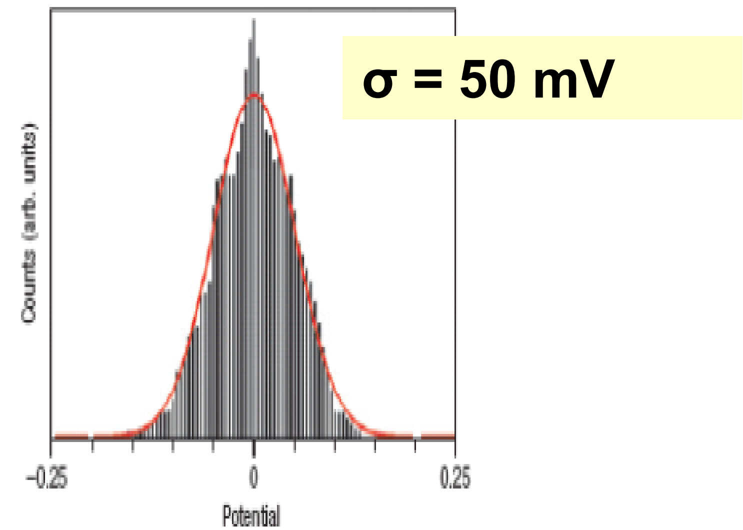
This work:





Martin et al. (Yacoby group):





SiO_2 surface potential

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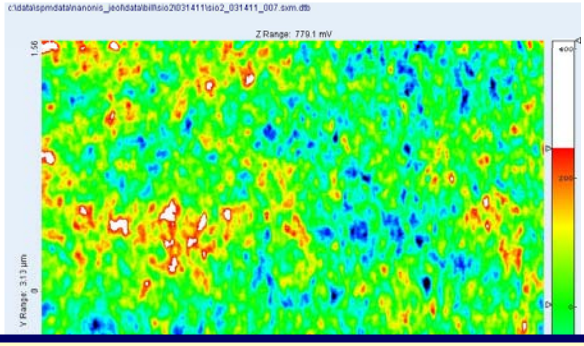
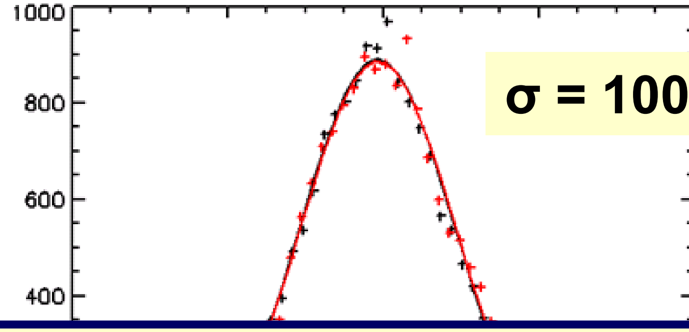


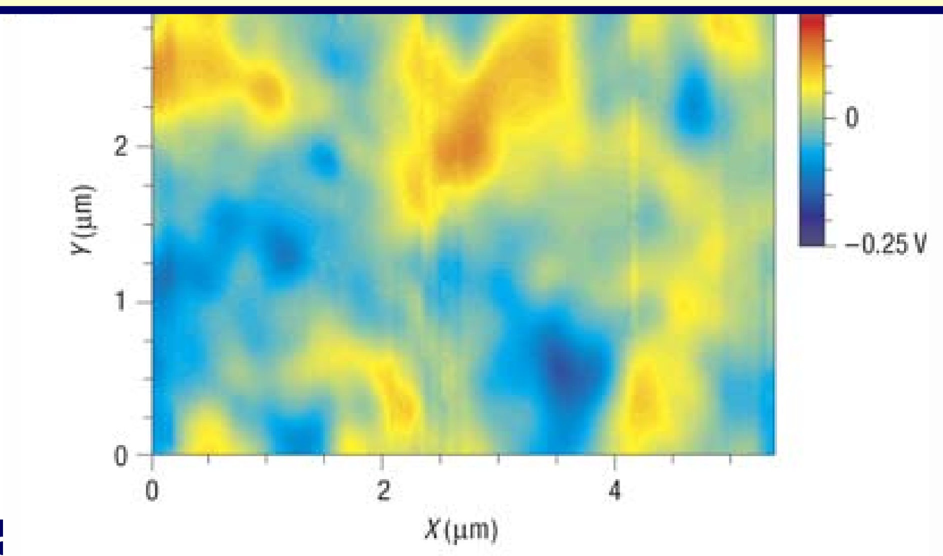


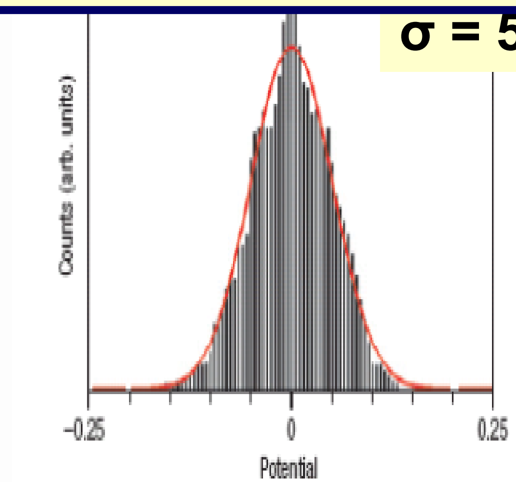
σ = 100 mV

But: variance of random 1/*r* potential diverges!

(experimentally: depends on details of spatial resolution, distance of impurities)

Ma





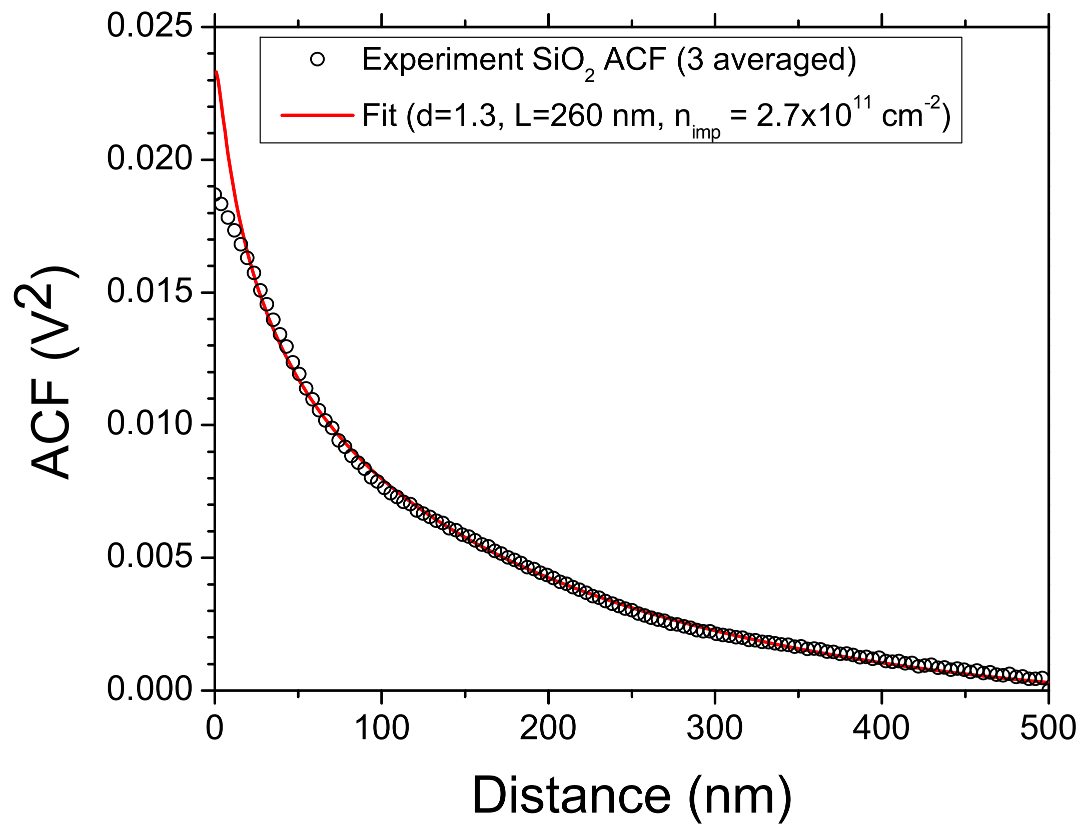
σ = 50 mV

Autocorrelation of the potential

Autocorrelation function of *V*(r):

$$R(r) = \langle V(0)V(r) \rangle = \left(\frac{e^2}{\kappa_{eff} r_{imp}} \right)^2 \int_{1/L}^{\infty} \frac{2\pi q e^{-2qd} J_0(r)}{[q\varepsilon(q)]^2} dq$$

