

# Electrical Characterization of Materials: II

Mark Lundstrom

Electrical and Computer Engineering  
Purdue University  
West Lafayette, IN USA

# Coupled charge and heat current equations

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electrical current:

$$\mathcal{E} = \rho J + S \frac{dT}{dx}$$

heat current (electronic):

$$J_Q = \pi J - \kappa_e \frac{dT}{dx}$$

heat current (lattice):

$$q_x = -\kappa_L \left( \frac{dT}{dx} \right)$$

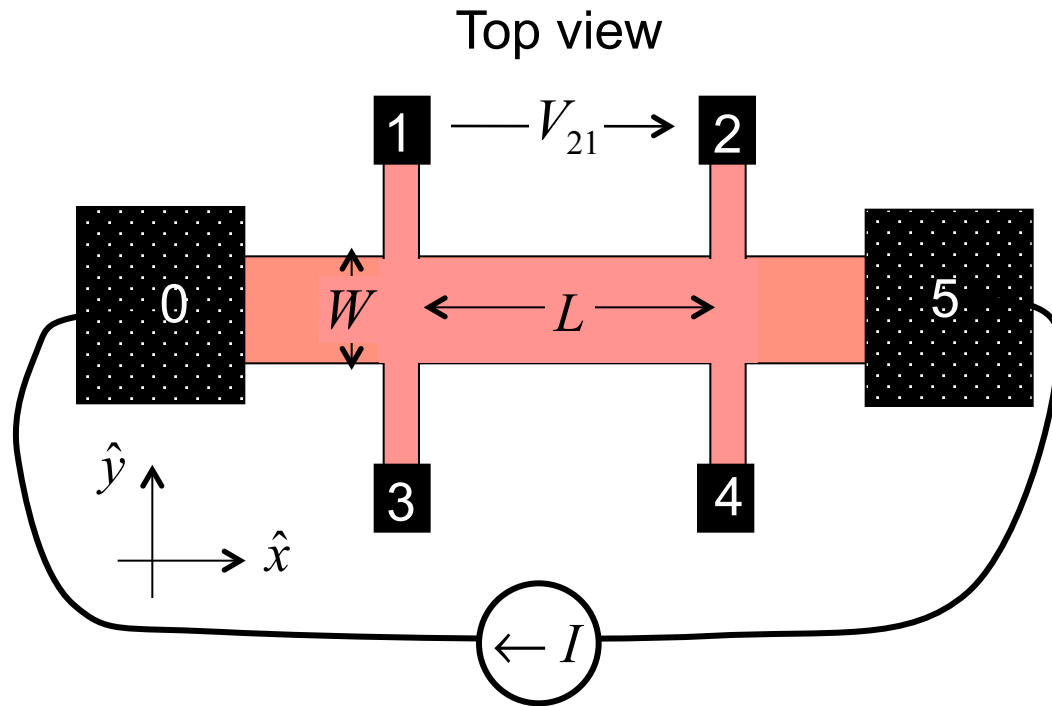
We have been discussing how to measure conductivity (and carrier density and mobility), and we briefly the Seebeck coefficient.

# Outline

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1. **Review**
2. Temperature-dependent measurements
3. Errors in Hall effect measurements
4. Graphene: a case study
5. Summary

# Hall bar geometry



$$R_H = \frac{-V_H}{I_x B_z} = \frac{1}{(-q)(n_S/r_H)}$$

Hall coefficient

$$\frac{V_{21}}{I} = \rho_S \frac{L}{W}$$

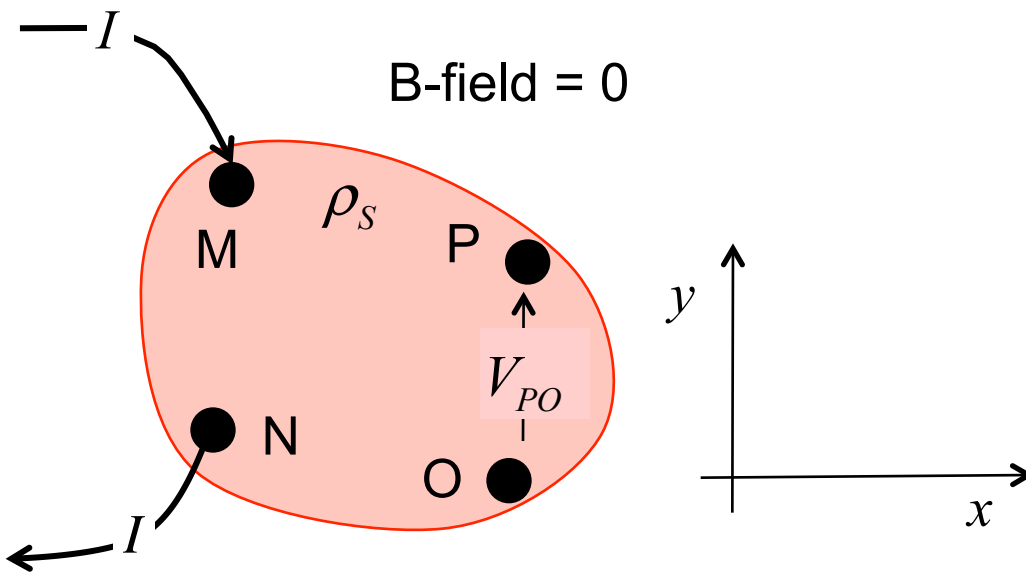
$$\mu_H = r_H \mu_n = \frac{1}{n_H q \rho_S}$$

$$r_H \equiv \frac{\langle\langle \tau_m^2 \rangle\rangle}{\langle\langle \tau_m \rangle\rangle^2}$$

“Hall factor”

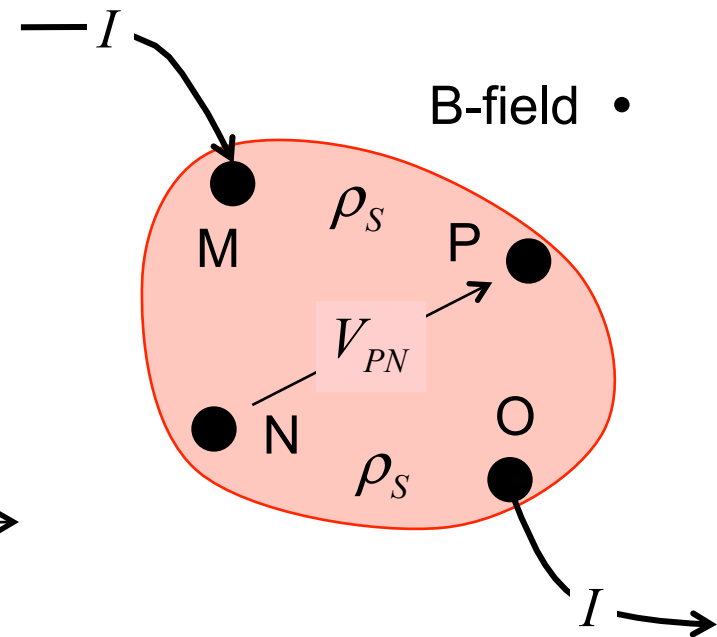
# van der Pauw geometry

## Resistivity



- 1) force a current in  $M$  and out  $N$
- 2) measure  $V_{PO}$
- 3)  $R_{MN, OP} = V_{PO} / I$  related to  $\rho_S$

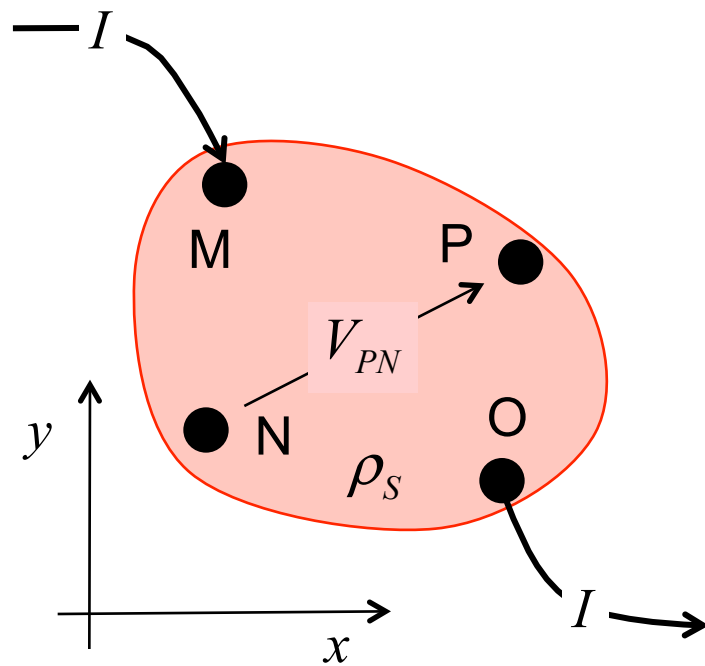
## Hall effect



- 1) force a current in  $M$  and out  $O$
- 2) measure  $V_{PN}$
- 3)  $R_{MO, NP} = V_{PN} / I$  related to  $V_H$

# van der Pauw approach: Hall effect

## Hall effect



$$\vec{J}_n = nq\mu_n \vec{E} - (\sigma_n \mu_n r_H) \vec{E} \times \vec{B}$$

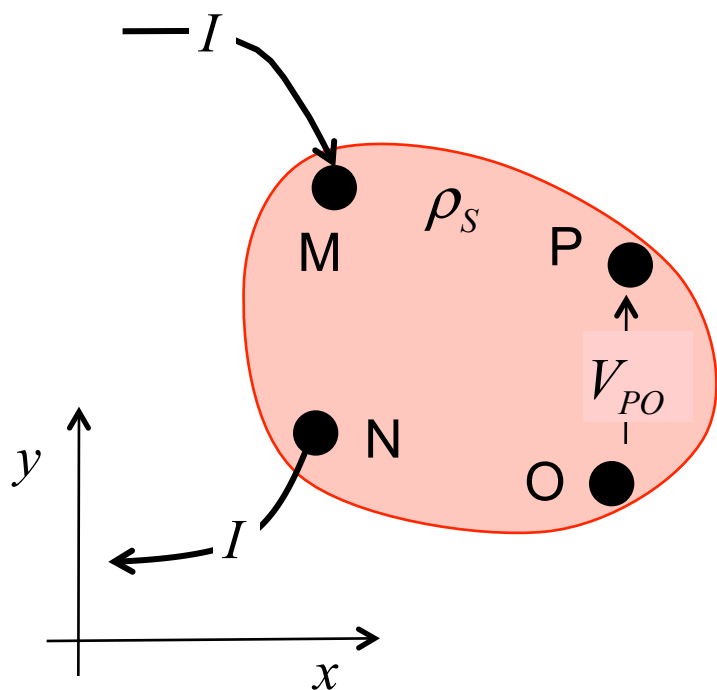
$$V_H \equiv \frac{1}{2} [V_{PN}(+B_z) - V_{PN}(-B_z)]$$

$$V_H = \rho_n \mu_H B_z I$$

**So we can do Hall effect measurements on such samples.**

See Lundstrom, *Fundamentals of Carrier Transport*, 2<sup>nd</sup> Ed., Sec. 4.7.1.

