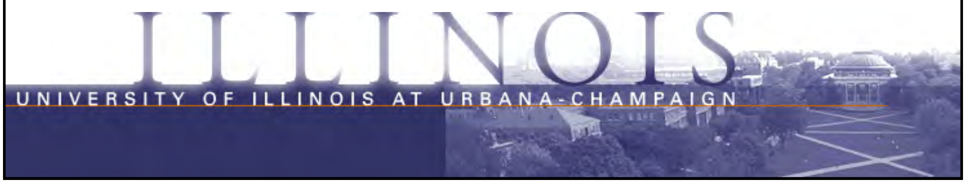
# Graphene Thermal Physics

**Eric Pop**

**Dept. of Electrical & Computer Engineering**
**Micro & Nanotechnology Lab, Beckman Institute**

***http://poplab.ece.illinois.edu***

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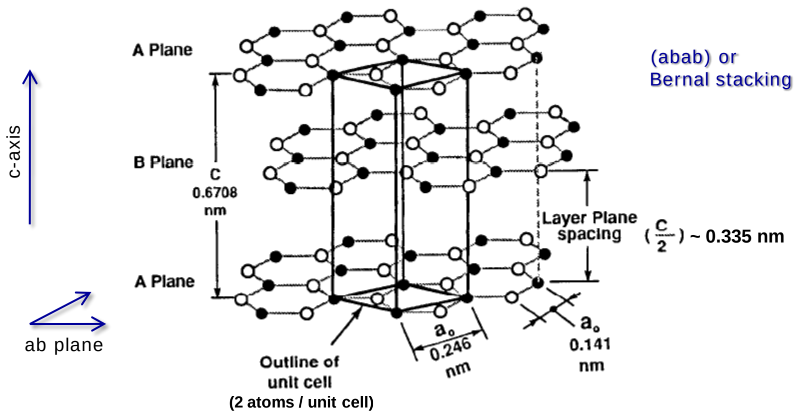
- Heat capacity of graphene
- Thermal conductivity of graphene

} + Background

- Graphene devices
- "Other" physics
  (thermoelectrics, asymmetry, quantum thermal conductance…)

# Structure of Graphite




- Graphite is highly asymmetric
- Strong in-plane covalent $sp^2$ sigma bonds (524 kJ/mol)
- Weak out-of-plane van der Waals pi bonds (7 kJ/mol)



# Heat Capacity: Energy Stored ΔU=CΔT

- Heat capacity of a solid: $C = C_{electrons} + C_{lattice}$

$$C_L = \frac{du}{dT} = \int \hbar\omega \frac{df(\omega)}{dT} g(\omega) d\omega$$

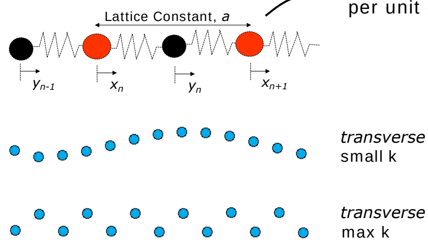
ω(k)
phonon dispersion

density of states

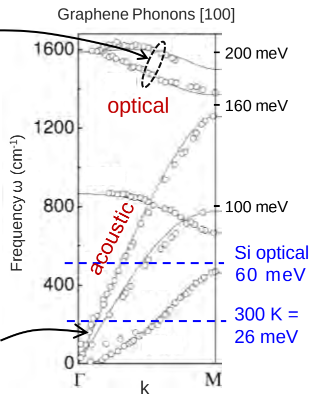
phonon distribution
(e.g. Bose-Einstein)

- High temperature: classically recall $C = 3N_A k_B$
  Dulong-Petit Law (1819) → most solids → 25 J/mol/K at high T
- Low temperature: experimentally C → 0

# Phonons: Atomic Lattice Vibrations




2 atoms per unit cell



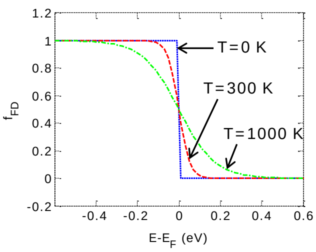

$$\mathbf{u}(\mathbf{r},t) = \mathbf{A}\exp[i(\mathbf{k}\cdot\mathbf{r} - i\omega t)]$$

$$v_S = \frac{d\omega}{dk}$$

- Transverse ($u \perp k$) vs. longitudinal modes ($u \parallel k$)
- Acoustic (transport sound & heat) vs. optical (scatter light & electrons)
- "Hot phonons" = highly occupied modes above room temperature

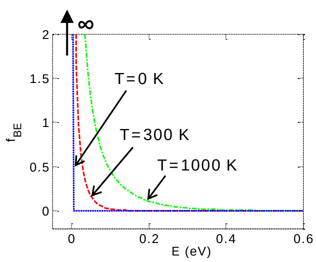


# Fermi-Dirac vs. Bose-Einstein Statistics




$$f_{FD}(E) = \frac{1}{\exp\left(\frac{E-E_F}{k_B T}\right)+1}$$

*Fermions = half-integer spin*
*(e.g. electrons, protons)*



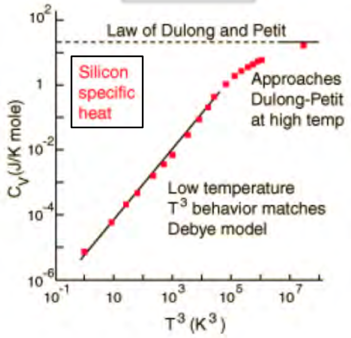

$$f_{BE}(E) = \frac{1}{\exp\left(\frac{E}{k_B T}\right)-1}$$

*Bosons = integer spin*
*(e.g. phonons, photons)*

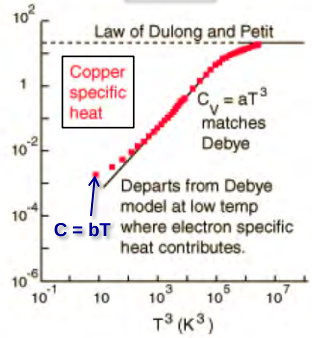
- In the limit of high energy, both reduce to the classical Maxwell-Boltzmann statistics: $f_{MB}(E) \approx \exp(-E/k_B T)$

