

Thermal Conductivity of Graphene

Twists and Turns in Understanding Phonon Transport in
Quasi-Two-Dimensional Materials

Alexander A. Balandin

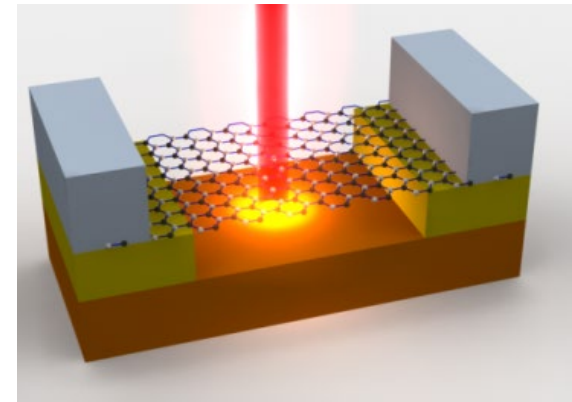
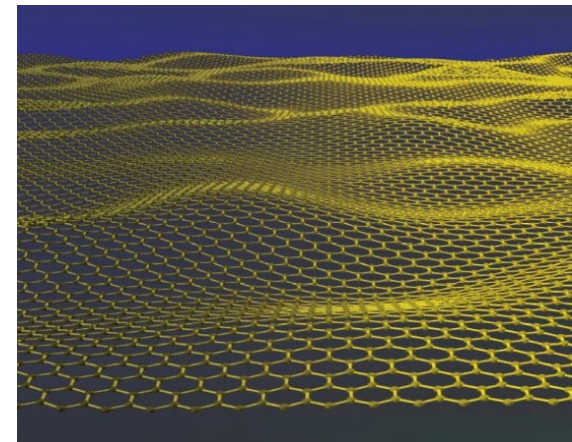
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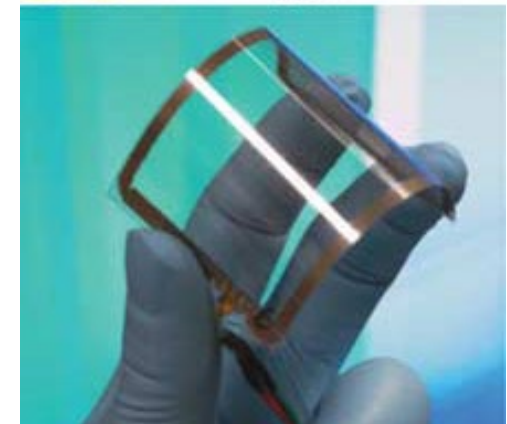
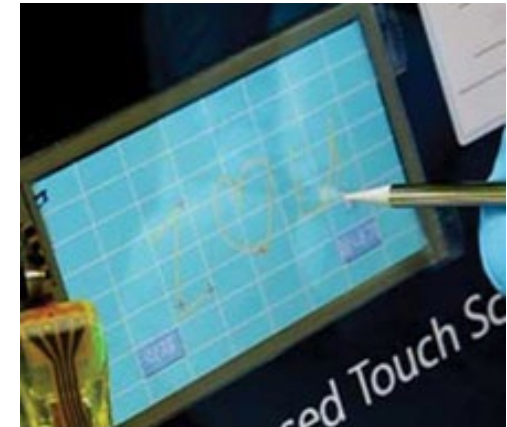
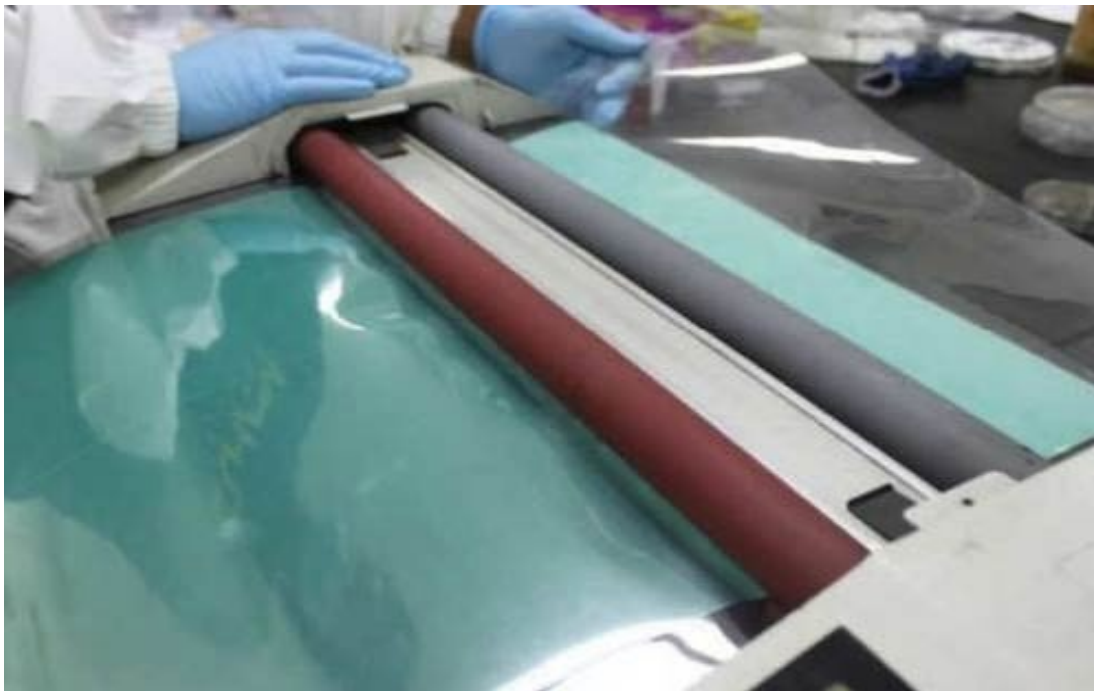