# van der Pauw approach: resistivity



***So we can do resistivity measurements on such samples.***

$$R_{MN,OP} = \frac{\rho_S}{\pi} \ln \left( \frac{(a+b)(b+c)}{b(a+b+c)} \right)$$

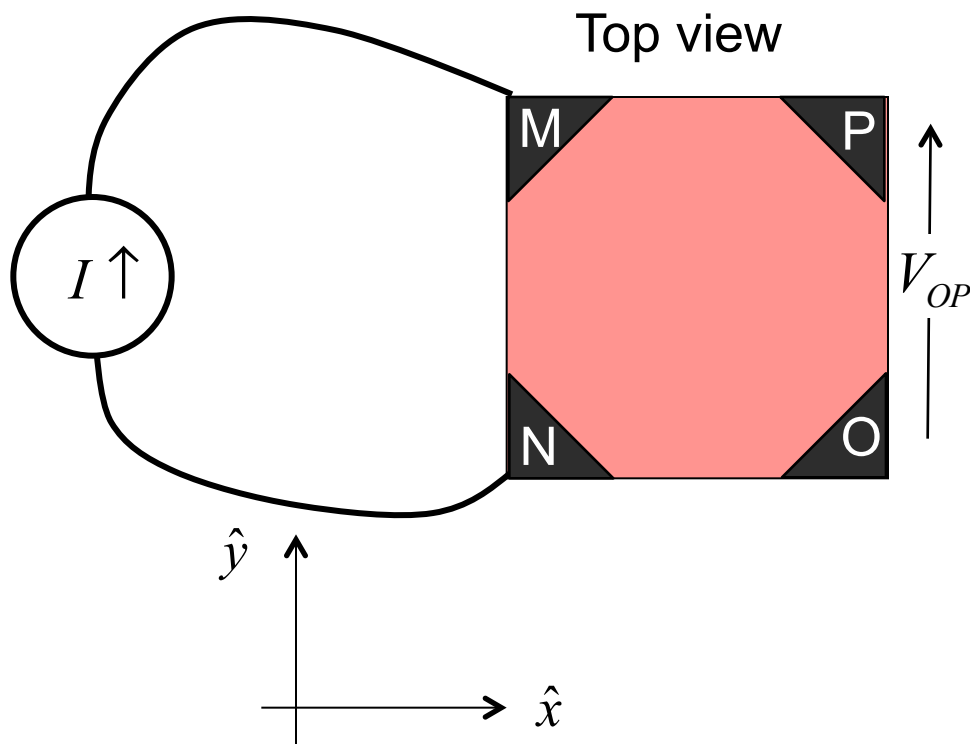
$$R_{NO,PM} = \frac{\rho_S}{\pi} \ln \left( \frac{(a+b)(b+c)}{ac} \right)$$

it can be shown that:

$$e^{-\frac{\pi}{\rho_S} R_{MN,OP}} + e^{-\frac{\pi}{\rho_S} R_{NO,PM}} = 1$$

Given two measurements of resistance, this equation can be solved for the sheet resistance.

# van der Pauw technique: regular sample



$$e^{-\frac{\pi}{\rho_S} R_{MN,OP}} + e^{-\frac{\pi}{\rho_S} R_{NO,PM}} = 1$$

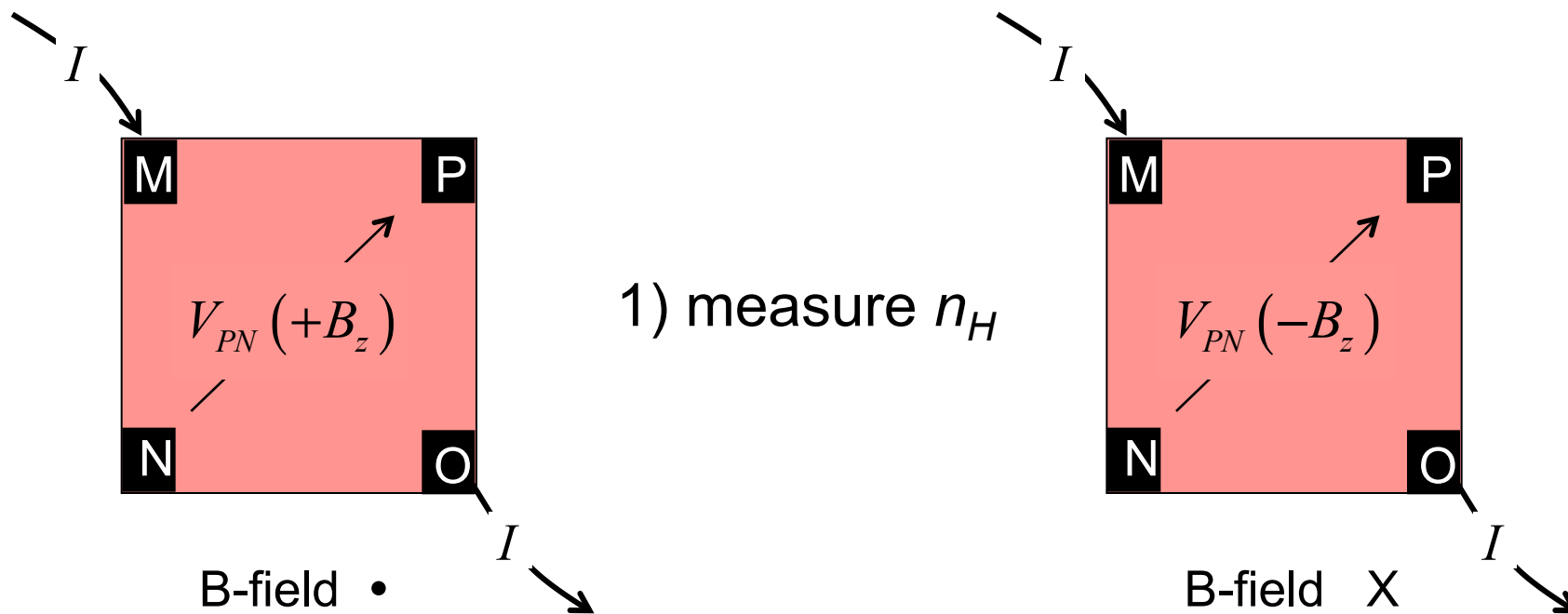
$$R_{MN,OP} = R_{NO,PM} = \frac{V}{I}$$

$$\rho_S = \frac{\pi}{\ln 2} \frac{V}{I}$$

Force  $I$  through two contacts, measure  $V$  between the other two contacts.

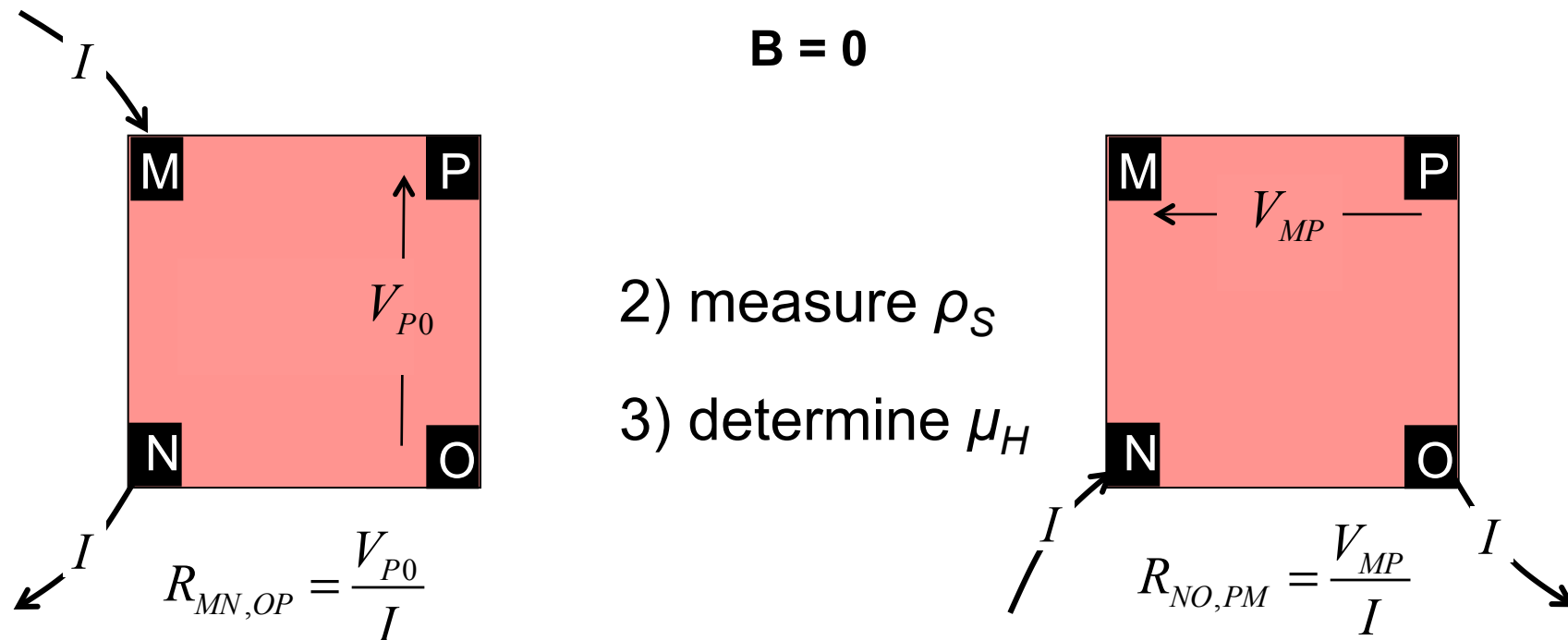


# van der Pauw technique: summary



$$V_H = \frac{1}{2} [V_{PN}(+B_Z) - V_{PN}(-B_Z)] = \frac{r_H}{qn_S} B_z I = \frac{B_z I}{qn_H}$$