Shaffique Adam
(to be published)



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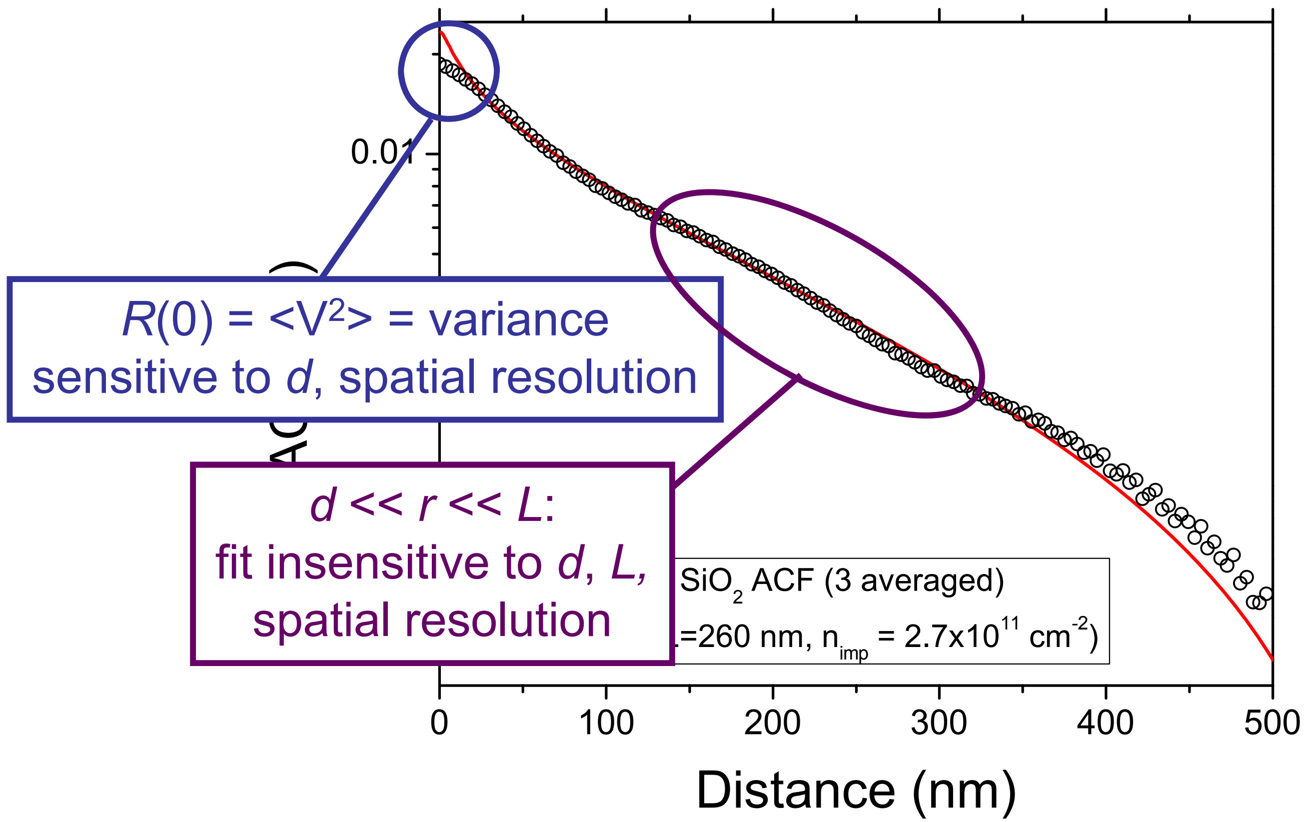

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$$R(r) = \langle V(0)V(r) \rangle = \left(\frac{e^2}{\kappa_{eff} r_{imp}} \right)^2 \int_{1/L}^{\infty} \frac{2\pi q e^{-2qd} J_0(r)}{[q\varepsilon(q)]^2} dq$$

Shaffique Adam
(to be published)

Fit parameters:

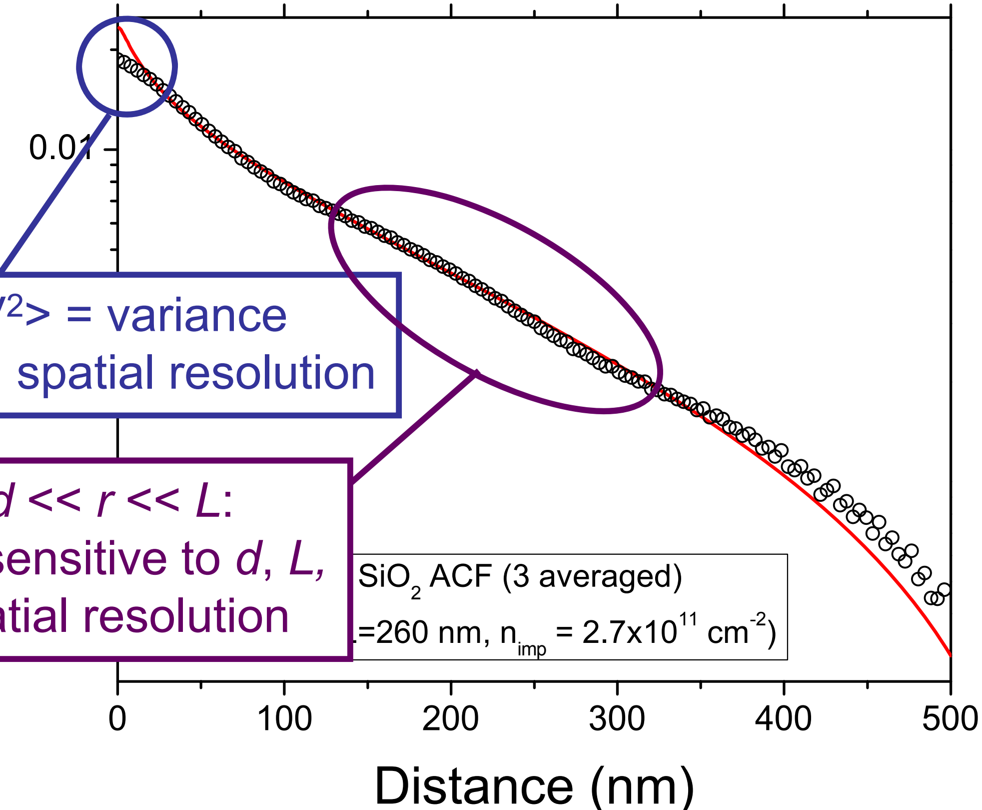
d = 1.3 nm

L = 260 nm (~SiO_2 thickness)

n_{imp} = 2.7 x 10^{11} cm^{-2}

$$\mu = \frac{5 \times 10^{15}\, V^{-1}s^{-1}}{n_{imp}}$$

Expt. by Chen, et al.
***Nat. Phys.* 4, 377 (2008)**



R(0) = <V²> = variance
sensitive to *d*, spatial resolution

d << *r* << *L*:
fit insensitive to *d*, *L*,
spatial resolution

n_{imp} = 2.7 x 10^{11} cm^{-2} → μ = 18,500 cm^2/Vs

Random unit charges at surface of SiO_2 can explain observed mobility in graphene