## Heat Capacity: Dielectrics vs. Metals

Dielectrics




Metals



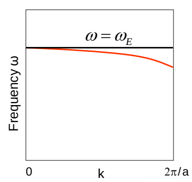

- Very high T: C = $3nk_B$ (constant) both dielectrics & metals
- Intermediate T: C ~ $aT^D$ both dielectrics & metals in D dimensions*
- Very low T: C ~ bT metals (electron contribution only)

* assuming linear ω=vk phonon dispersion



## Heat Capacity: Einstein Model (1907)

- Key assumption: all oscillators have same frequency
- High-T (correct, recover Dulong-Petit Law):




$$C_E(T) \approx 3Nk_B$$

- Low-T (incorrect, drops too fast)

$$C_E(T) \approx 3Nk_B \left(\frac{\hbar\omega_E}{k_B T}\right)^2 e^{-\hbar\omega_E/k_B T}$$

But... Einstein model OK for optical phonon heat capacity

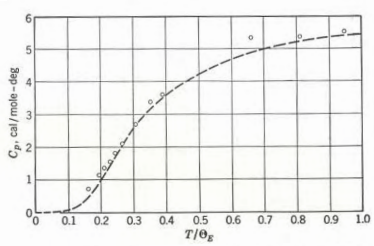
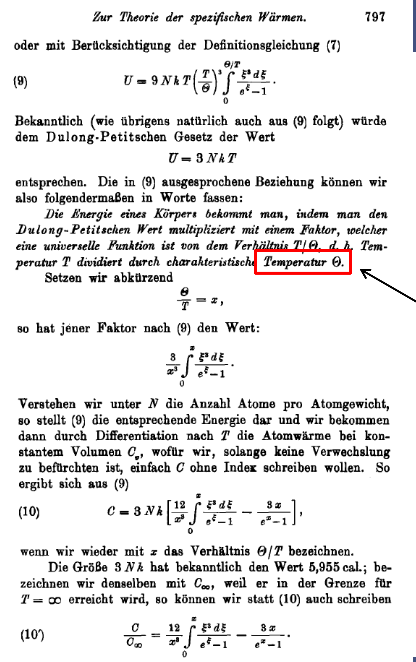



Fig. 6.2. Comparison of experimental values of the heat capacity of diamond and values calculated on the Einstein model, using $\Theta_E$ = 1320°K. [After A. Einstein, Ann. Physik **22**, 180 (1907).]



*Zur Theorie der spezifischen Wärmen.* 797

oder mit Berücksichtigung der Definitionsgleichung (7)

$$U = 9NkT\left(\frac{T}{\Theta}\right)^3 \int_0^{\Theta/T} \frac{\xi^3 d\xi}{e^\xi - 1}. \quad (9)$$

Bekanntlich (wie übrigens natürlich auch aus (9) folgt) würde dem Dulong-Petitschen Gesetz der Wert

$$U = 3NkT$$

entsprechen. Die in (9) ausgesprochene Beziehung können wir also folgendermaßen in Worte fassen:

*Die Energie eines Körpers bekommt man, indem man den Dulong-Petitschen Wert multipliziert mit einem Faktor, welcher eine universelle Funktion ist von dem Verhältnis $T/\Theta$, d. h. Temperatur $T$ dividiert durch charakteristische Temperatur $\Theta$.*

Setzen wir abkürzend

$$\frac{\Theta}{T} = x,$$

so hat jener Faktor nach (9) den Wert:

$$\frac{3}{x^3}\int_0^x \frac{\xi^3 d\xi}{e^\xi - 1}.$$

Verstehen wir unter $N$ die Anzahl Atome pro Atomgewicht, so stellt (9) die entsprechende Energie dar und wir bekommen dann durch Differentiation nach $T$ die Atomwärme bei konstantem Volumen $C_v$, wofür wir, solange keine Verwechslung zu befürchten ist, einfach $C$ ohne Index schreiben wollen. So ergibt sich aus (9)

$$C = 3Nk\left[\frac{12}{x^3}\int_0^x \frac{\xi^3 d\xi}{e^\xi - 1} - \frac{3x}{e^x - 1}\right], \quad (10)$$

wenn wir wieder mit $x$ das Verhältnis $\Theta/T$ bezeichnen.

Die Größe $3Nk$ hat bekanntlich den Wert 5,955 cal.; bezeichnen wir denselben mit $C_\infty$, weil er in der Grenze für $T = \infty$ erreicht wird, so können wir statt (10) auch schreiben

$$\frac{C}{C_\infty} = \frac{12}{x^3}\int_0^x \frac{\xi^3 d\xi}{e^\xi - 1} - \frac{3x}{e^x - 1}. \quad (10')$$

*Annalen der Physik* 39(4) p. 789 (1912)

*Peter Debye (1884-1966)*



# Heat Capacity: Debye Model (1912)

- Key assumption: oscillators have linear ω-k with cutoff $\omega_D$
- Often in terms of Debye temperature

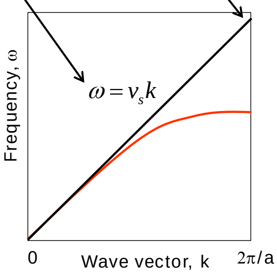
$$\theta_D = \frac{\hbar v_s}{k_B}\left(6\pi^2 N\right)^{1/3}$$

- … roughly corresponds to max acoustic phonon frequency cutoff $k_B\theta_D = \hbar\omega_D$
- Low-T (< $\theta_D$/10):

$$C_D(T) \approx \frac{12\pi^4}{5} Nk_B\left(\frac{T}{\theta_D}\right)^3$$

- High-T: (> 0.8 $\theta_D$)