# van der Pauw technique: summary



$$e^{-\frac{\pi}{\rho_S} R_{MN,OP}} + e^{-\frac{\pi}{\rho_S} R_{NO,PM}} = 1 \quad \sigma_S = n_S q \mu_n = \frac{n_S}{r_H} q r_H \mu_n = n_H q \mu_H$$

# outline

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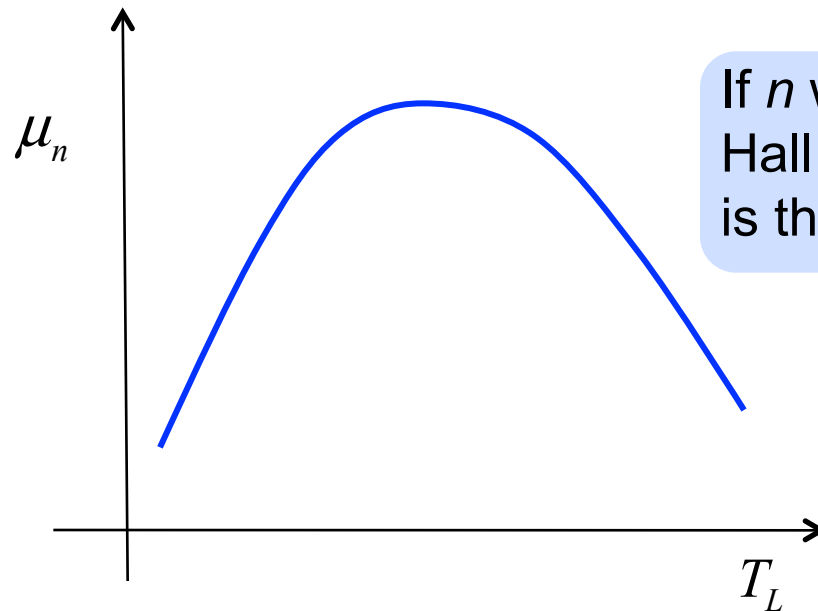
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# Temperature-dependent measurements

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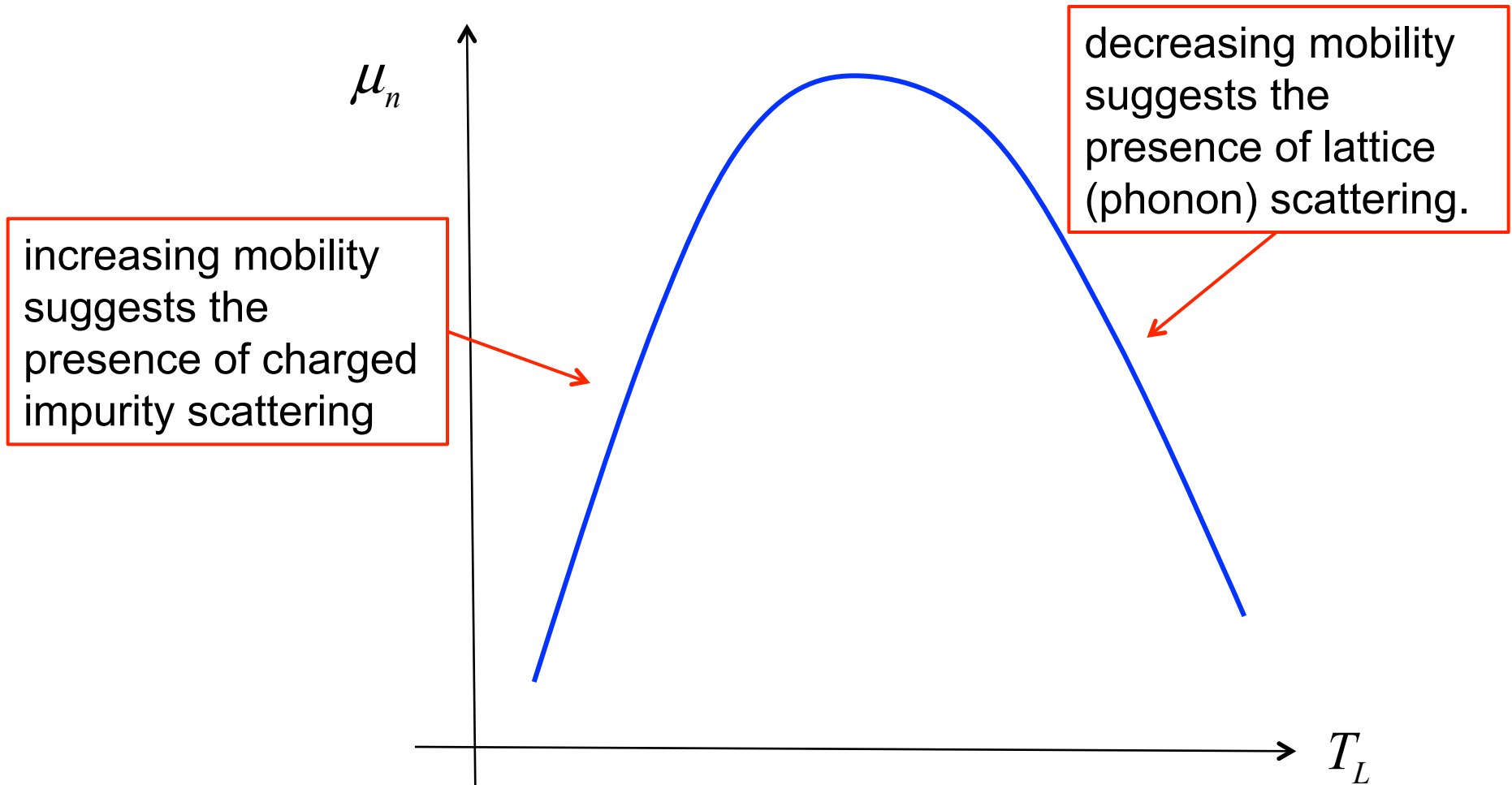
It is common practice to measure the temperature-dependent conductivity.

Assuming that the carrier density is known (or can be measured), a mobility is then extracted from:  $\sigma = n q \mu_n$



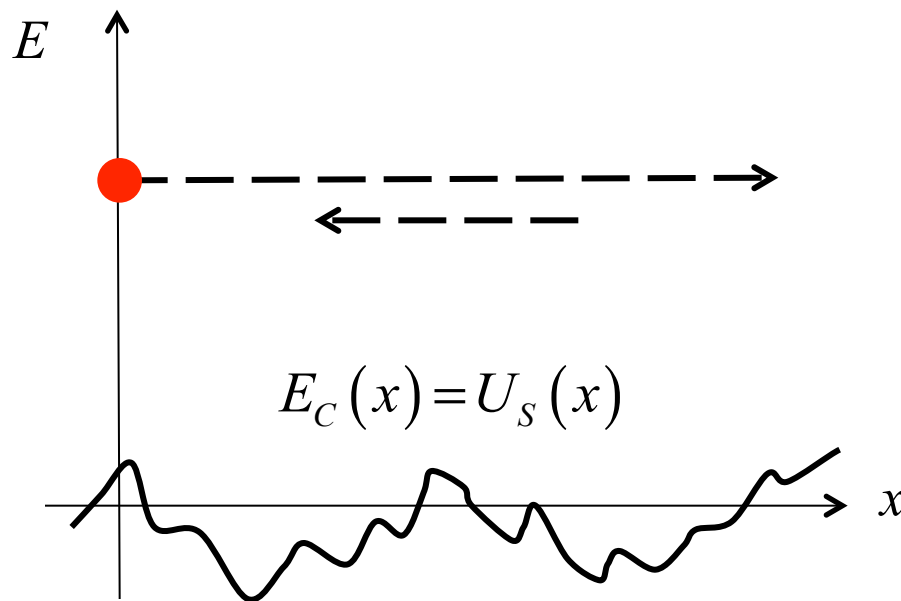
If  $n$  was measured by Hall effect, then mobility is the Hall mobility.

# Interpretation



# Charged impurity scattering

$$\tau(E) \uparrow \text{ as } E \uparrow$$



Random charges introduce random fluctuations in  $E_C$ , which act as scattering centers.

High energy electrons don't "see" these fluctuations and are not scattered as strongly.

Average carrier energy  $\sim k_B T_L$ .