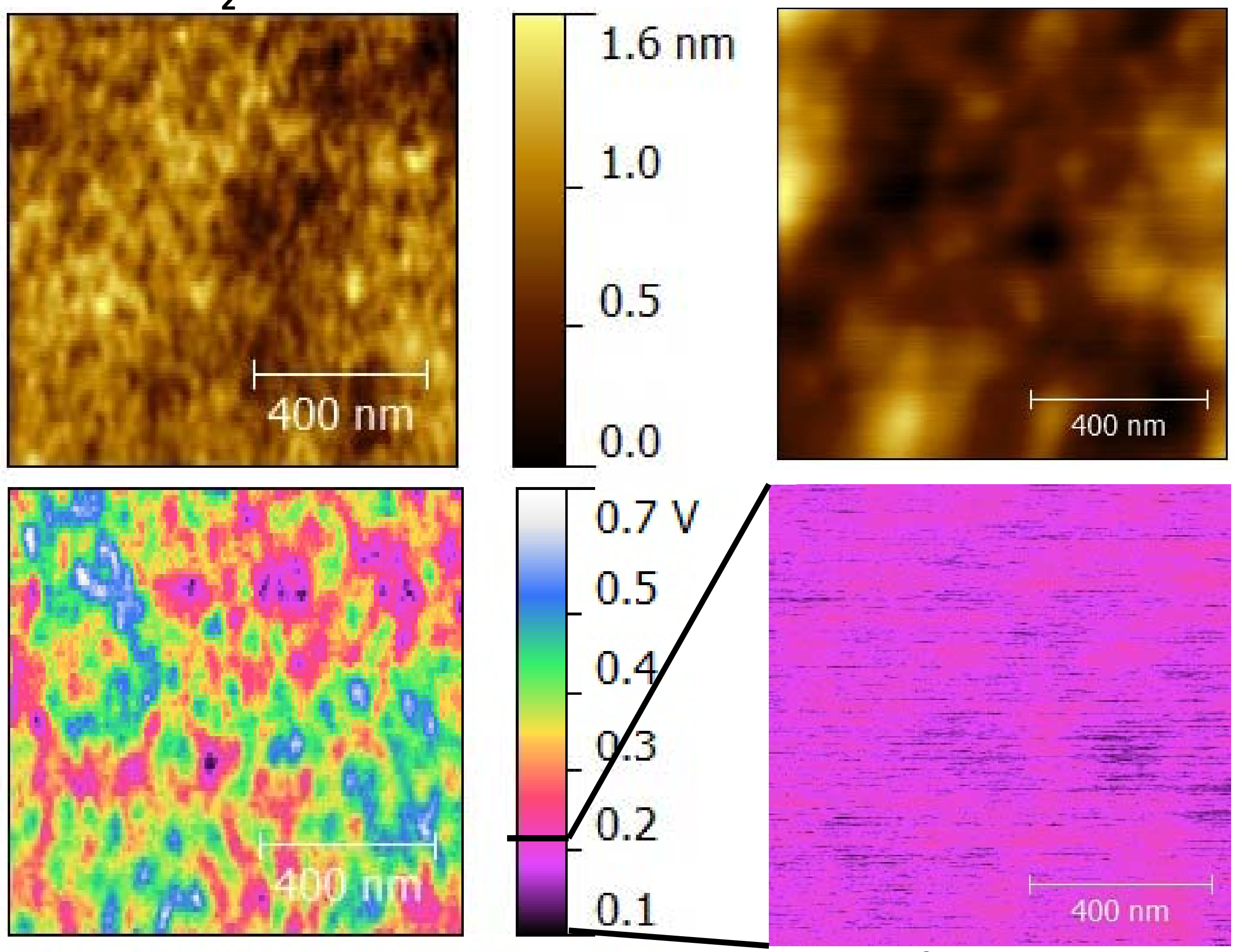
Initial measurement of h-BN

Direct comparison of SiO_2 charge disorder with that of 40-nm thick h-BN flake exfoliated onto SiO_2.

SiO_2 | **40 nm BN**

AFM topography

KPFM potential



z-scale: 0.208 V

σ = 21 mV for this flake.

I. Transport/scanned probe expts. on graphene in UHV

Correlated charged impurities [*PRL* **107**, 206601 (2011)]

Imaging charge disorder of bare SiO_2 [in preparation]

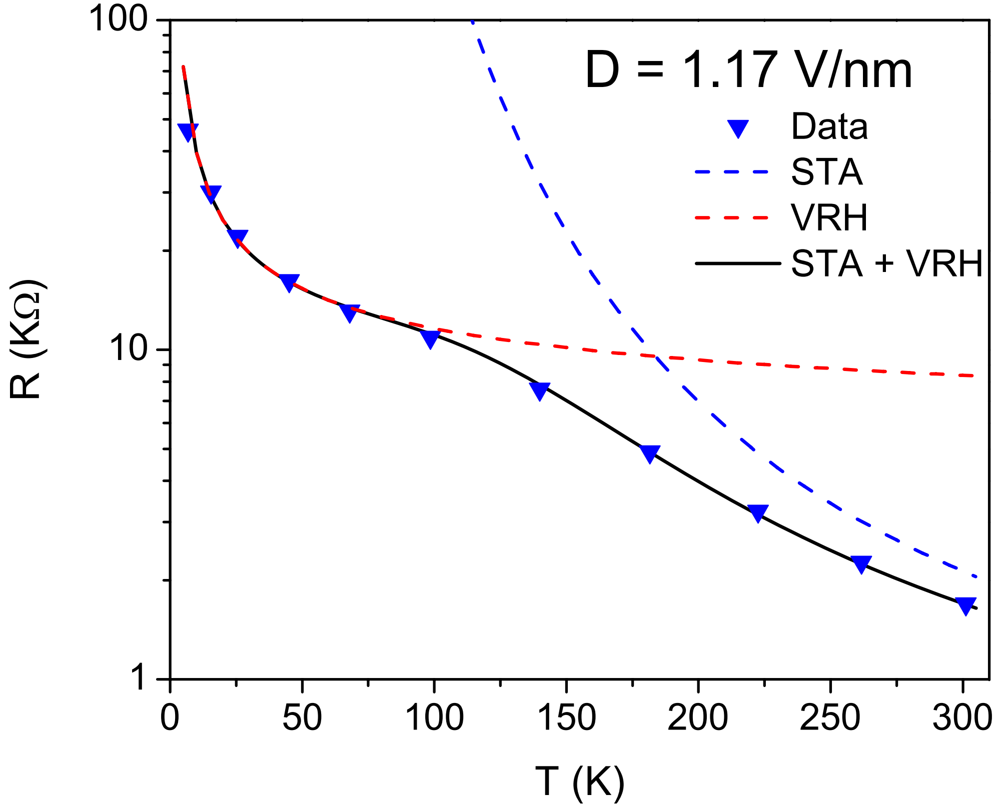
II. Graphene devices

Bandgapped bilayer graphene [*Nano Lett.* **10**, 4521 (2010)]

A bilayer graphene hot electron bolometer [ArXiv:1111.1202]

Dual-gated bilayer graphene: Resistance vs. Temperature

J. Yan and M.S. Fuhrer, *Nano Letters* **10**, 4521 (2010)



$$\sigma = \sigma_{STA} + \sigma_{VRH}$$

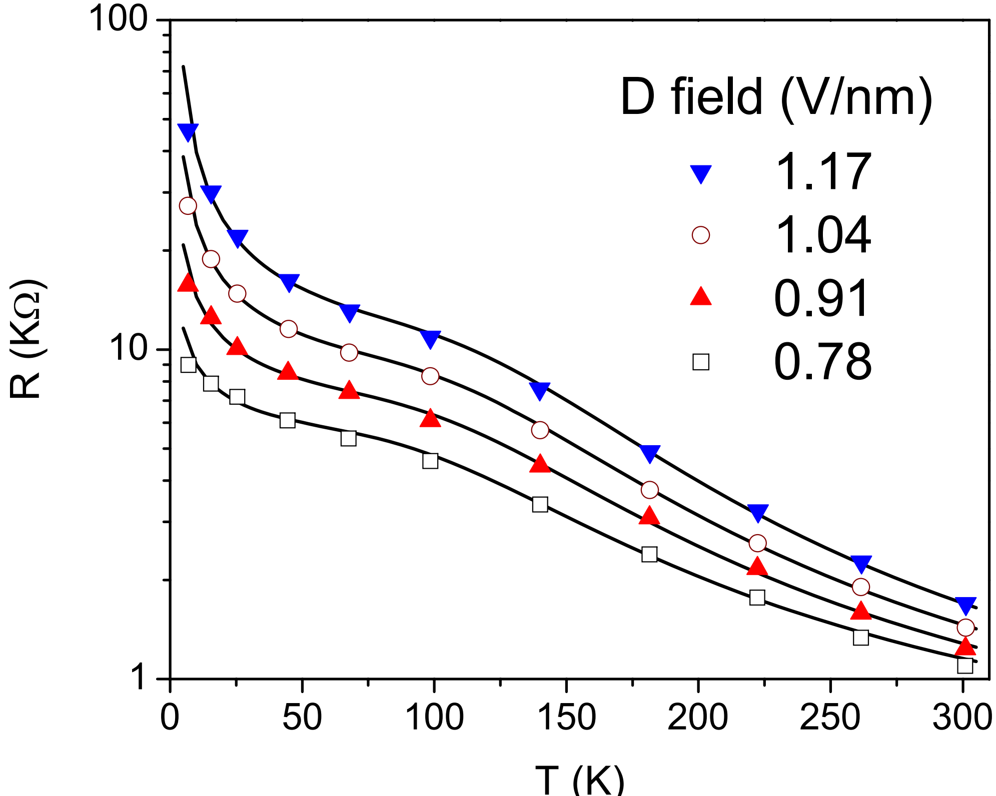
$$\sigma_{STA}^{-1} = R_{S0} e^{\frac{\Delta / 2}{k_B T}}$$