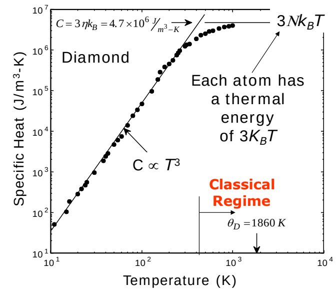
$$C_D(T) \approx 3Nk_B$$




In D-dimensions when T << $\theta_D$: $C_D \propto T^D$

# Debye Model at Low- and High-T

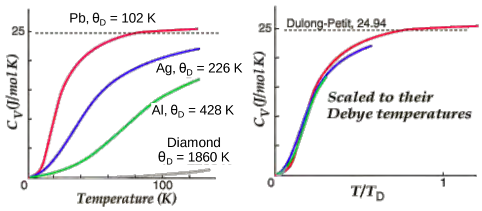
- In practice: $\theta_D$ ~ fitting parameter to heat capacity data
- $\theta_D$ is related to "stiffness" of solid and melting temperature




Debye Temperatures

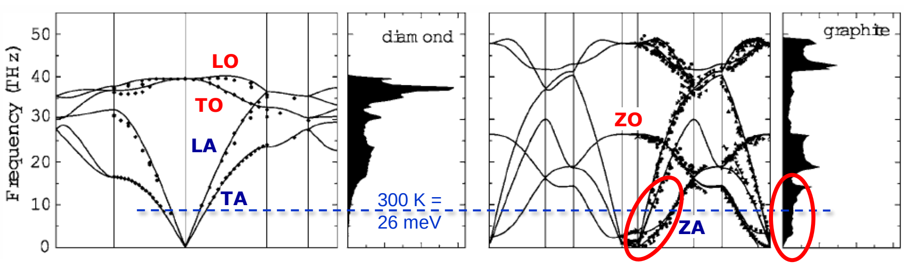
| Element | $\theta_D$, °K | Compound | $\theta_D$, °K |
|---|---|---|---|
| Li | 335 | NaCl | 280 |
| Na | 156 | KCl | 230 |
| K | 91.1 | $CaF_2$ | 470 |
| Cu | 343 | LiF | 680 |
| Ag | 226 | $SiO_2$ (quartz) | 255 |
| Au | 162 | | |
| Al | 428 | | |
| Ga | 325 | | |
| Pb | 102 | | |
| Ge | 378 | | |
| Si | 647 | | |
| C | 1860 | | |
| Graphite | 2480 → in-plane (sp2); 180 → out-of-plane (vdW) | | |

in-plane (sp2) and out-of-plane (vdW) values: to resolve low-temperature heat capacity "quandary" since graphene data was neither 2-D ($T^2$) nor 3-D ($T^3$)





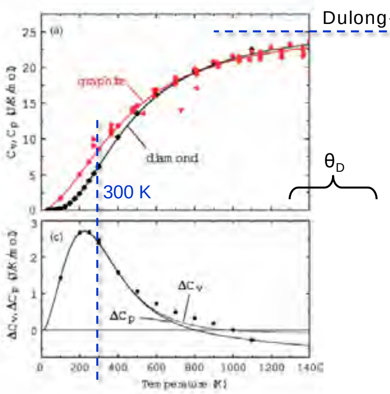

# Phonon Dispersion of Diamond & Graphite




- Diamond "like" silicon:
  - Longitudinal & transverse (x2) acoustic (LA, TA)
  - Longitudinal & transverse (x2) optical (LO, TO)
- Graphite is unusual:
  - Layer-shearing, -breathing, and -bending modes (ZA, ZO)
  - Higher optical freq. than diamond, strong $sp^2$ bond stretching modes
  - Graphite has more low-frequency modes

Tohei, Phys. Rev. B (2006)

# Heat Capacity of Diamond & Graphite



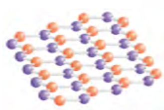

| | $C_p$ at 25˚C and 1 atm. (kJ/kg·K) |
|---|---|
| Graphite | 0.690 - 0.719 |
| Diamond | 0.502 - 0.519 |
| Boron | 1.025 |
| Aluminum | 0.900 |
| Titanium | 0.523 |
| Copper | 0.385 |
| Tungsten | 0.130 |
| Water | 4.186 |
| **Silicon** | **0.80** |
| **$SiO_2$** | **0.71** |

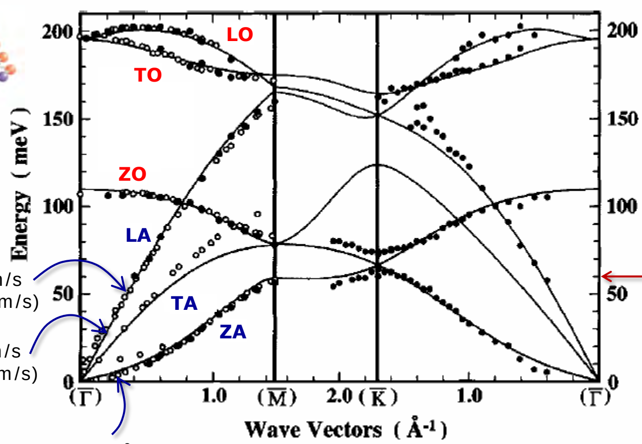
- Graphite has higher phonon DOS at low frequency → higher heat capacity than diamond at room T
- Both increase up to Debye temperature range, then reach "classical" $3N_Ak_B$ limit

Pierson (1993)
Tohei, Phys. Rev. B (2006)



# Phonon Dispersion in Graphene






$v_{LA}$ ~ 23 km/s (silicon ~ 9 km/s)

$v_{TA}$ ~ 16 km/s (silicon ~ 5 km/s)

Si optical phonons

$\omega \sim k^2$
$v_{ZA} \sim 0$ ← out-of-plane bending or flexing mode important for heat capacity at very low T may be affected (suppressed) by substrate