**Mobility should increase with increasing temperature.**

# Lattice (phonon) scattering

$$\frac{1}{\tau(E)} \propto n_{ph}$$

$$n_{ph} = \frac{1}{e^{\hbar\omega/k_B T_L} - 1}$$

$$n_{ph} \uparrow \text{ as } T_L \uparrow$$

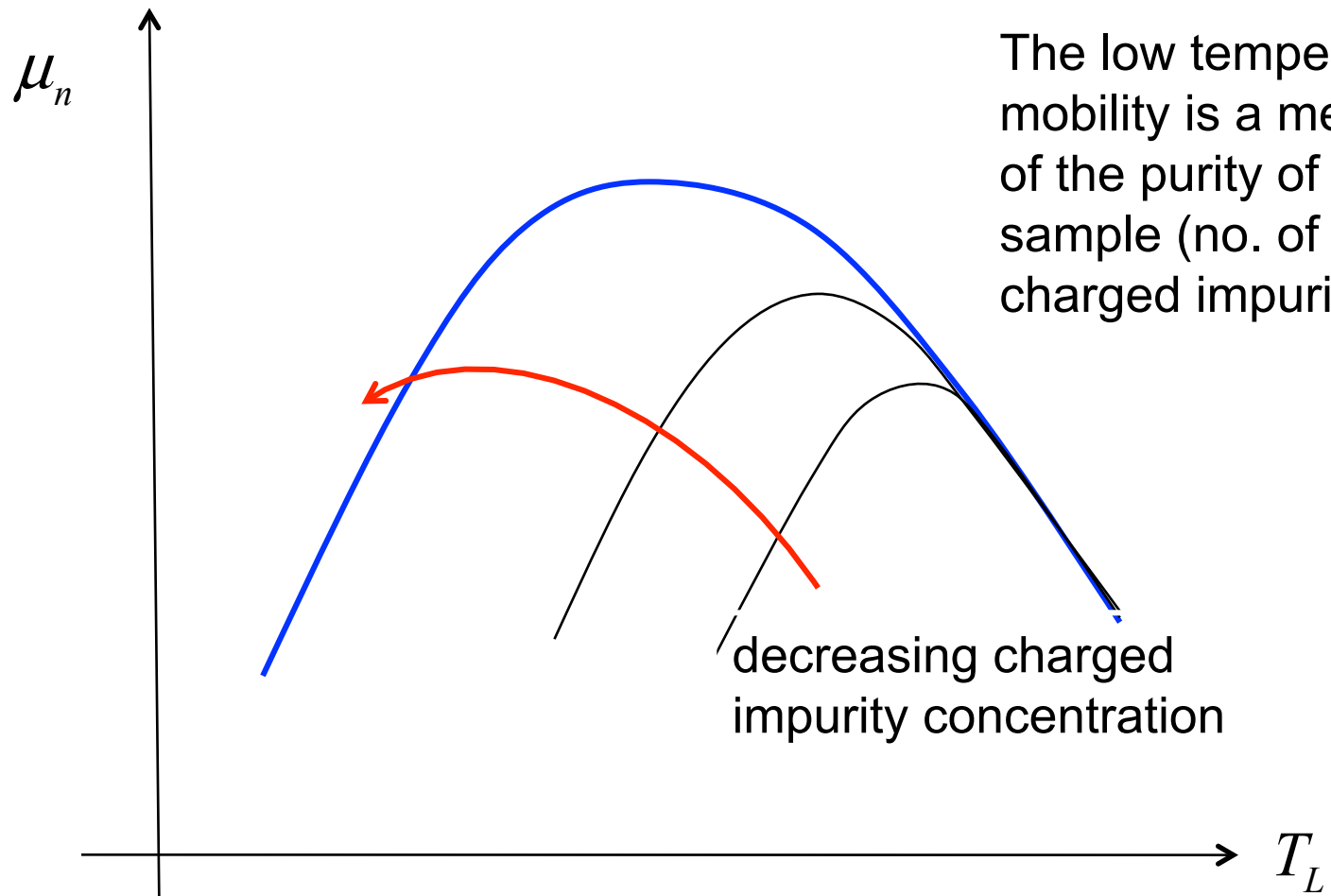
Carrier scattering rate is proportional to the number of phonons.

Phonon occupation number given by the Bose-Einstein distribution.

Number of phonons increases as temperature increase.  
Scattering time decreases.

**Mobility should decrease with increasing temperature.**

# Mobility vs. temperature



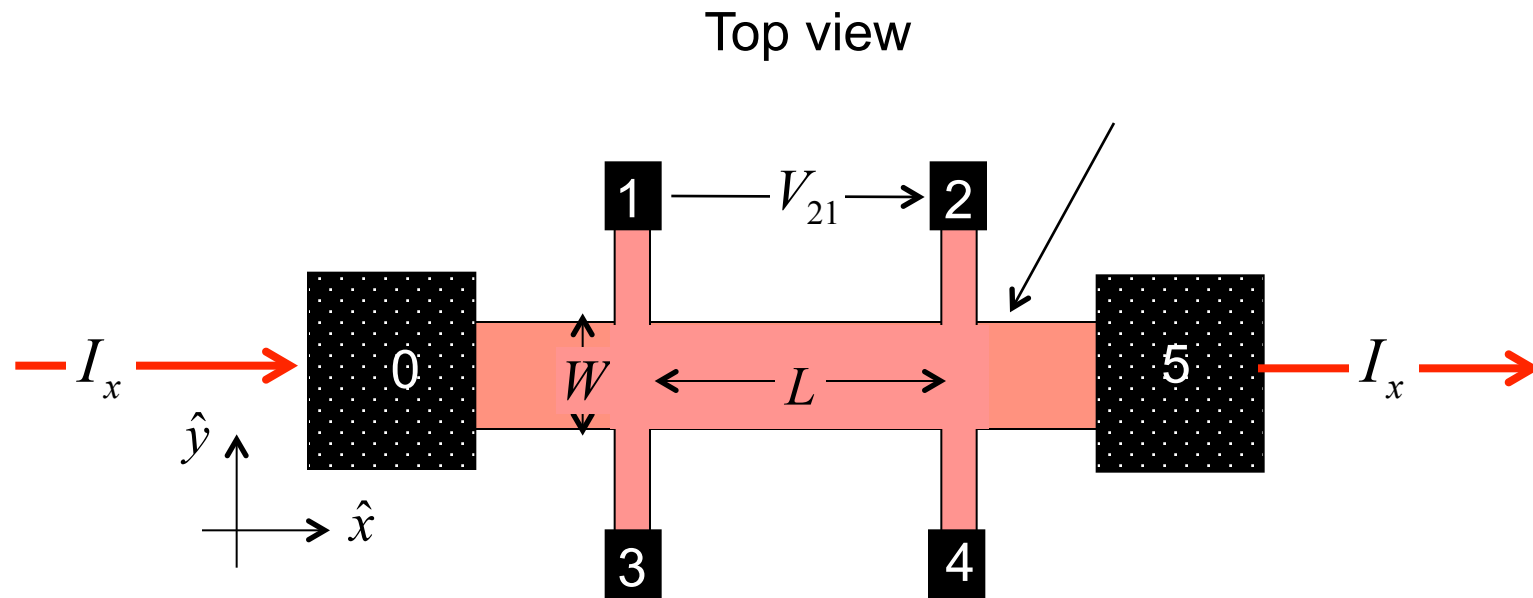


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# Hall effect measurements (errors)



We have assumed isothermal conditions to compute the Hall voltage, but we expect Peltier cooling at contact 0 and Peltier heating at contact 1. If the sample is not isothermal, how does the Hall voltage change?

# Magnetoconductivity tensor

$$\begin{pmatrix} J_{nx} \\ J_{ny} \end{pmatrix} = \begin{pmatrix} \sigma_S & -\sigma_S \mu_H B_z \\ +\sigma_S \mu_H B_z & \sigma_S \end{pmatrix} \begin{pmatrix} \mathcal{E}_x \\ \mathcal{E}_y \end{pmatrix}$$

$$J_{ni} = \sum_j \sigma_{ij}(B_z) \mathcal{E}_j \quad \begin{pmatrix} \sigma_{11} & \sigma_{12} \\ \sigma_{21} & \sigma_{22} \end{pmatrix} = \begin{pmatrix} \sigma_S & -\sigma_S \mu_H B_z \\ +\sigma_S \mu_H B_z & \sigma_S \end{pmatrix}$$

$$J_{ni} = \sigma_{ij}(B_z) \mathcal{E}_j \quad (\text{summation convention})$$

$$J_i = \sigma_S \mathcal{E}_i - \sigma_S \mu_H \varepsilon_{ijk} \mathcal{E}_j B_k$$

$$\begin{aligned} \varepsilon_{ijk} &= +1(i, j, k \text{ cyclic}) \\ &= -1(i, j, k \text{ anti-cyclic}) \\ &= 0(\text{otherwise}) \end{aligned}$$

## Recall...

$$\mathcal{E}_i = \rho_{ij}(\vec{B}) J_j + S_{ij}(\vec{B}) \partial_j T_L$$

$$J_i^Q = \pi_{ij}(\vec{B}) J_j - \kappa_{ij}^e(\vec{B}) \partial_j T_L$$

$$\rho_{ij}(\vec{B}) = \rho_0 + \rho_0 \mu_H \varepsilon_{ijk} B_k + \dots$$

$$S_{ij}(\vec{B}) = S_0 + S_1 \varepsilon_{ijk} B_k + \dots$$

$$\pi_{ij}(\vec{B}) = \pi_0 + \pi_1 \varepsilon_{ijk} B_k + \dots$$

$$\kappa_{ij}^e(\vec{B}) = \kappa_0^e + \kappa_1 \varepsilon_{ijk} B_k + \dots$$

For parabolic energy bands

# Nernst effect

Assume that there is a temperature gradient in the x-direction. How is the electric field (Hall voltage) affected?)

$$\mathcal{E}_i = \rho_{ij}(\vec{B})J_j + S_{ij}(\vec{B})\partial_j T_L$$

$$\rho_{ij}(\vec{B}) = \rho_0 + \rho_0 \mu_H \epsilon_{ijk} B_k + \dots$$

$$S_{ij}(\vec{B}) = S_0 + S_1 \epsilon_{ijk} B_k + \dots$$

$$\mathcal{E}_y = \rho_0 J_y + \rho_0 \mu_H \epsilon_{yzx} B_z J_x + S_0 \partial_y T_L + S_1 \epsilon_{yzx} B_z \partial_x T_L$$

$$\mathcal{E}_y = +\rho_0 \mu_H \epsilon_{yxz} B_z J_x + S_1 \epsilon_{yxz} B_z \partial_x T_L$$

$$\mathcal{E}_y = -\rho_0 \mu_H B_z J_x - S_1 B_z \partial_x T_L$$

## Nernst voltage

Reverse direction of  $B_z$  and  $J_x$  and average results to eliminate.

## Other effects

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Other “thermomagnetic effects” such as the Ettingshausen and Righi-Leduc effects also occur and affect the measured Hall voltage. See Lundstrom, Chapter 4, Sec. 4.6.2 for a discussion.