$$\sigma_{VRH}^{-1} = R_{V0} e^{\left(\frac{T_0}{T}\right)^{\frac{1}{3}}}$$

see also: Jun Zhu group, Jarillo-Herrero group

Dual-gated bilayer graphene: Resistance vs. Temperature

J. Yan and M.S. Fuhrer, *Nano Letters* **10**, 4521 (2010)



$$\sigma = \sigma_{STA} + \sigma_{VRH}$$

$$\sigma_{STA}^{-1} = R_{S0} e^{\frac{\Delta/2}{k_B T}}$$

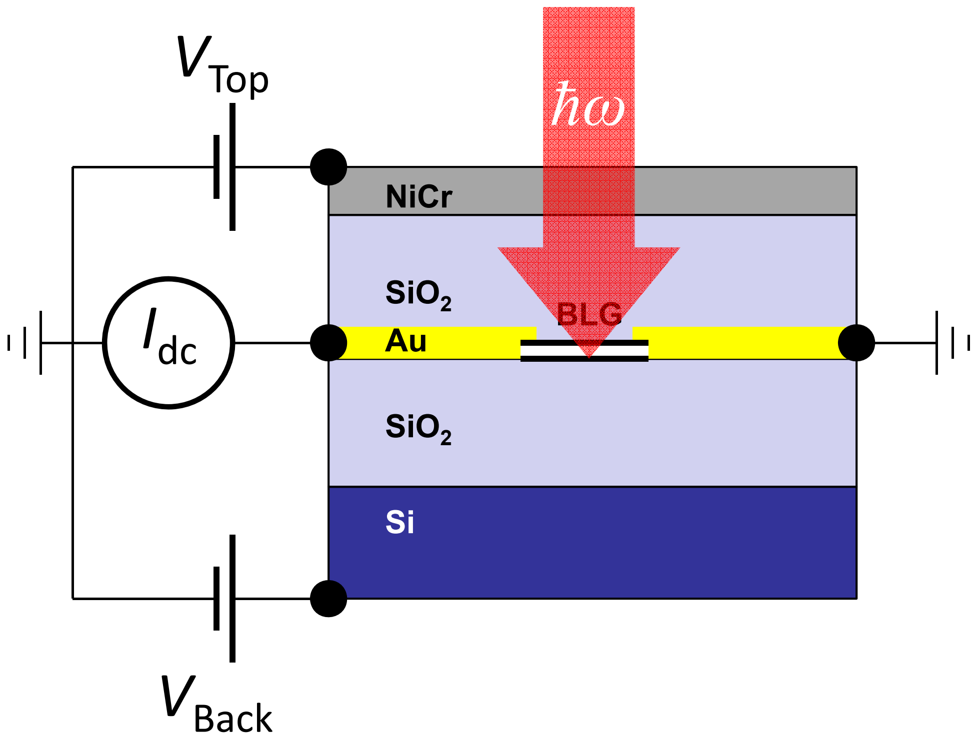
$$\sigma_{VRH}^{-1} = R_{V0} e^{\left(\frac{T_0}{T}\right)^{\frac{1}{3}}}$$

D (V/nm)	Gap (meV)	T_0 (K)	R_{S0}(Kohm)	R_{V0}(Kohm)
1.17	116±6	102±19	0.23±0.02	4.3±0.5
1.04	109±6	55±9	0.22±0.02	3.9±0.3
0.91	102±6	26±8	0.22±0.02	4.0±0.7
0.78	89±8	8±3	0.27±0.03	3.7±0.6

Bandgapped Bilayer Graphene as Photodetector

ArXiv:1111.1202

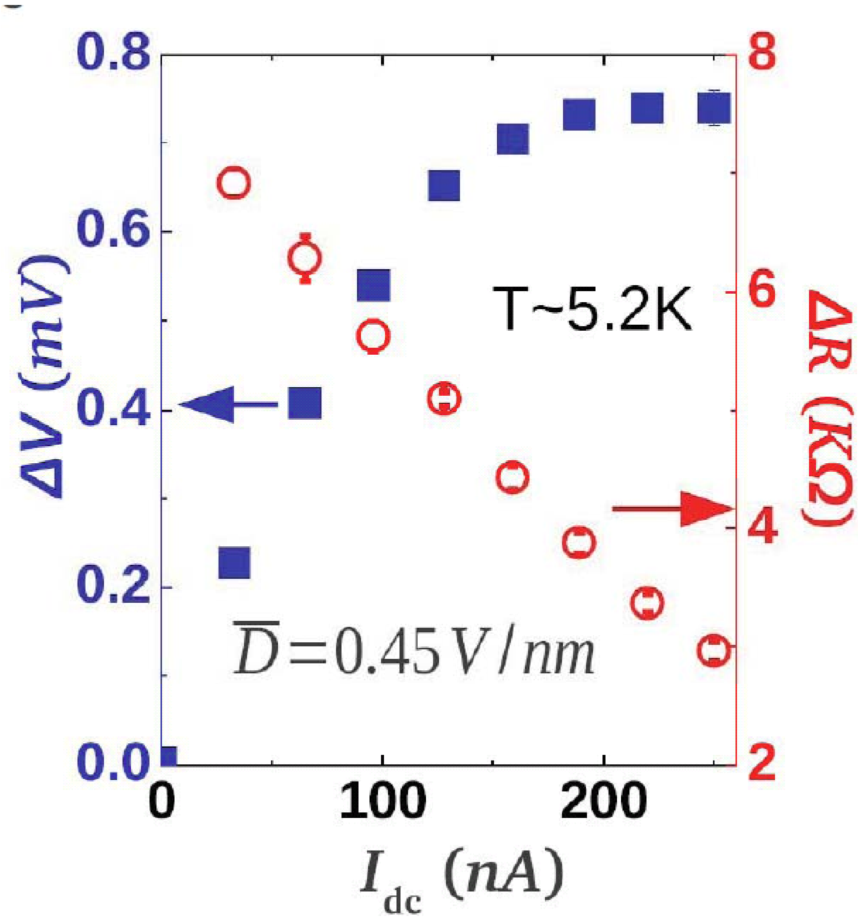
CO_2 laser, photon energy 117meV



Source dc current I_{dc}, measure

$\Delta V = I_{dc} \Delta R$

V_{sd} (light off)- V_{sd} (light on)



Absorbed light energy: 3.7nW

Sensor responsivity: 2×10^5 V/W

Comparable to commercial silicon bolometers!

Origin of the photo-response

ArXiv:1111.1202

- **Photoconductive: Photo-excited carriers Δ*n* enhance conductivity Δσ = (Δ*n*)eμ:**

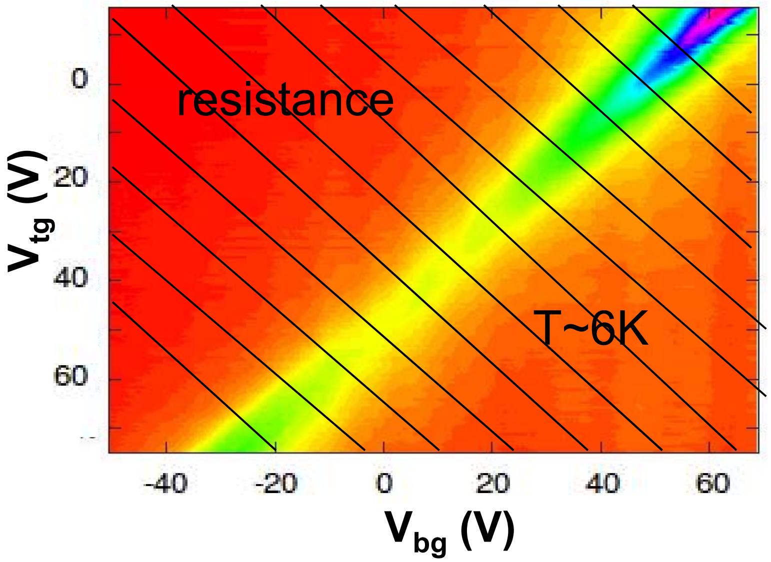
$$\Delta R = \Delta\left(\frac{1}{\sigma}\right) = R^2 \Delta\sigma$$