Yanagisawa *et al.*, *Surf. Interf. Analysis* 37, 133 (2005)

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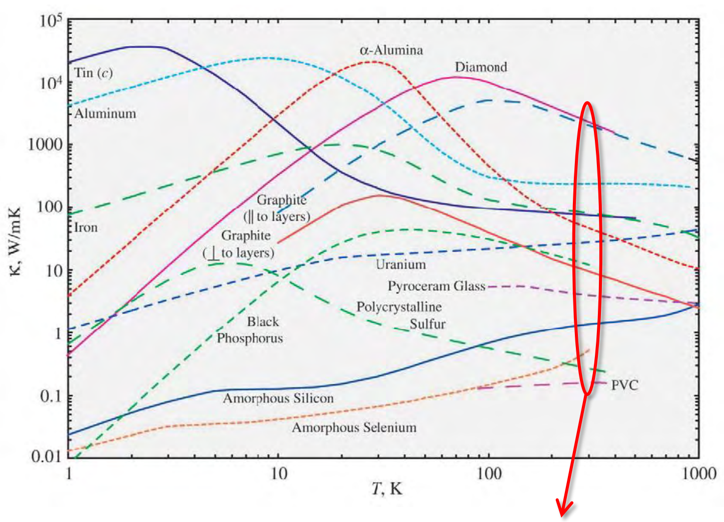
+ Background

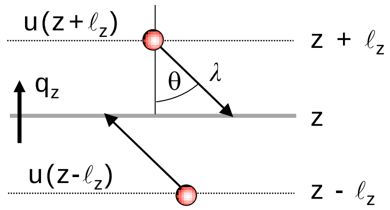


# Thermal Conductivity of Solids




Unlike electrical conductivity, thermal spans "only" 4 orders of magnitude

# Thermal Conductivity: Kinetic Theory




*compare to…* $J_{diff} = -qD_n \frac{dn}{dz}$

Integrate energy flux over all angles:

$$J_q = -\frac{1}{3} v\lambda \frac{du}{dT}\frac{dT}{dz} \equiv -k\frac{dT}{dz}$$

heat capacity

Thermal conductivity: $k = \frac{1}{3} C v \lambda$

mean free path (MFP)

in 3-D

phonon velocity dω/dk

$$v = \left[\frac{1}{3}\left(\frac{2}{v_{TA}} + \frac{1}{v_{LA}}\right)\right]^{-1}$$

heat gen. per unit volume (W/m³)

Heat diffusion eq:

$$\nabla\cdot(k\nabla T) - q''' = 0$$

*Poisson eq:*

*compare to…* $\nabla\cdot(\epsilon\nabla V) - \rho = 0$

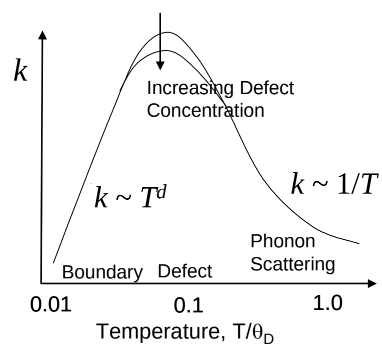


# Thermal Conductivity and Phonon MFP

- Heat capacity depends on → ω(k) and T
- Phonon group velocity depends only → ω(k)
- Mean free path λ → phonon scattering mechanisms:
  - Boundary & edge scattering
  - Defect & impurity scattering
  - Phonon-phonon scattering ~ 1/T

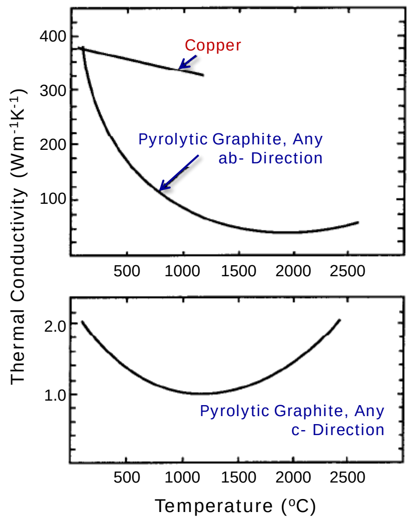
| | C | λ | k |
|---|---|---|---|
| low T | $\propto T^d$ | $\lambda \rightarrow L$ (size) | $\propto T^d$ |
| high T | $3Nk_B$ | $\propto 1/T$ | $\propto 1/T$ |

$$k \approx \frac{1}{3} C v\lambda \approx \frac{1}{3} C v^2 \tau$$



# Thermal Conductivity of Graphite*

* *typical commercial pyrolitic graphite*




| | W/m·K at 25°C |
|---|---|
| Pyrolytic graphite: | |
| *ab* directions | 390 |
| *c* direction | 2 |
| Graphite fiber (pitch-based) | 1180 |
| Diamond (Type II) | 2000 - 2100 |
| Silver | 420 |
| Copper | 385 |
| Beryllium oxide | 260 |
| Aluminum nitride | 200 |
| Alumina | 25 |
| **Silicon** | **140** |
| **$SiO_2$** | **1.4** |

comparable to copper (ab directions)

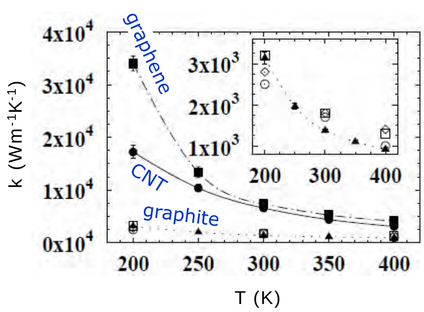
comparable to plastics, $SiO_2$ (c direction)

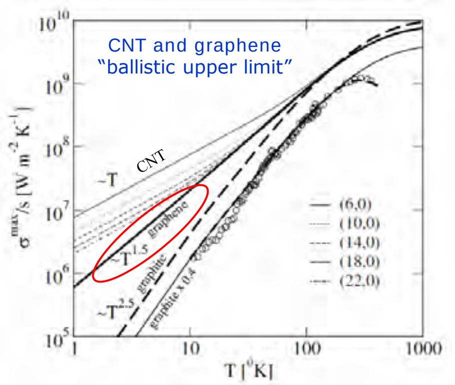
*High (100-1000x) thermal anisotropy in all graphite*