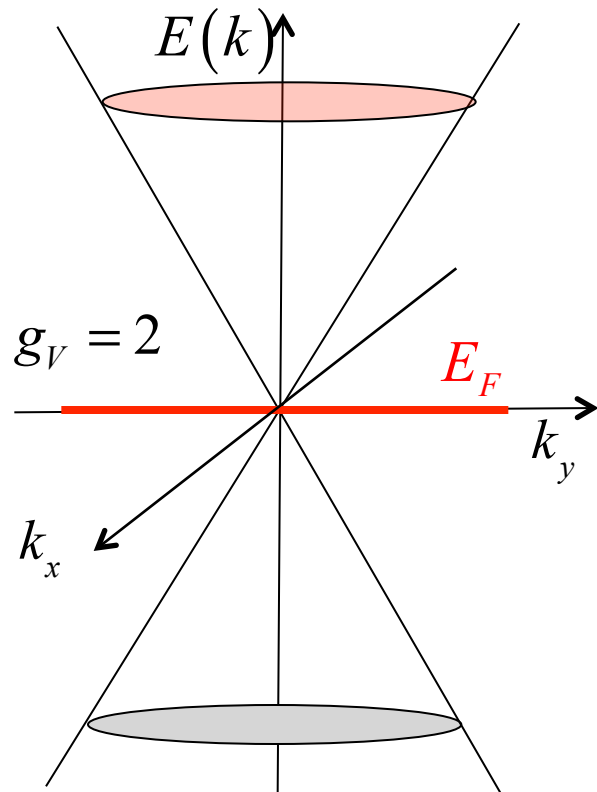
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Taken from: 2011 NCN Summer School, “Near-equilibrium Transport, Lecture 10: Case study-Near-equilibrium Transport in Graphene,” <http://nanohub.org/resources/11873>

# Graphene



$$E(k) = \pm \hbar v_F k = \pm \hbar v_F \sqrt{k_x^2 + k_y^2}$$

$$v(k) = v_F \approx 1 \times 10^8 \text{ cm/s}$$

$$D(E) = 2|E| / \pi \hbar^2 v_F^2$$

$$n_S(E_F) = \int_0^\infty D(E) f_0(E) dE \approx \frac{E_F^2}{\pi \hbar^2 v_F^2}$$

$$M(E) = W \frac{2|E|}{\pi \hbar v_F}$$

$$\sigma_S(0\text{K}) = \frac{2q^2}{h} \lambda_{app} \left( \frac{2E_F}{\pi \hbar v_F} \right)$$

$$\frac{1}{\lambda_{app}} = \frac{1}{\lambda(E_F)} + \frac{1}{L}$$



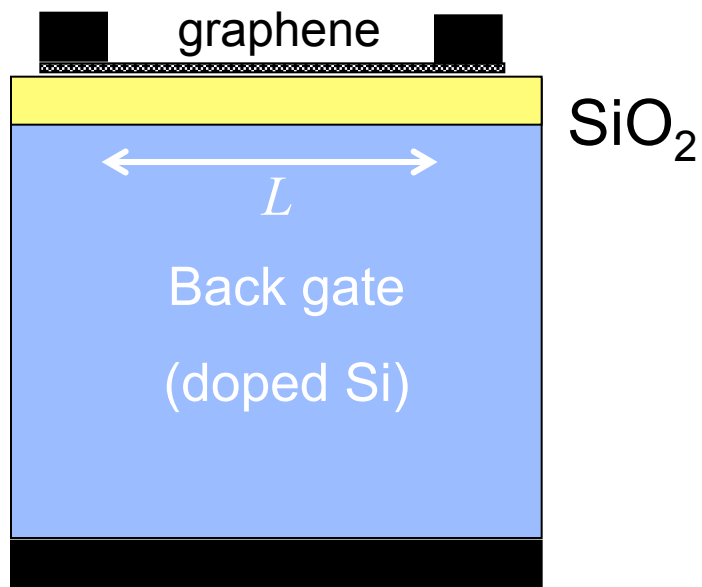
# Gate-modulated conductance in graphene

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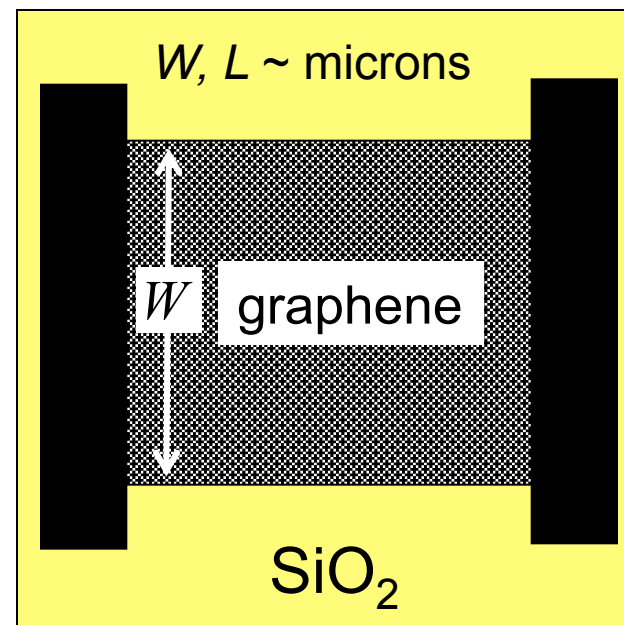
- 1) The location of the Fermi level (or equivalently the carrier density) is experimentally controlled by a “gate.”
- 2) In a typical experiments, a layer of graphene is placed on a layer of  $\text{SiO}_2$ , which is on a doped silicon substrate. By changing the potential of the Si substrate (the “back gate”), the potential in the graphene can be modulated to vary  $E_F$  and, therefore,  $n_S$ .

## Experimental structure (2-probe)

(4-probe techniques are used to eliminate series resistance and for Hall effect measurements.)



Side

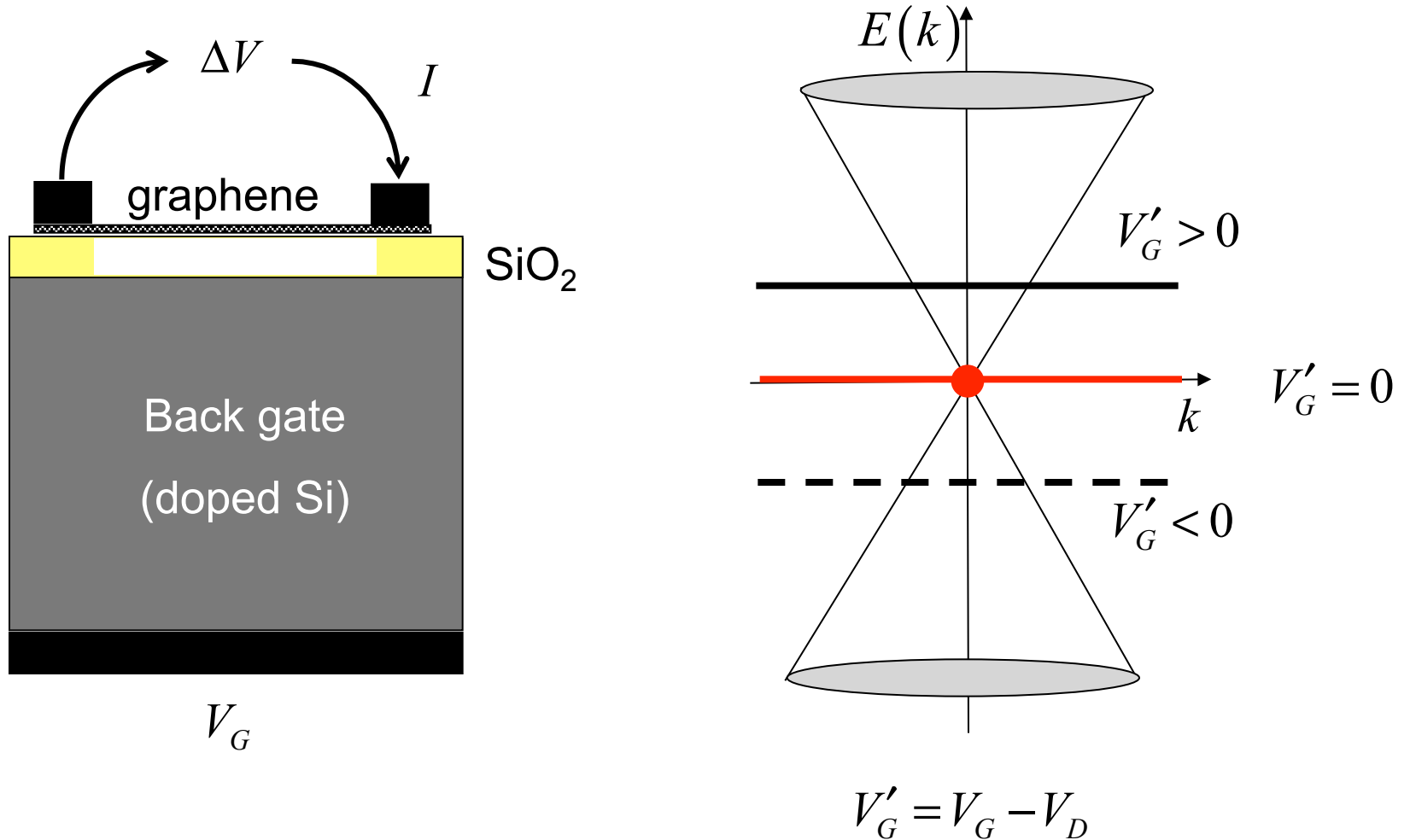


Top view

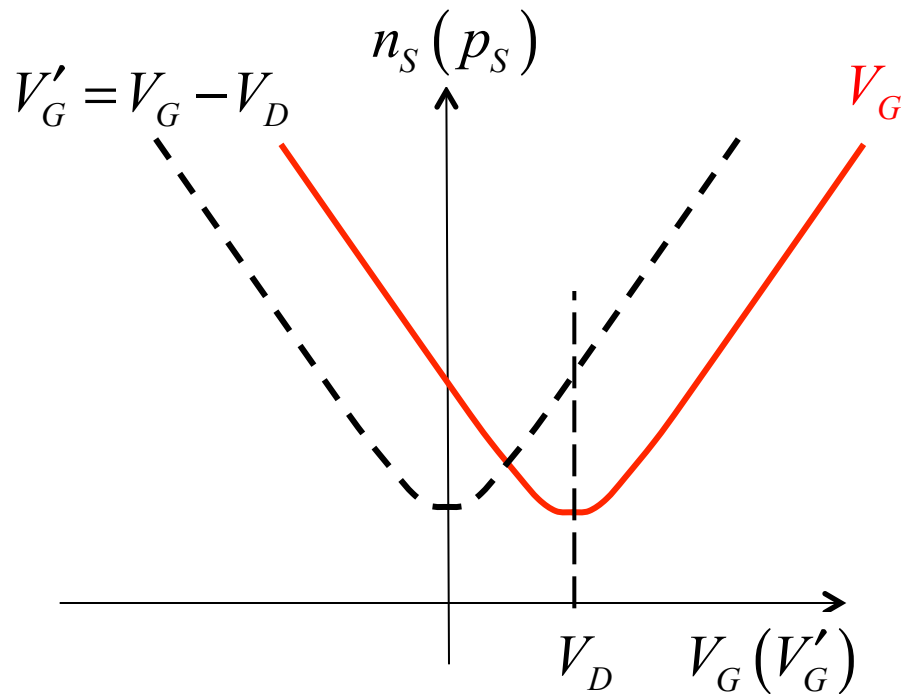
Typically, Cr/Au or Ti/Au are used for the metal contacts.