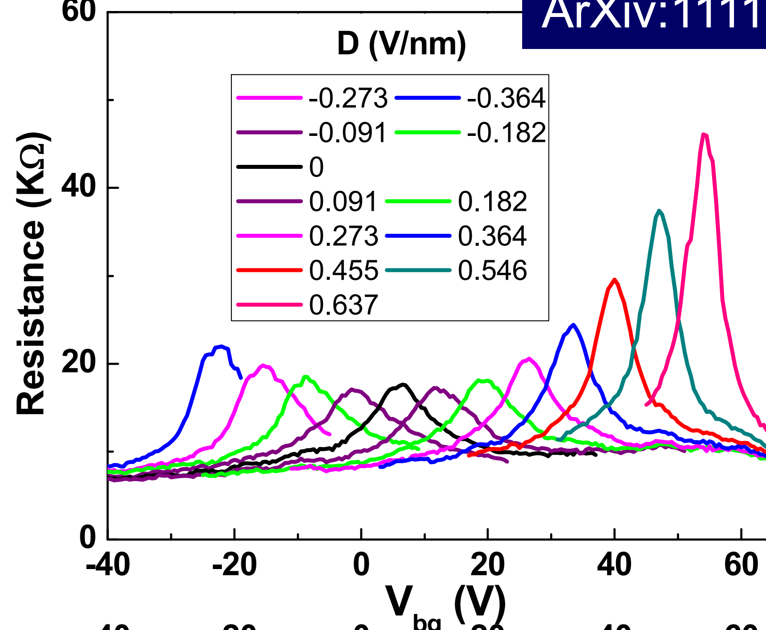
- **Bolometric: Absorbed radiation heats device; temperature-dependent resistance leads to a response:**

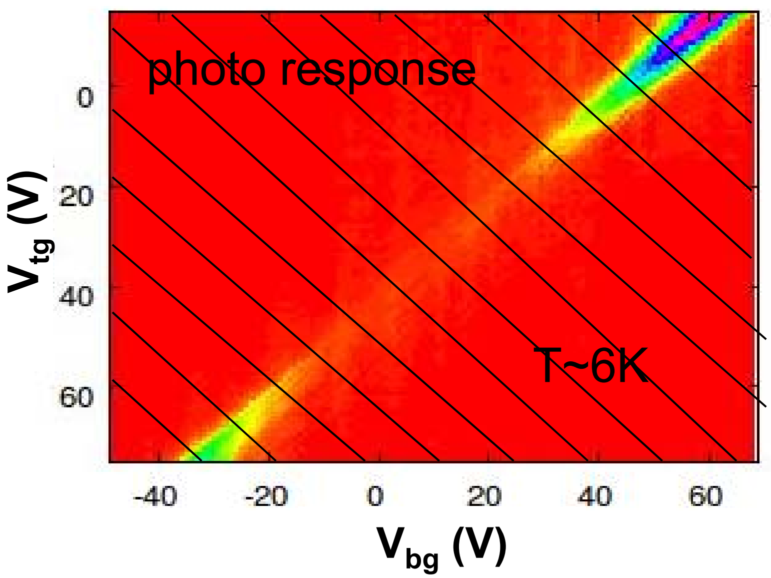
$$\Delta R = \left(\frac{\partial R}{\partial T}\right) \Delta T$$