Pierson (1993)



# Graphene Thermal Conductivity (Theoretical)

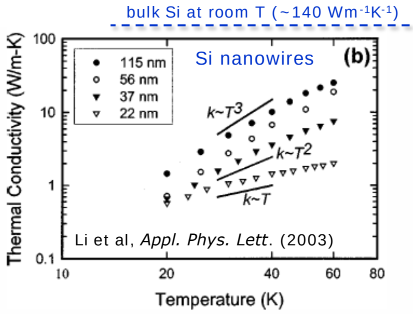


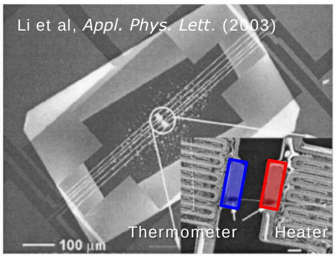



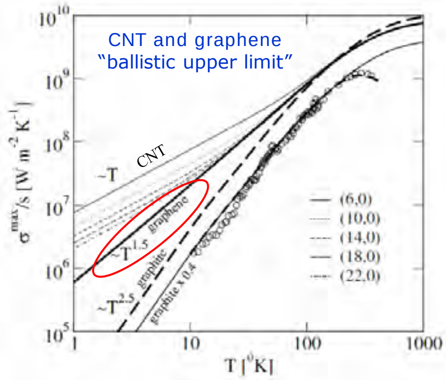

- *Graphite* ab- thermal conductivity generally lower because of interlayer interactions (could be same for graphene on substrate)
- *Graphite* $\sim T^{2.5}$ at very low T due to bending modes ($\omega \sim k^2$)
- Graphene $\sim T^{1.5}$ at very low T due to -------""-------""------- ($\omega \sim k^2$)

Berber et al, *Phys. Rev. Lett.* (2000); Mingo et al, *Phys. Rev. Lett*. (2005)

# The Imprint of Dimensionality



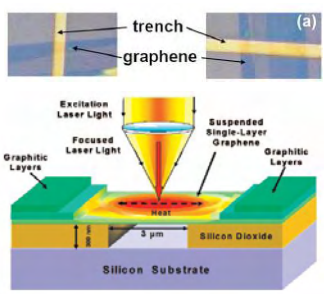




- Phonon dispersion → phonon DOS → heat capacity → k ~ Cvλ
- Information about dimensionality of system can be obtained from low-temperature k-T (or C-T) measurements
- Ex: Si nanowires ~ from 1-D to 3-D features!



# Graphene Thermal Conductivity (Experimental)

- Graphene flakes suspended over $SiO_2$ trenches (~3 um wide)
- Thermometry using Raman G-band (1583 $cm^{-1}$) shift
- *Room temperature* results in-line with existing CNT data



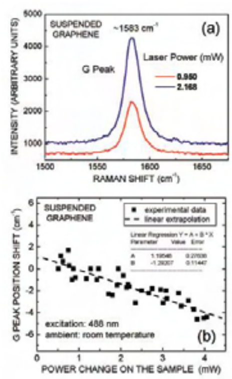




Table 1. Room Temperature Thermal Conductivity in Graphene and CNTs

| sample type | K (W/mK) | method | comments | ref |
|---|---|---|---|---|
| SLG | ~3080-5150 | optical | individual; suspended | Balandin et al. |
| MW-CNT | >3000 | electrical | individual; suspended | Kim et al.[15] |
| SW-CNT | ~3500 | electrical | individual; suspended | Pop et al.[16] |
| SW-CNT | 1750–5800 | thermocouples | bundles | Hone et al.[17] |
| Bulk graphite | 500–2000 max: >2000 | Variety | In-plane (basal); high quality | Ho *et al.*[8] |

Balandin et al, *Nano Lett*. (2008); Ghosh et al, *Appl. Phys. Lett*. (2008)

## Wiedemann-Franz Law

- Q: what is the electronic contribution to thermal transport?
- Wiedemann & Franz (1853) empirically saw $k_e/\sigma$ = const(T)
- Lorenz (1872) noted $k_e/\sigma$ proportional to T

$$\begin{cases} \kappa_e = \frac{1}{3}\left(\frac{\pi^2}{2} k_B^2 n \frac{T}{E_F}\right) v_F^2 \tau \\ \sigma = q\mu n = \frac{q^2 \tau}{m} n \end{cases}$$

taking the ratio: $$L_0 = \frac{\kappa_e}{\sigma T} = \frac{\pi^2 k_B^2}{3q^2}$$

Remarkable: independent of n, m, and even $\tau$!

- Graphene electronic contribution appears <10% throughout T range

Experimentally

| | L = κ/σT 10$^{-8}$ WΩ/K$^2$ | |
|---|---|---|
| Metal | 0 ° C | 100 °C |
| Cu | 2.23 | 2.33 |
| Ag | 2.31 | 2.37 |
| Au | 2.35 | 2.40 |
| Zn | 2.31 | 2.33 |
| Cd | 2.42 | 2.43 |
| Mo | 2.61 | 2.79 |
| Pb | 2.47 | 2.56 |

$$L_0 = 2.45 \times 10^{-8}\ \text{W}\Omega/\text{K}^2$$



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- Heat capacity of graphene
- Thermal conductivity of graphene
  } + Background
- Graphene devices
- "Other" physics
  (thermoelectrics, asymmetry, quantum thermal conductance…)