The thickness of SiO<sub>2</sub> is typically 300nm or 90nm, which makes it possible to see a single layer of graphene.

# Using a gate voltage to change the Dirac point (or $E_F$ )



# Gate voltage - carrier density relation



If the oxide is not too thin (so that the quantum capacitance of the graphene is not important), then

$$qn_S = C_{ins} V'_G$$

$$C_{ins} = \frac{\epsilon_{ins}}{t_{ins}}$$

# Sheet conductance vs. $V_G$

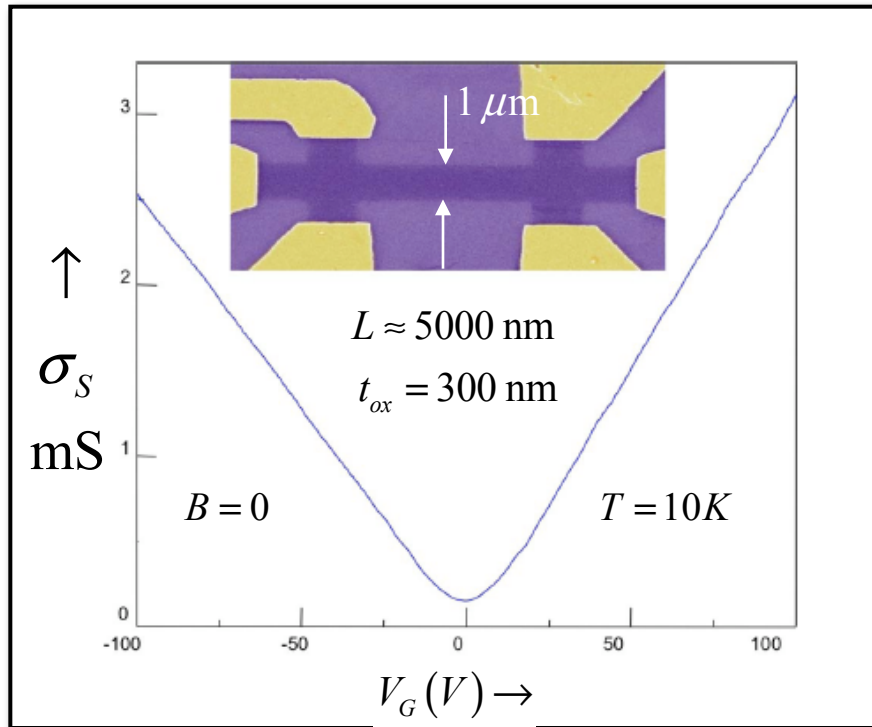


Fig. 30 in A. H. Castro, et al., "The electronic properties of graphene," Rev. of Mod. Phys., **81**, 109, 2009.

$$G = \sigma_S W/L$$

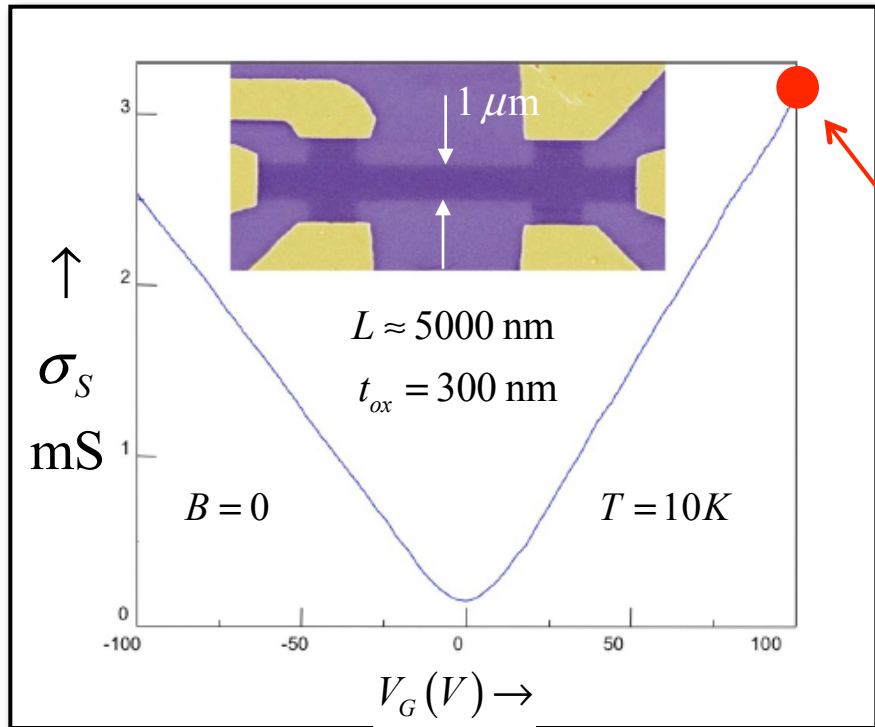
$$\sigma_S(E_F) \approx \frac{2q^2}{h} \lambda_{app}(E_F) \left( \frac{2E_F}{\pi \hbar v_F} \right)$$

$$n_S = C_{ox} V_G \approx \frac{1}{\pi} \left( \frac{E_F}{\hbar v_F} \right)^2$$

$$\lambda_{app}(E_F) = \frac{\sigma_S / (2q^2/h)}{2\sqrt{n_S/\pi}}$$

$$(T_L = 0 \text{ K})$$

## Mean-free-path ( $V_G = 100V$ )



$$\sigma_S \approx 3.0 \text{ mS}$$

$$n_S \approx 7.1 \times 10^{12} \text{ cm}^{-2}$$

$$E_F \approx 0.3 \text{ eV}$$

$$\lambda_{app} (0.3 \text{ eV}) \approx 130 \text{ nm}$$

$$\lambda(0.3 \text{ eV}) \ll L$$

Fig. 30 in A. H. Castro, et al., "The electronic properties of graphene," Rev. of Mod. Phys., **81**, 109, 2009.

## Mean-free-path ( $V_G = 50V$ )

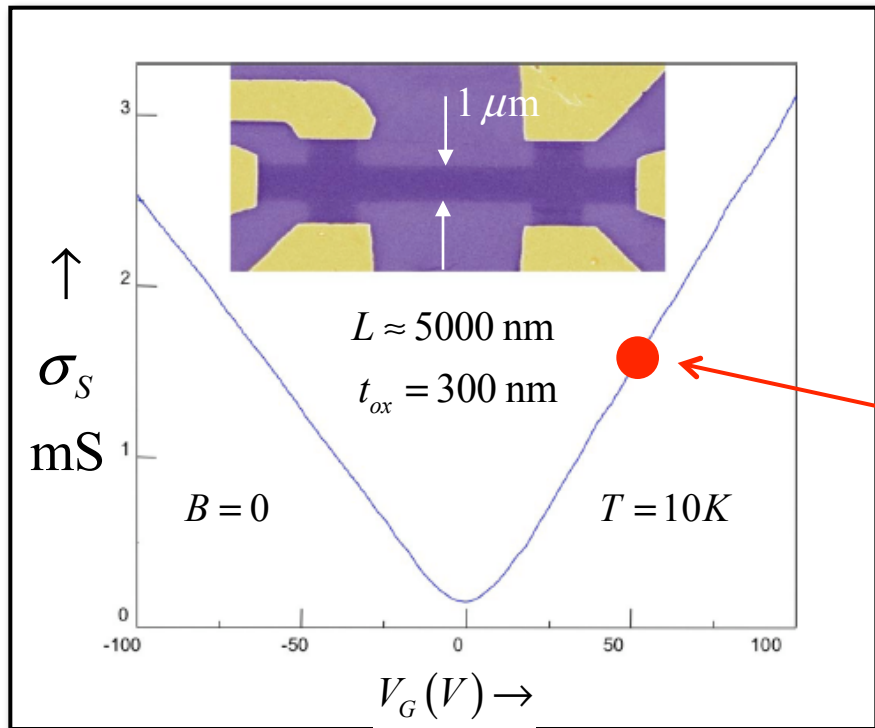


Fig. 30 in A. H. Castro, et al., "The electronic properties of graphene," Rev. of Mod. Phys., **81**, 109, 2009.

$$\sigma_S \approx 1.5 \text{ mS}$$

$$n_S \approx 3.6 \times 10^{12} \text{ cm}^{-2}$$

$$E_F \approx 0.2 \text{ eV}$$

$$\lambda_{app} (0.2 \text{ eV}) \approx 90 \text{ nm}$$

$$\frac{\lambda(0.2 \text{ eV})}{\lambda(0.3 \text{ eV})} \approx 0.69$$

$$\frac{0.2 \text{ eV}}{0.3 \text{ eV}} \approx 0.67$$

$$\lambda(E_F) \propto E_F$$

# Mobility

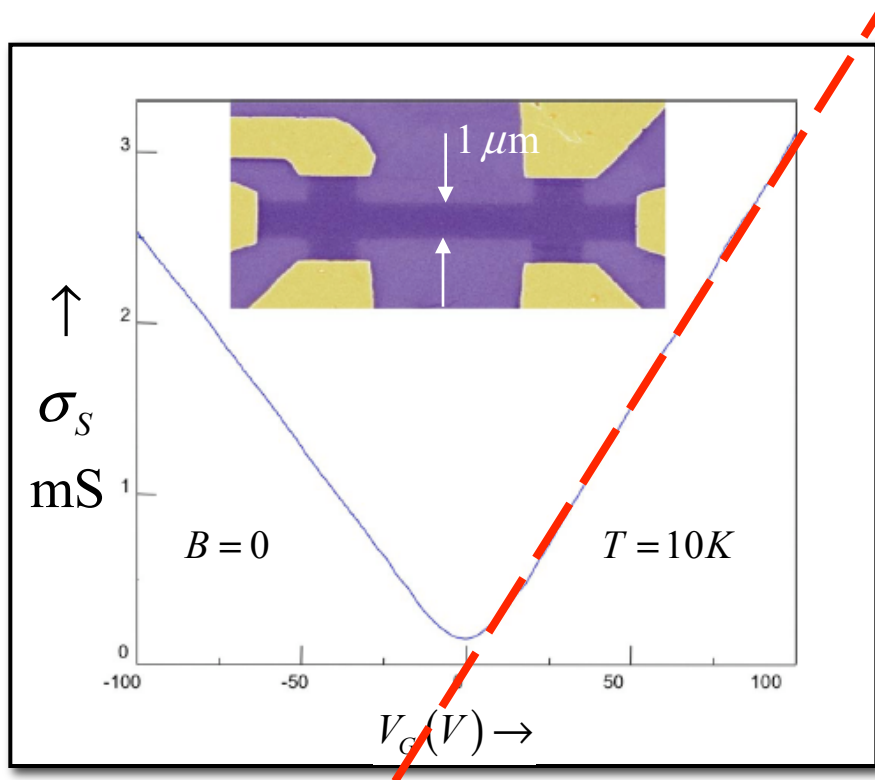


Fig. 30 in A. H. Castro, et al., “The electronic properties of graphene,” Rev. of Mod. Phys., **81**, 109, 2009.