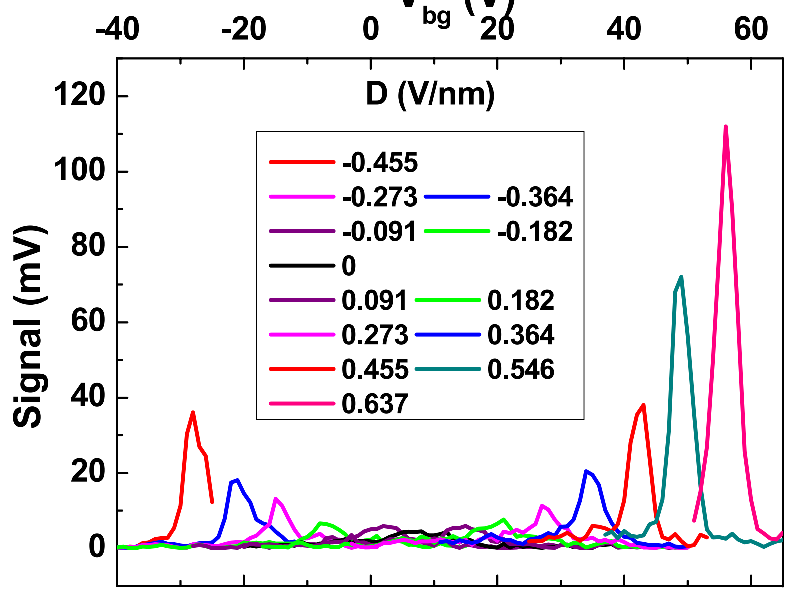
Charge transport vs. photo response

ArXiv:1111.1202



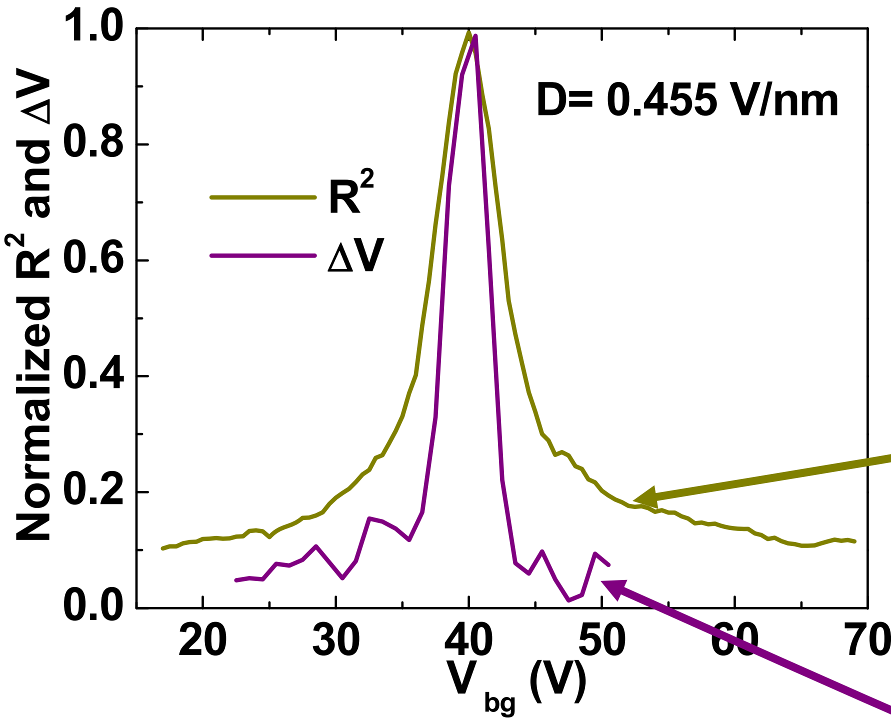


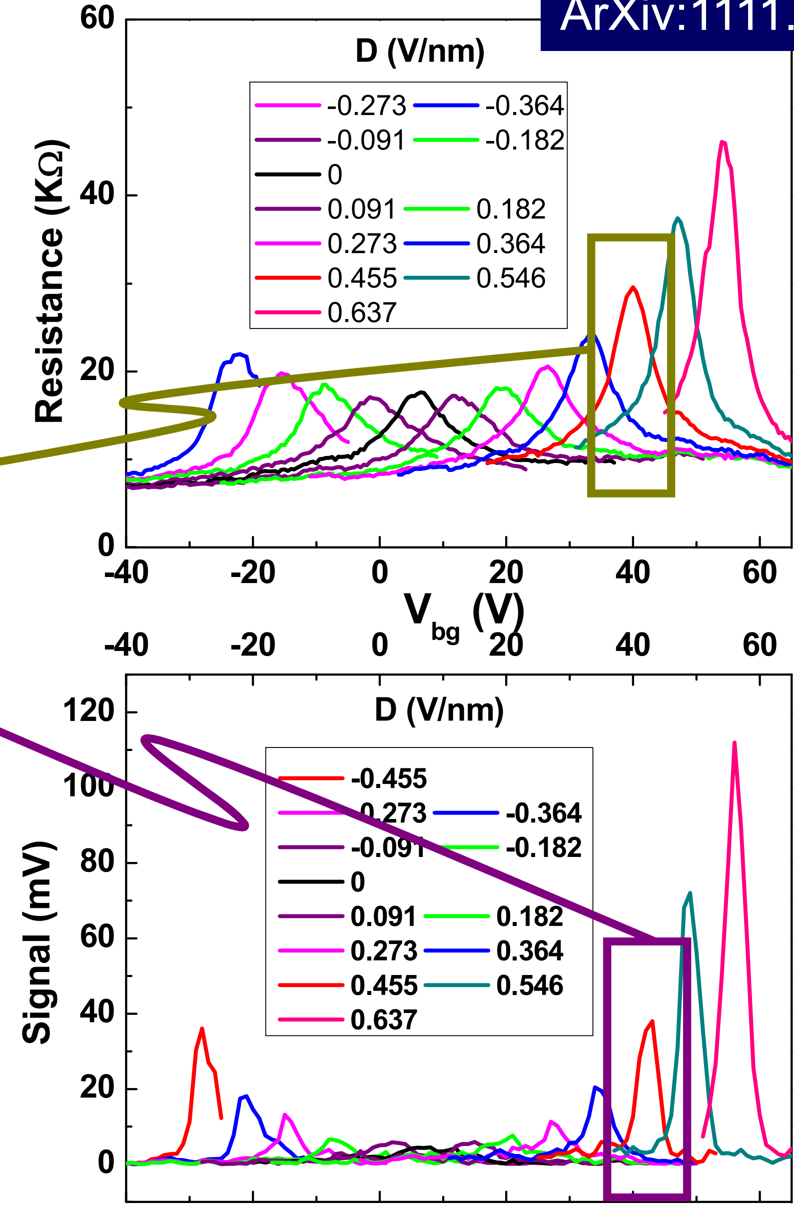




Charge transport vs. photo response

ArXiv:1111.1202





$$\Delta V = IR^2 \Delta\sigma$$

$$\Delta V \propto R^2$$

Origin of the photo-response

ArXiv:1111.1202

Photoconductive: Photo-excited carriers Δ*n* enhance conductivity Δσ = (Δ*n*)eμ:

Doesn't explain:

- **strong T dependence**
- **strong I_{dc} dependence**
- **carrier density dependence**
- **energy gap dependence**

$$\Delta R = \Delta\left(\frac{1}{\sigma}\right) = R^2 \Delta\sigma$$

Origin of the photo-response

ArXiv:1111.1202

Photoconductive: P… excited carriers Δn enhanc… ductivity $\Delta\sigma = (\Delta n)e\mu$:

Doesn't explain:

- **strong T dependence**
- **strong I_{dc} dependence**
- **carrier density dep…**
- **energy gap de… ce**

$$\ldots \Delta\left(\frac{1}{\sigma}\right) = R^2\Delta\sigma$$

Origin of the photo-response

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Photoconductive: P... excited carriers Δn enhanc... ductivity $\Delta\sigma = (\Delta n)e\mu$:

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- **carrier density dep...**
- **energy gap de... ...ce**

$$\ldots\Delta\left(\frac{1}{\sigma}\right) = R^2\Delta\sigma$$

Bolometric: Absorbed radiation heats device; temperature-dependent resistance leads to a response:

$$\frac{dR}{dP} = \frac{dR}{dT}\frac{dT}{dP} \qquad \frac{dT}{dP} \equiv R_h$$

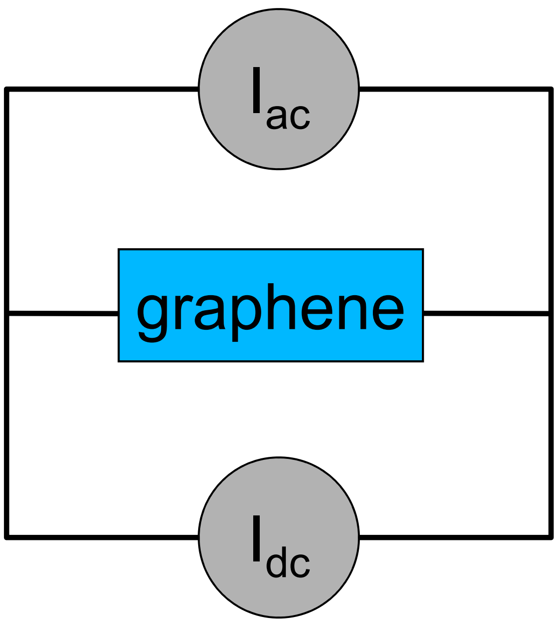
Heat resistance

Can we measure $R_h = dT/dP$ directly?

Electrical heating of graphene

$$I = I_{dc} + I_{ac}\cos(\omega t)$$

$$R = R_0 + \frac{dR}{dP} I^2 R_0 = R_0 + \frac{dR}{dP}\left(I_{dc}{}^2 + \frac{I_{ac}^2}{2} + 2 I_{dc} I_{ac}\cos(\omega t) + \frac{I_{ac}^2}{2}\cos(2\omega t) \right) R_0$$



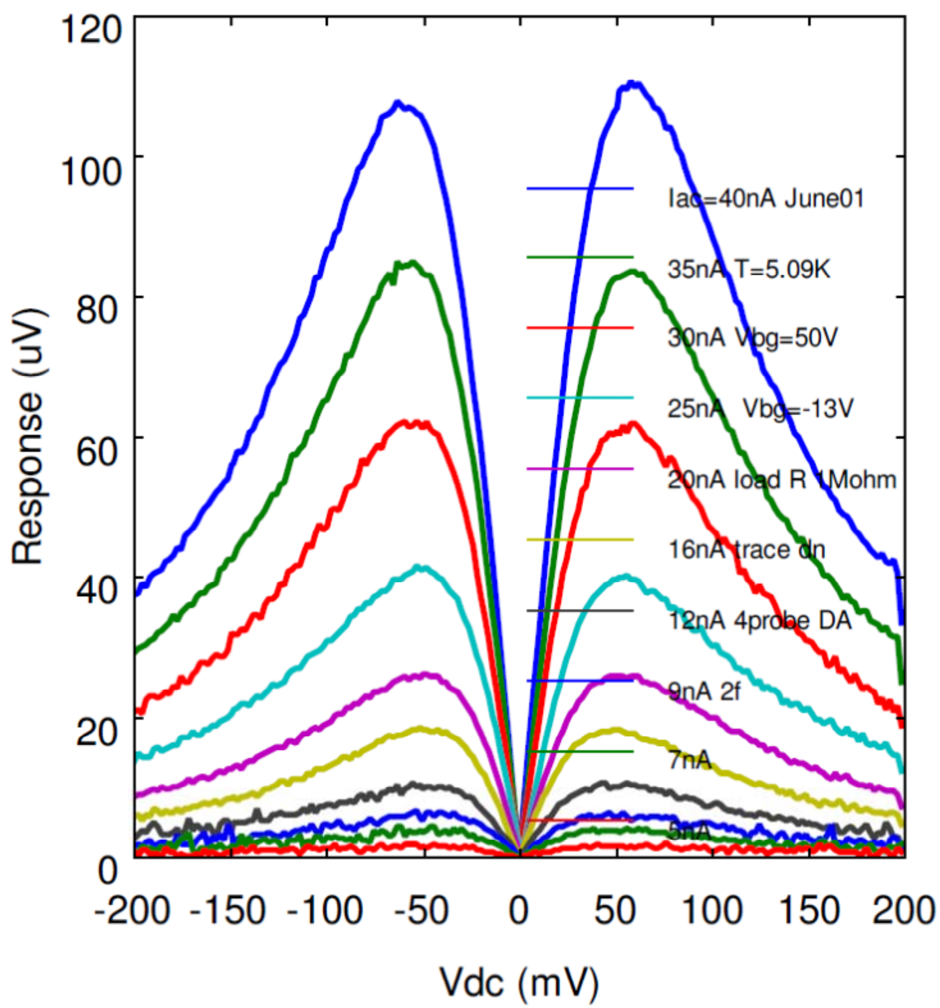
2ω signal given by:

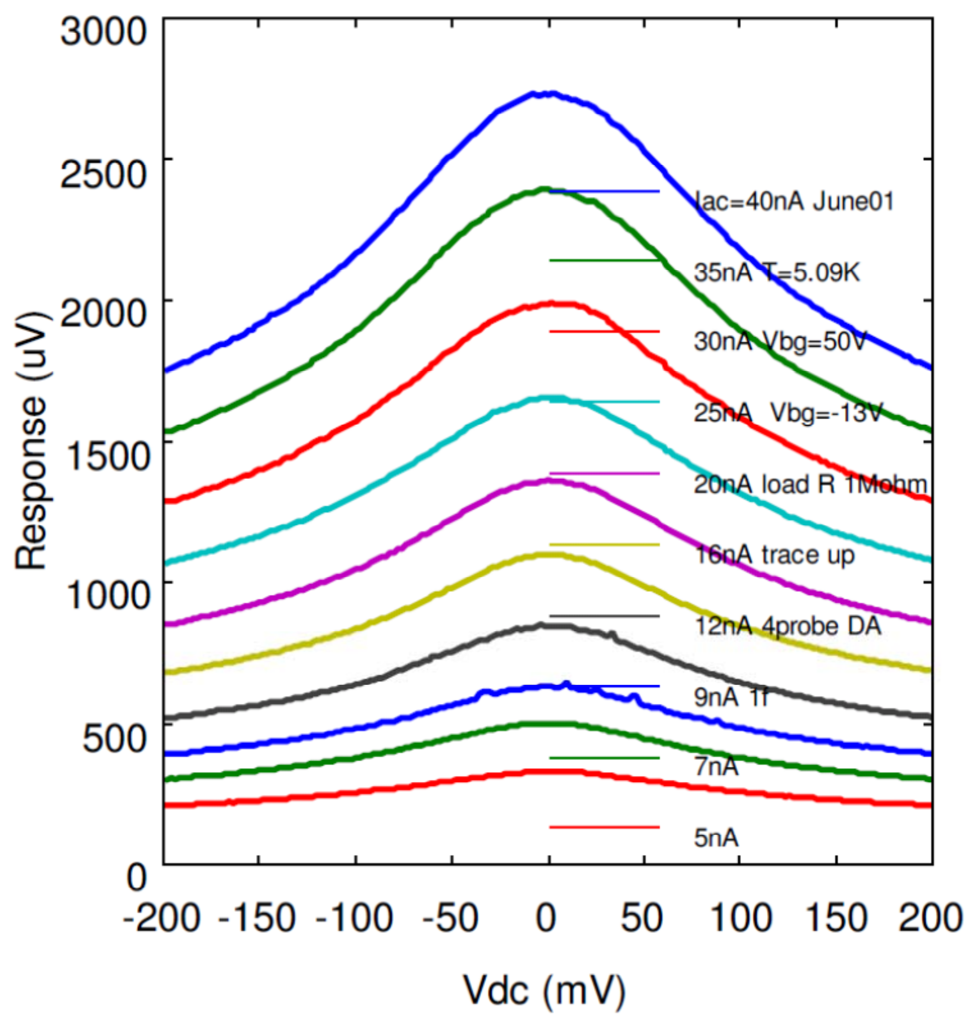
$$V(2\omega) = I_{dc}\frac{dR}{dP}\left(\frac{3}{2} I_{ac}{}^2 R_0\right)\cos 2\omega t$$

2nd harmonic signals of graphene: I_{ac} and I_{dc} dependence

$$V(2\omega) \Rightarrow \frac{dR}{dP}$$

$$V(1\omega) \Rightarrow R$$

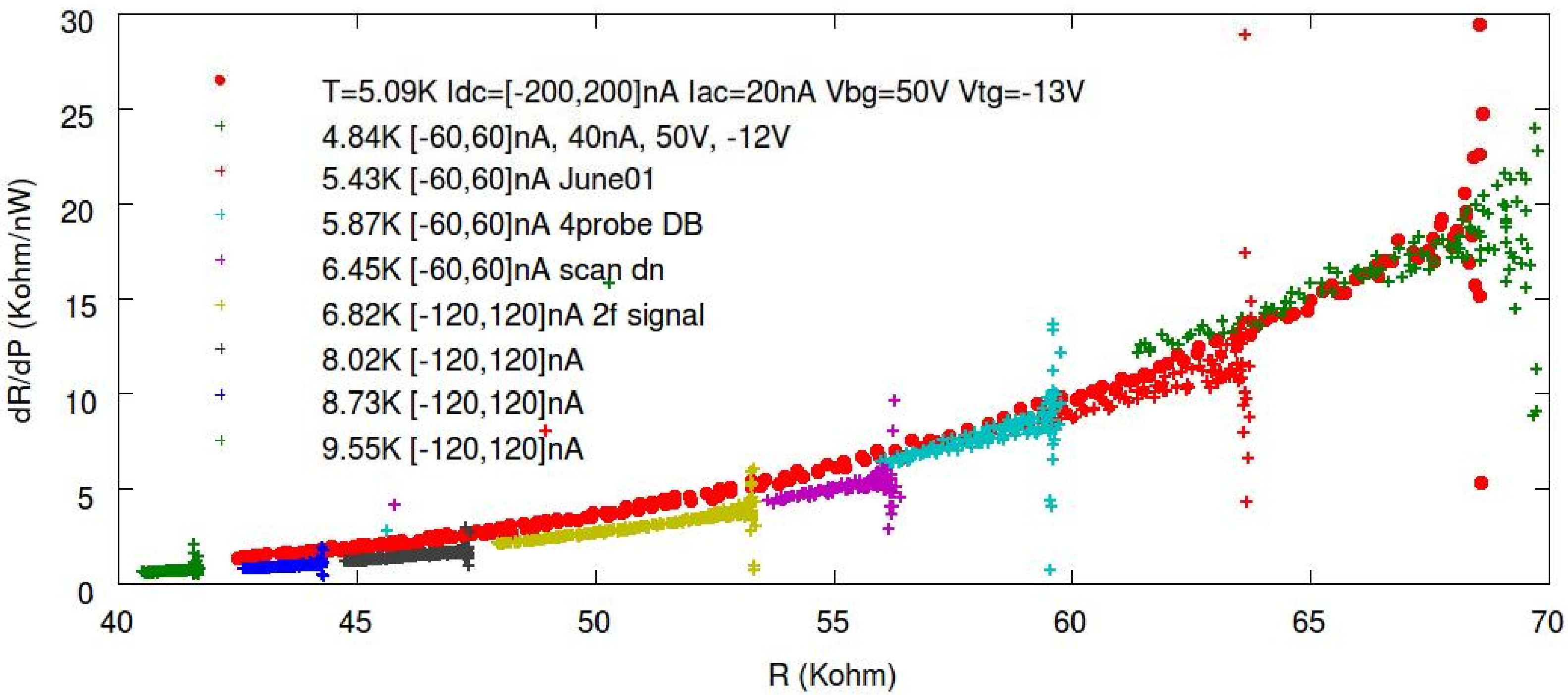




Temperature and I_{dc} dependence of 2f signal

ArXiv:1111.1202

dR/dP is a unique function of resistance R indicating that non-linearity is due to heating