# Background on Thermal Resistance

$$\mathcal{P} = \mathcal{I}^2 \times \mathcal{R}$$

$$\Delta T = \mathcal{P} \times \mathcal{R}_{th} \qquad \Delta V = \mathcal{I} \times \mathcal{R}_{el}$$

$$\mathcal{R} = f(\Delta T)$$

$$R_{th} = \frac{L}{kA} \qquad R_{el} = \frac{\rho L}{A}$$

**Fourier's Law (1822)**

**Ohm's Law (1827)**



# Thermal-Electrical Cheat Sheet

$$J_q = -k\nabla T \qquad J_{diff} = -qD\nabla n$$

| Thermal | | ⟺ | Electrical | |
|---|---|---|---|---|
| Temperature | T [K] | ⟺ | Voltage V | [V] |
| Heat | Q [J] | ⟺ | Charge Q | [C] |
| Heat transfer rate q [W] | | ⟺ | Current i | [A] |
| Thermal resistance | $R_T$ [K/W] | ⟺ | Electrical resistance | R [V/A] |
| Thermal capacitance $C_T$ [J/K] | | ⟺ | Electrical capacitance C [C/V] | |

**Governing equations**

Steady-State condition

| Temperature Rise | ⟺ | Voltage Difference |
|---|---|---|
| $\Delta T = q R_T$ | ⟺ | $\Delta V = i R$ |

Transient condition

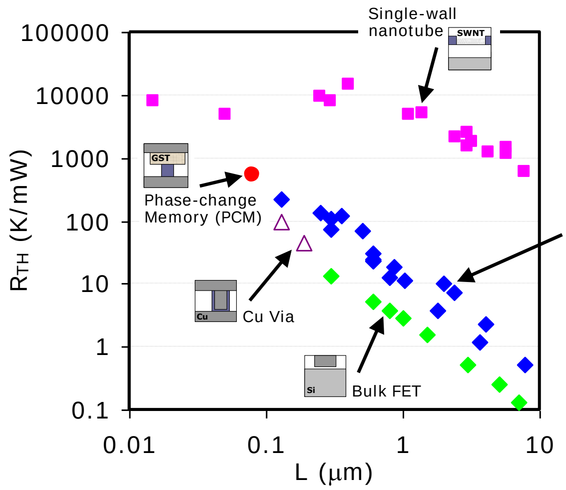
| Heat diffusion | ⟺ | RC transmission line |
|---|---|---|
| $\nabla^2 T = R_T C_T \frac{\partial T}{\partial t}$ | ⟺ | $\nabla^2 V = RC \frac{\partial V}{\partial t}$ |

Figure 1. Thermal-Electrical analogous quantities.

$$\nabla\cdot(k\nabla T) - q''' = 0 \qquad \nabla\cdot(\epsilon\nabla V) - \rho = 0$$

$$R_{th} = \frac{L}{kA} \qquad R_{el} = \frac{\rho L}{A}$$

## Thermal Resistance of Devices




High thermal resistances:

- SWNT due to small thermal conductance (very small d ~ 2 nm)
- Others due to low thermal conductivity, decreasing dimensions, increased role of interfaces

Power input also matters:

- SWNT ~ 0.01-0.1 mW
- Others ~ 0.1-1 mW

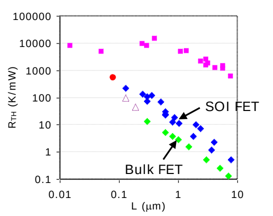
$$\Delta T = P \times R_{th}$$

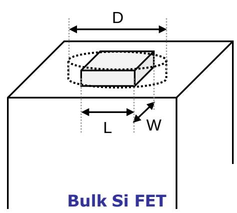
Data: Mautry (1990), Bunyan (1992), Su (1994), Lee (1995), Jenkins (1995), Tenbroek (1996), Jin (2001), Reyboz (2004), Javey (2004), Seidel (2004), Pop (2004-6), Maune (2006).

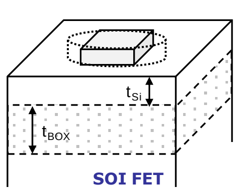


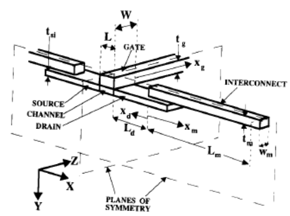
## Modeling Device Thermal Response

- Steady-state models
  - Lumped: Mautry (1990), Goodson-Su (1994-5), Pop (2004), Darwish (2005)
  - Finite-Element models
- Transient models



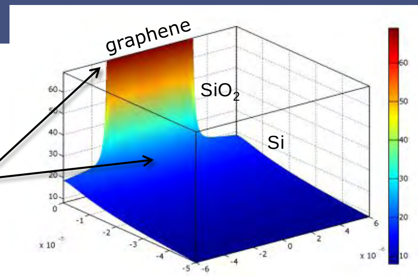




$$R_{TH} = \frac{1}{2k_{Si}D} \approx \frac{1}{2k_{Si}\sqrt{LW}}$$

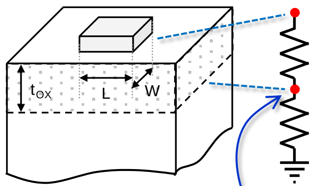
$$R_{TH} \approx \frac{1}{2W}\left(\frac{t_{BOX}}{k_{BOX}k_{Si}t_{Si}}\right)^{1/2}$$

# Graphene FET




$$\frac{T_{Si}}{T_G} \approx 0.29$$

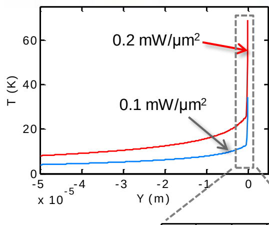
Assumptions:
$t_{ox}$ = 300 nm
L = 25 um
W = 6 um
$k_{ox}$ = 1.3 $Wm^{-1}K^{-1}$
$k_{si}$ = 80 $Wm^{-1}K^{-1}$