Since,  $\sigma_S \sim n_S$ , we can write:

$$\sigma_S \equiv n_S q \mu_n$$

and deduce a mobility:

$$\mu_n \approx 12,500 \text{ cm}^2/\text{V-sec}$$

Mobility is constant, but mean-free-path depends on the Fermi energy (or  $n_S$ ).



# Electron-hole puddles

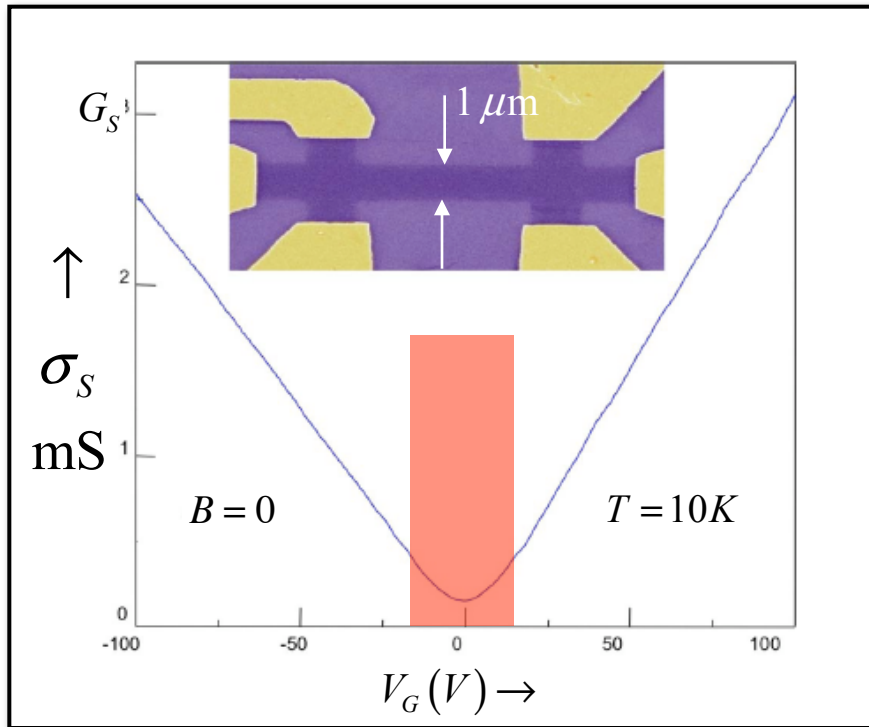
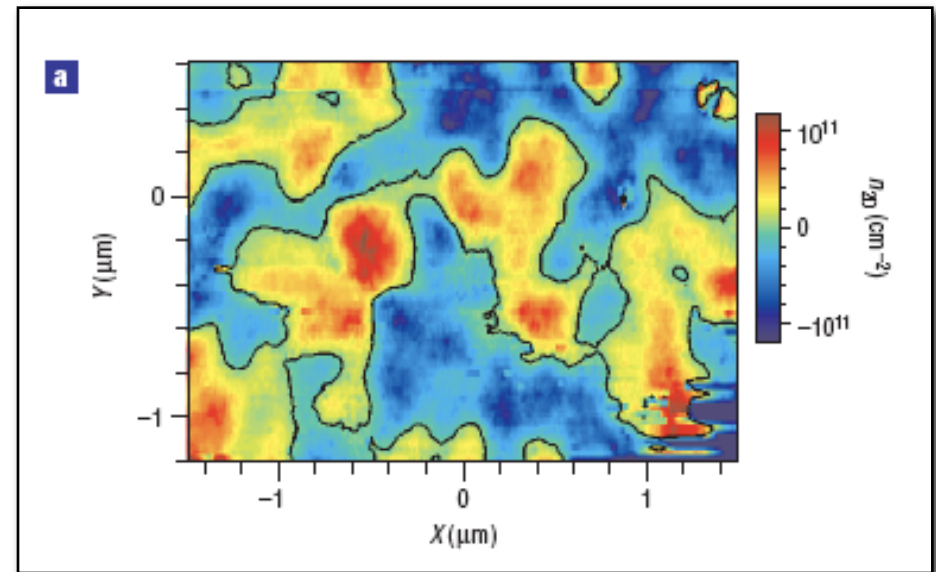
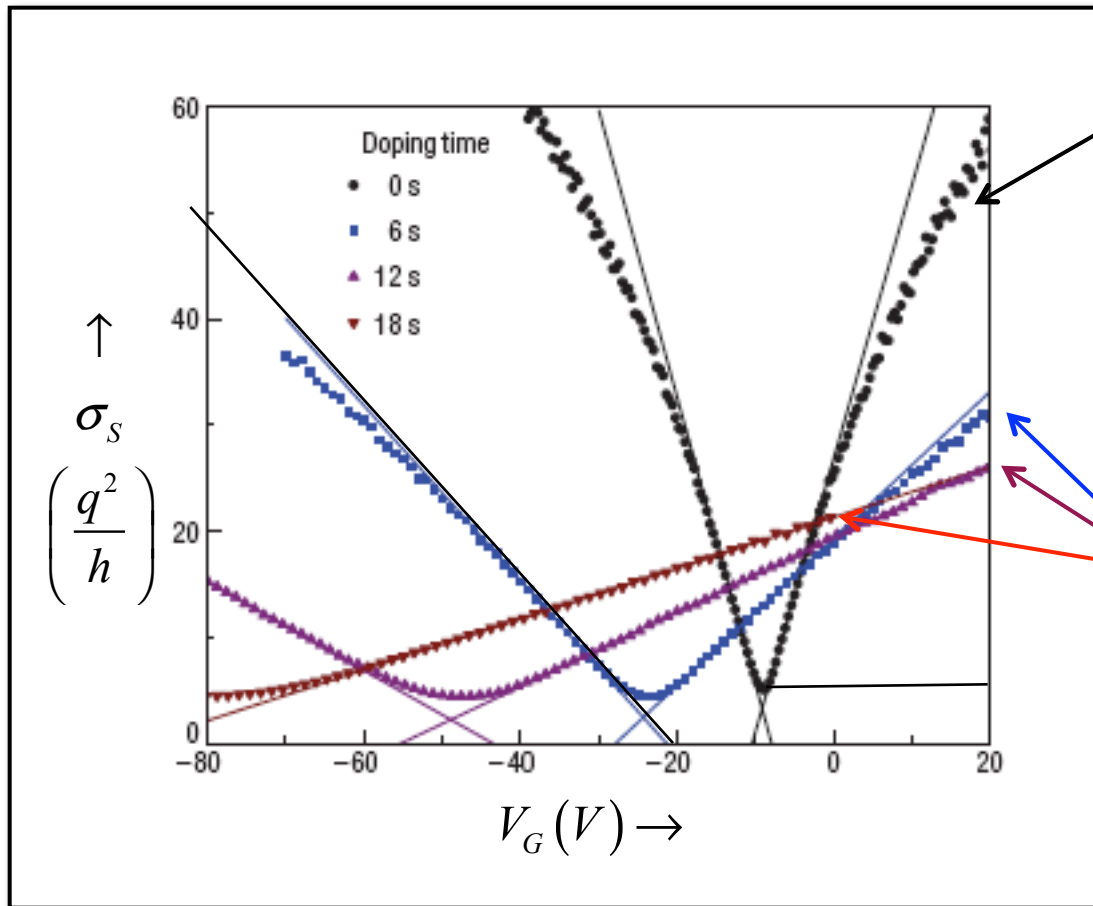


Fig. 30 in A. H. Castro, et al., "The electronic properties of graphene," Rev. of Mod. Phys., **81**, 109, 2009.