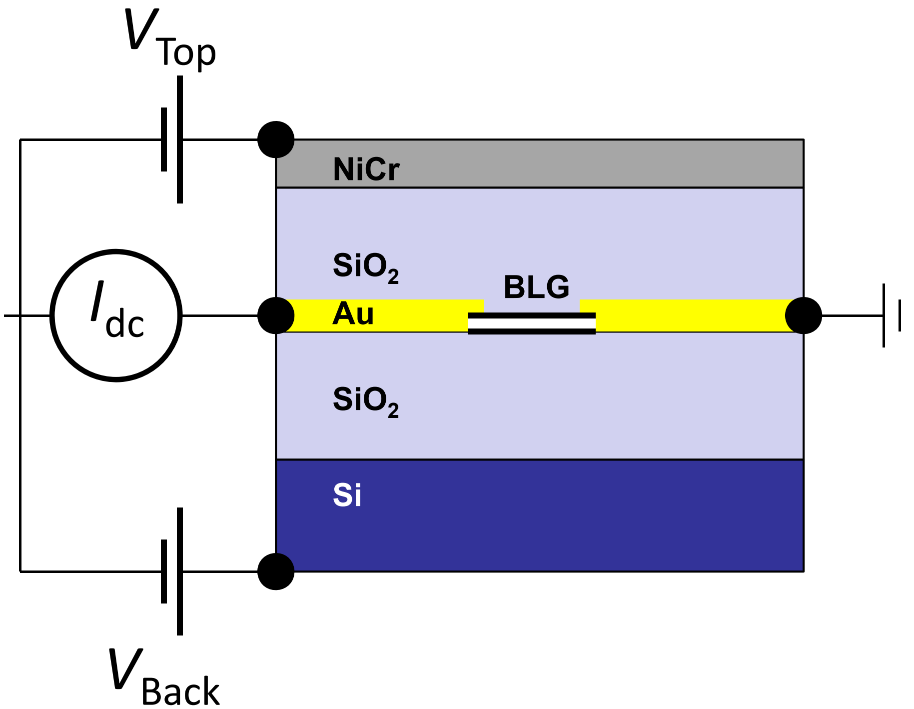


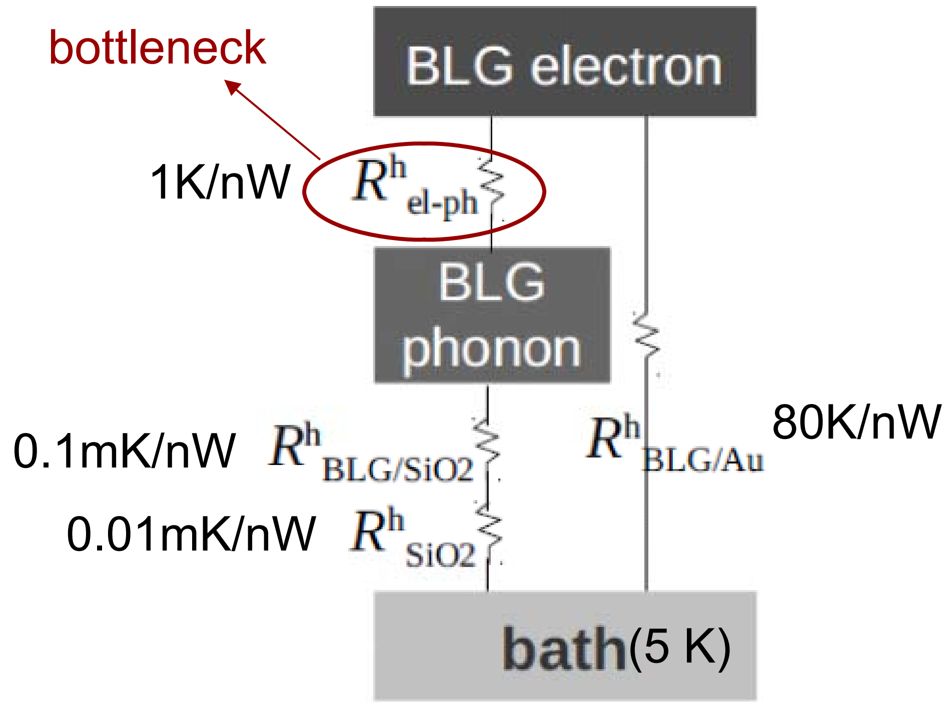
$$\frac{dR}{dP} = \frac{dR}{dT}\frac{dT}{dP} \qquad \frac{dT}{dP} \equiv R_h$$

Heat resistance

Heat conduction pathways

ArXiv:1111.1202





Assuming charge density of 10^{12} cm^{-2} , $\Theta_{BG} = 70K$

Theory: $$R^h = \frac{dT}{dP} = 0.6\left(\frac{T}{5}\right)^{-3} K/nW$$

Viljas and Heikkila PRB, 81, 245404 (2010)

Experiment: $$R^h = \frac{dT}{dP} = 2\left(\frac{T}{5}\right)^{-3.45} K/nW$$

Good agreement betw. theory and experiment
→ small electron-phonon coupling allows large bolometric effects!

Time-resolved photoresponse

ArXiv:1111.1202

Time constant: $\tau = R^h C$

$$R^h = \frac{dT}{dP} = 2\left(\frac{T}{5}\right)^{-3.45} K/nW \quad \text{(expt.)}$$

$$C = (\pi^2/3)\upsilon(E_F)k_B^2 T \approx \left(1.1\times10^{-19}\right)\times\left(\frac{T}{5}\right) \quad \text{(estimate)}$$

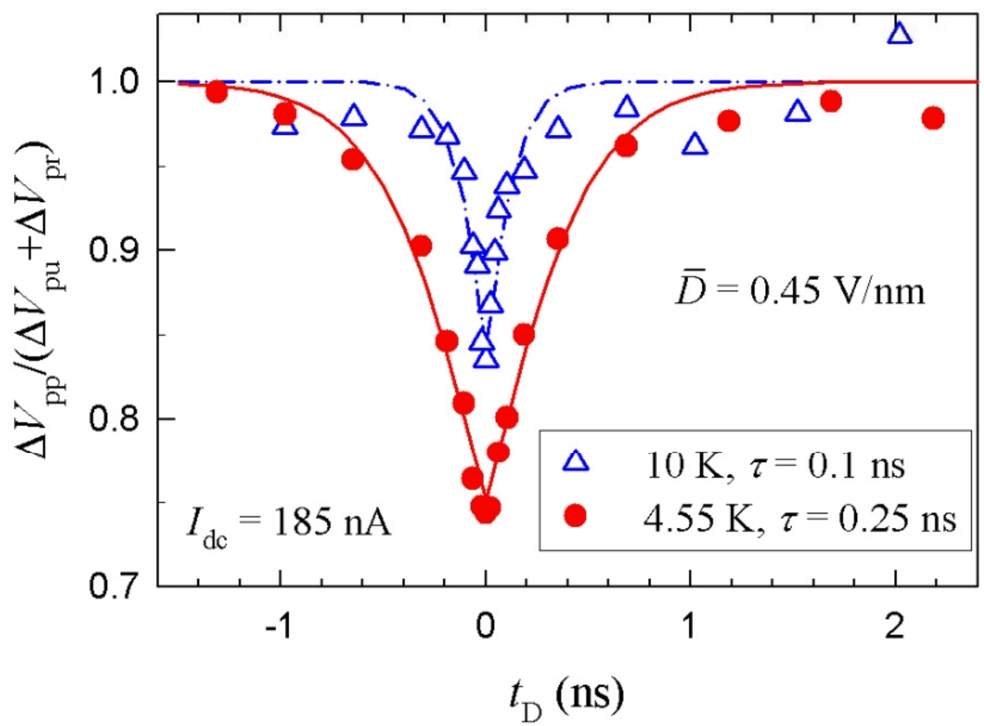
$$\tau = R^h C \approx 0.2\times(T/5)^{-2.45}\,\text{ns}$$

Pump-probe experiment:

Pump/probe in optical, detect dc

Non-linearity gives reduced response when pump/probe are coincident

Excellent agreement with estimate!



Photoresponse time agrees with estimate of *e-ph* relaxation time from DC measurements

Graphene Hot Electron Bolometer: results and prospects

Current device: Graphene hot electron bolometer @ T = 5K:

$NEP_{phonon} = 2.6 \times 10^{-16}$ W/Hz$^{1/2}$

$NEP_{Nyquist} = 3.3 \times 10^{-14}$ W/Hz$^{1/2}$

$\tau = 0.2$ ns

Compare:

Superconducting bolometer @ T = 4K:

NEP = 10^{-13} W/Hz$^{1/2}$

$\tau = 1$ ms

[Skidmore et al. *APL* **82**, 469 (2003)]

Graphene HEB @ T = 100 mK:

$NEP_{phonon} = 6 \times 10^{-21}$ W/Hz$^{1/2}$

$NEP_{Nyquist}$ = ?

$\tau = 3$ μs

Future improvements:

Proximity-induced superconductivity (transition-edge detection) in graphene

Cleaner devices (stronger activated behavior) using BN substrates

Lower impedance devices/higher quantum efficiency using multilayer graphene

Thank you!

Correlated charged impurity scattering in graphene
Jun Yan, Michael S. Fuhrer
Physical Review Letters **107**, 206601 (2011)

Imaging charge disorder of bare SiO_2
K. Burson, M.S. Fuhrer, W. G. Cullen, in preparation

Charge transport in dual gated bilayer graphene with Corbino geometry
Jun Yan and Michael S. Fuhrer
Nano Letters **10**, 4521 (2010)

Dual-gated bilayer graphene hot electron bolometer
J. Yan, M.-H. Kim, J.A. Elle, A.B. Sushkov, G.S. Jenkins, H.M. Milchberg, M.S. Fuhrer, and H.D. Drew
arXiv:1111.1202

Prof. Michael S. Fuhrer
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Prof. Sankar Das Sarma
Dr. Shaffique Adam (now @ NIST), **Dr. Euyheon Hwang**, Prof. Enrico Rossi (now @ Wm.&Mary), Wang-Kong Tse, Dr. Dimi Culcer, **Qiuzi Li**

Prof. Dennis Drew
Dr. Greg Jenkins, Dr. Andrei Sushkov, Dr. Myoung-Hwan Kim

Prof. Howard Milchberg
Jennifer Elle