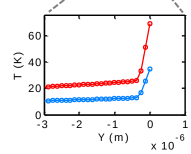



$$R_{ox} \approx \frac{t_{ox}}{k_{ox}LW} \approx 1538\ K/W$$

$$R_{Si} \approx \frac{1}{2k_{Si}\sqrt{LW}} \approx 510\ K/W$$

$$\frac{T_{Si}}{T_G} = \frac{R_{Si}}{R_{ox}+R_{Si}} \approx 0.25$$

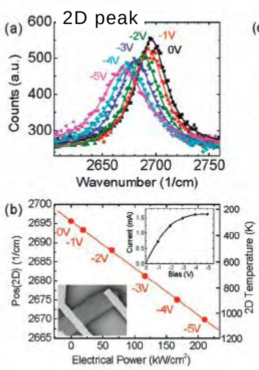


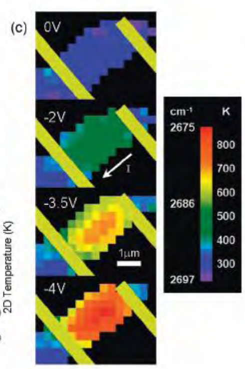



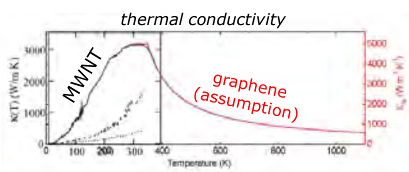

Bae, Ong, Estrada, Pop (2009)

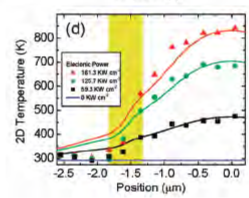


# Raman Thermal Imaging of Graphene FET




- Spatially resolved (~0.5 um) Raman (2D ~ 2700 $cm^{-1}$) thermal imaging
- Suggests power dissipation directly with SO phonons (60 meV) in $SiO_2$
- Suggests thermal boundary $R_B$ (graphene-$SiO_2$) ~ 4x$10^{-8}$ $m^2K/W$

Freitag et al, *Nano Lett*. (2009)

# Table of Contents

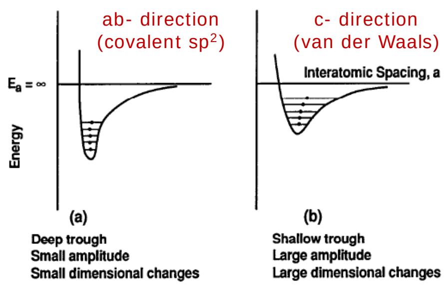
- Heat capacity of graphene
- Thermal conductivity of graphene

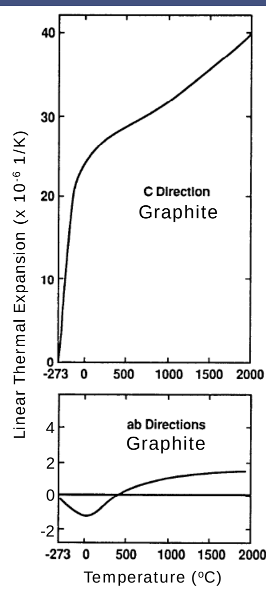
+ Background

- Graphene devices
- "Other" physics
  (thermoelectrics, asymmetry, quantum thermal conductance…)



# Thermal Expansion of Graphite

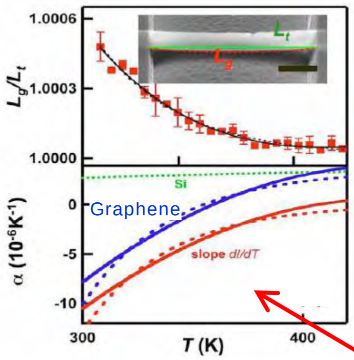


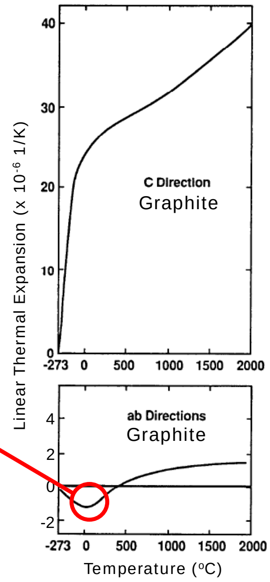




- Thermal expansion coefficient (TEC) is
  - Positive and large in c- direction
  - Negative (at room temperature) in ab- plane
- Graphite thermal expansion anisotropy can result in large internal stresses

Pierson (1993)

# Thermal Expansion of Graphene



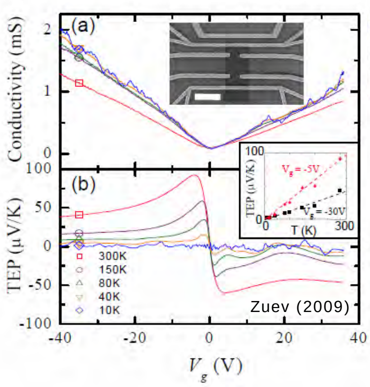



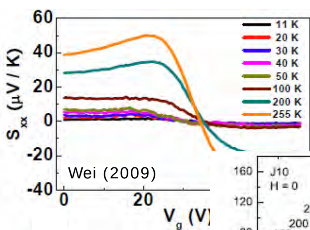

- Recent estimate of thermal contraction in single-layer suspended graphene*
- TEC ~ $-7 \times 10^{-6}$ $K^{-1}$, 5-6x greater than in graphite ab- plane

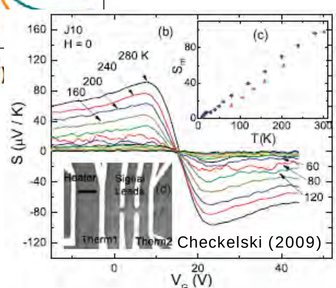
*Bao *et al*, arxiv/cond-mat (2009)