J. Martin, et al, "Observation of electron-hole puddles in graphene using a scanning single-electron transistor," Nature Phys., **4**, 144, 2008

# Effect of potassium doping

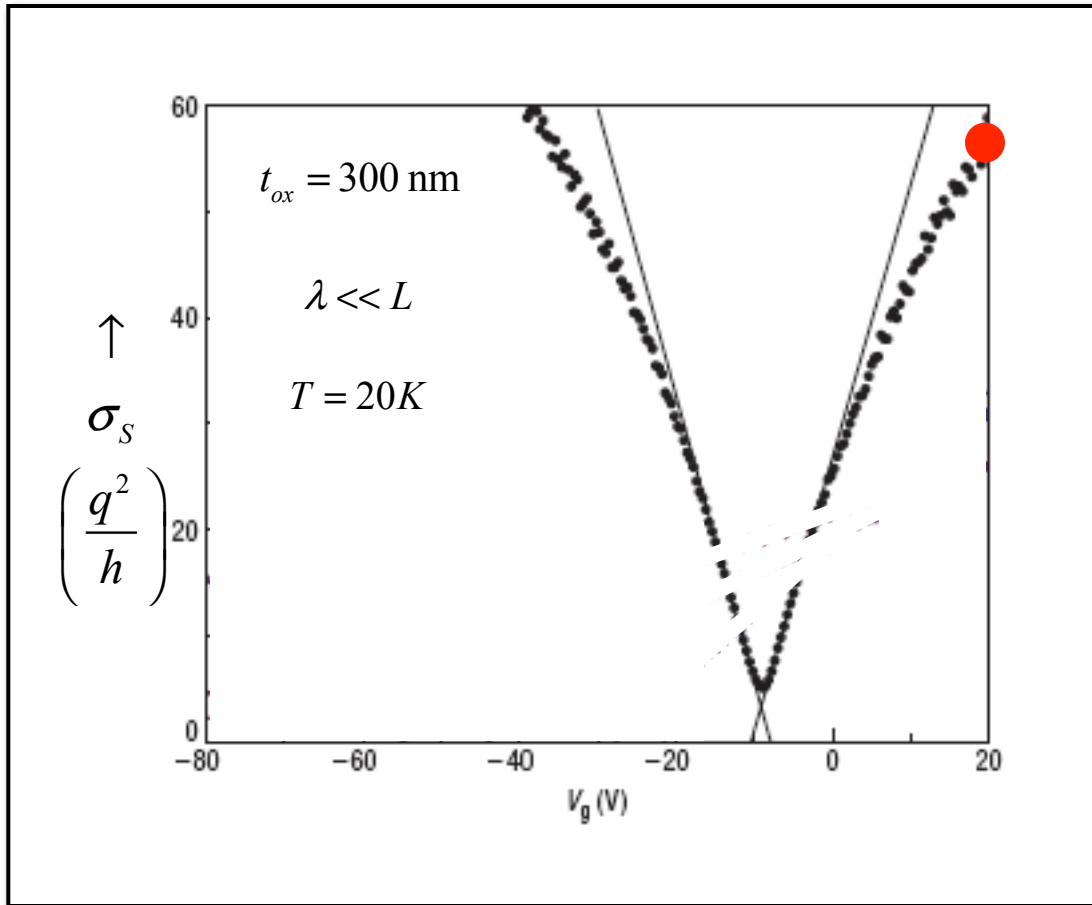


For nominally undoped samples,  $\sigma_S$  vs.  $n_S$  is non-linear.

As doping increases,  $\sigma_S$  vs.  $n_S$  becomes more linear, mobility decreases, and the NP shifts to the left.

J.-H. Chen, C. Jang, S. Adam, M. S. Fuhrer, E. D. Williams, and M. Ishigami, "Charged-impurity scattering in graphene," *Nature Phys.*, **4**, 377-381, 2008.

# Nominally undoped sample: is it ballistic?



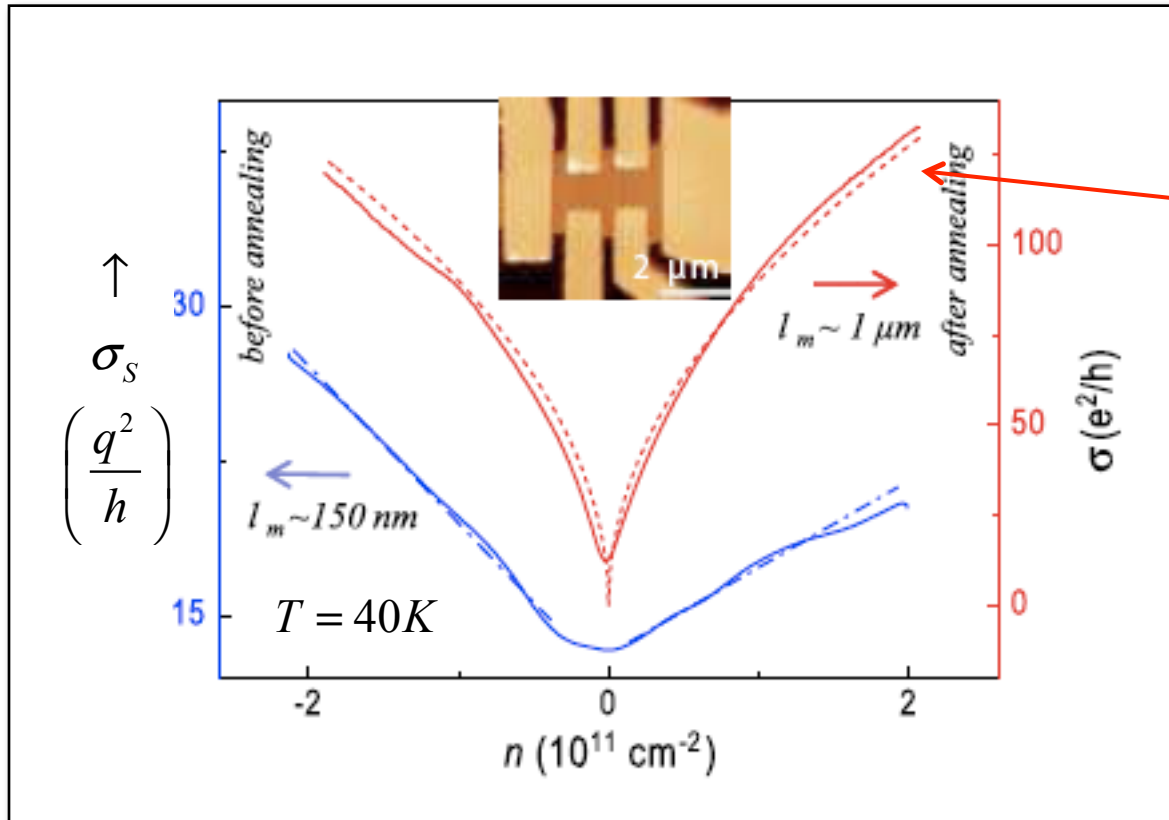
$$\frac{1}{\lambda_{app}} = \frac{1}{\lambda} + \frac{1}{L}$$

$$\lambda_{app} = \frac{\sigma_s / (2q^2/h)}{2\sqrt{n_s/\pi}} \approx 164 \text{ nm}$$

$$\lambda \ll L$$

J.-H. Chen, C. Jang, S. Adam, M. S. Fuhrer, E. D. Williams, and M. Ishigami, "Charged-impurity scattering in graphene," *Nature Phys.*, **4**, 377-381, 2008.

# Unannealed vs. annealed suspended graphene



$$\sigma_s \propto \sqrt{n_s}$$

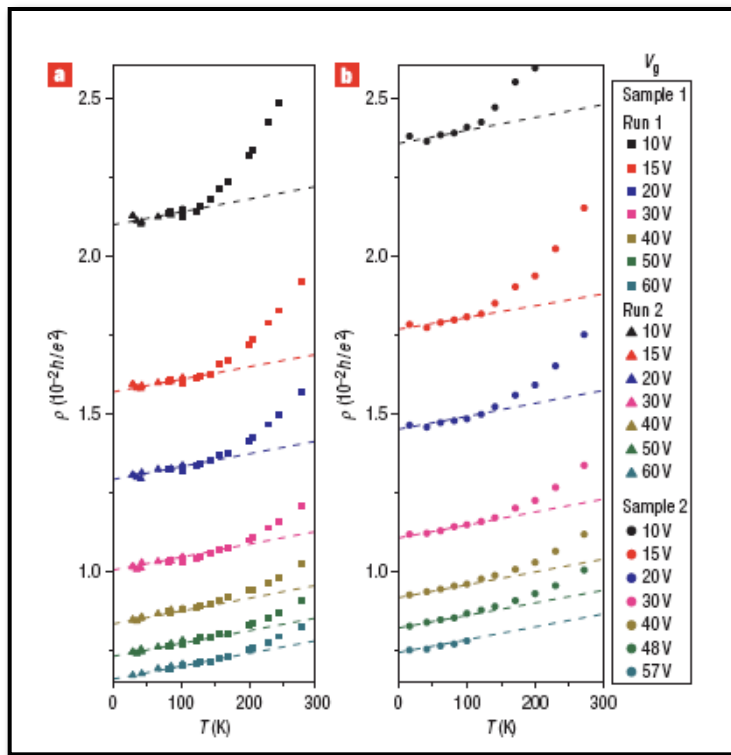
$$\lambda_{app} \approx 1300 \text{ nm}$$

expected from  
ballistic theory

K. I. Bolotin, K. J. Sikes, J. Hone, H. L. Stormer, and P. Kim, "Temperature dependent transport in suspended graphene," 2008

# Temperature dependence

Away from the conductance minimum, the conductance decreases as  $T_L$  increases (or resistivity increases as temperature increases).



$T_L < 100K$ :  $R_S \propto T_L$   
(acoustic phonon scattering - intrinsic)

$T_L > 100K$ :  $R_S \propto e^{h\omega_0/k_B T_L}$   
(optical phonons in graphene or surface phonons at  $\text{SiO}_2$  substrate)

J.-H. Chen, J. Chuan, X. Shudong, M. Ishigami, and M.S. Fuhrer, "Intrinsic and extrinsic performance limits of graphene devices on  $\text{SiO}_2$ ," Nature Nanotechnology, **3**, pp. 206-209, 2008.

# Phonons and temperature dependence

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$$R_S = \frac{1}{G_S} \propto \frac{1}{\lambda} \propto \frac{1}{\tau} \propto n_0$$

$$n_0 = \frac{1}{e^{\hbar\omega(\beta)/k_B T_L} - 1}$$

**acoustic phonons:**

$$\hbar\omega < k_B T_L$$

$$N_\beta \approx \frac{k_B T_L}{\hbar\omega}$$

$$R_S \propto T_L$$

**optical phonons:**

$$\hbar\omega_0 \approx k_B T_L$$

$$n_0 = \frac{1}{e^{\hbar\omega_0/k_B T_L} - 1}$$

$$R_S \propto \frac{1}{e^{\hbar\omega_0/k_B T_L} - 1}$$

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- 1) Hall bar or van der Pauw geometries allow measurement of both resistivity and Hall concentration from which the Hall mobility can be deduced.
- 2) Temperature-dependent measurements provide information about the dominant scattering mechanisms.
- 3) Care must be taken to exclude thermoelectric effects.



# References

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Kasper A. Borup, Johannes de Boor, Heng Wang, cFivos Drymiotis, Franck Gascoin, Xun Shi, Lidong Chen, Mikhail I. Fedorov, Eckhard Muller, Bo B. Iversen, and G. Jeffrey Snyder, “Measuring thermoelectric transport properties of Materials,” *Energy & Environmental Science*, **8**, 423–435, 2015.

# Questions

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