# Thermoelectric Properties of Graphene

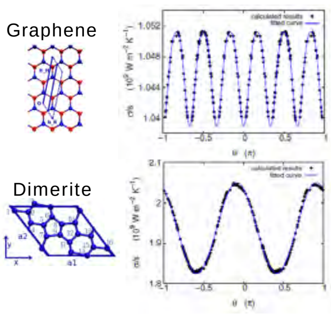


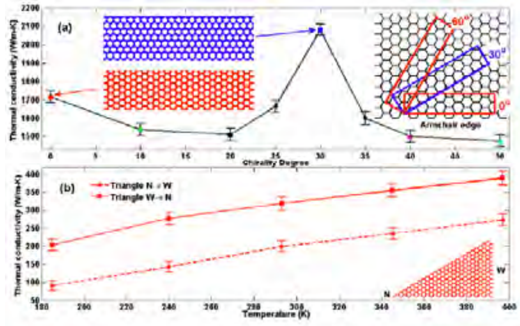




- Measured graphene thermopower $S \leq 100$ μV/K
  - note: $|S_{graphene}| >> |S_{Pd}, S_{Au}|$ at $T > 300$ K
- Peltier coefficient at metal junction: $\Pi = (S_1 - S_2)T$
- Peltier heating/cooling $Q = \Pi I \approx \Pi_{graphene} I$

# Thermal Anisotropy in Graphene




- Thermal conductivity variation with angle:
  - ~1% variation in graphene (π/3 angle period) } ballistic simulation**
  - ~10% variation in dimerite
  - ~25% variation in nanoribbon by Molecular Dynamics*
- Triangular graphene nanoribbon with 30° vertex → up to 2x thermal anisotropy (rectification)*

*Hu, *arxiv/cond-mat* (2009)
**Jiang, *Phys. Rev. B* (2009)



# Wiedemann-Franz Law in Nanoribbons

- Graphene nanoribbons: does the WFL hold in 1D? → YES
- 1D ballistic electrons carry energy too, what is their equivalent thermal conductance?

$$G_{th} = L\sigma_e T = \left(\frac{\pi^2 k_B^2}{3e^2}\right)\left(\frac{e^2}{h}\right)T = \boxed{\frac{\pi^2 k_B^2 T}{3h}}$$

(x2 if electron spin included)

$$G_{th} \approx 0.28 \text{ nW/K at } 300 \text{ K}$$

VOLUME 78, NUMBER 6 PHYSICAL REVIEW LETTERS 10 FEBRUARY 1997

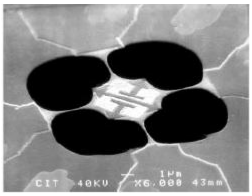
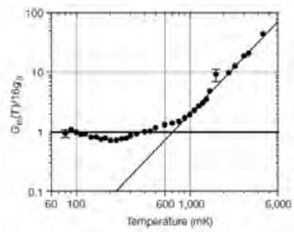
**Thermal Conductivity and Lorenz Number for One-Dimensional Ballistic Transport**

We study the thermal conductivity and Lorenz number of charge carriers for one-dimensional ballistic transport within the correlation function formalism. The carrier transit time between two ideal contacts is found to substitute for the collision time in the definition of a ballistic thermal conductivity. A universal thermal conductance $K = 2\pi^2 k_B^2 T/3h$ is naturally obtained for the degenerate case.

Greiner, Phys. Rev. Lett. (1997)

# Phonon Quantum Thermal Conductance

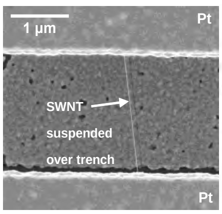
- Same thermal conductance quantum, irrespective of the carrier statistics (Fermi-Dirac vs. Bose-Einstein)

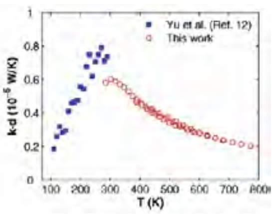




Phonon $G_{th}$ measurement in GaAs bridge at T < 1 K

Schwab, *Nature* (2000)

$$G_{th} = \frac{\pi^2 k_B^2 T}{3h} \approx 0.28 \text{ nW/K at 300 K}$$






Single nanotube $G_{th}$=2.4 nW/K at T=300K

Pop, *Nano Lett.* (2006)



# Graphene Thermal Properties Summary

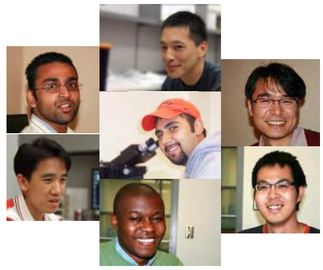
*What we think we know…*

- Thermal conductivity 3000-5000 $Wm^{-1}K^{-1}$ at 300 K (k↓↑T)
  - Phonon-dominated, electron contribution < 10%
  - Phonon mean free path ~0.7 µm
- Heat capacity (graphite) ~0.7 kJ/kg at room temperature (C↑↑T)
- Thermal boundary resistance with $SiO_2$ ~$4x10^{-8}$ $m^2K/W$ (SO phonon)
- Role of dimensionality:
  - Neither "2-D" nor "3-D"
  - Flexing mode ($\omega \sim k^2$) dominates heat capacity at T<50 K

*Much we don't know…*

- Temperature dependence of k, C, TBR
- Role of substrate interaction on all of the above
- Role of edges, defects, doping, isotopes…

# Acknowledgements



- Post-doc:
  - Dr. Myung-Ho Bae
- Grads:
  - V. Dorgan, D. Estrada, A. Liao, Z.-Y. Ong
  - B. Ramasubramanian, T. Tsafack, F. Xiong
- Undergrads:
  - Aidee, Gautam, Ryan, Shreya, Sumit, Tim, Yang
- Collaborators:
  - D. Jena (Notre Dame), M. Hersam (NWU), W. King (MechSE), J.-P. Leburton (ECE), J. Lyding (ECE), N. Mason (Physics), U. Ravaioli (ECE), M. Shim (MatSE)
- Funding:
  - NASA, NRI, UIUC (Beckman Award)
  - ONR, NSF, DARPA Young Faculty Award