Outline

- ◆ **Introduction and Motivations**
 - *Thermal conductivity in 2D*
- ◆ **Thermal Conductivity of Graphene**
 - *Optothermal Raman technique*
- ◆ **Theory**
 - *Graphene vs. graphite*
 - *ZA or not ZA*
 - *Other twists and turns*
- ◆ **Phononics**
 - *Engineering phonon spectrum in 2D*
- ◆ **Graphene Composites**
 - *TIMs and related*
- ◆ **Outlook**



Commercial Graphene's Applications We Expect – Based on Electronic Properties



- Wish list: electronic applications that use high electron mobility, Fermi level gating, mechanical flexibility, etc.
- **What is needed: CVD growth on dielectric substrates**

Commercial Graphene's Applications We Have – Based on Thermal Properties

Large-scale commercial graphene applications we got so far - graphene thermal

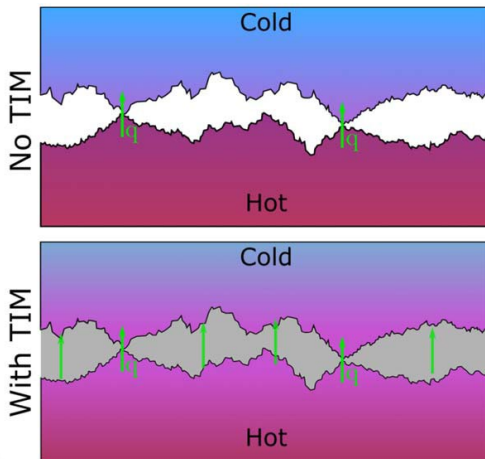


GRAPHENE-X Alpha 3-Layer Shell Jacket: 70% Graphene-integrated nylon and 30% polyester



“Graphene’s thermal properties have revolutionized the sport and athletic textile industry. The material acts as a filter between your skin and the environment, expelling heat in warmer weather and preserving and evenly distributing body heat in colder climates. Graphene enhanced fabrics have the potential to reinforce the natural way in which we adjust our body temperature, all while remaining breathable and comfortable.” ACS Material

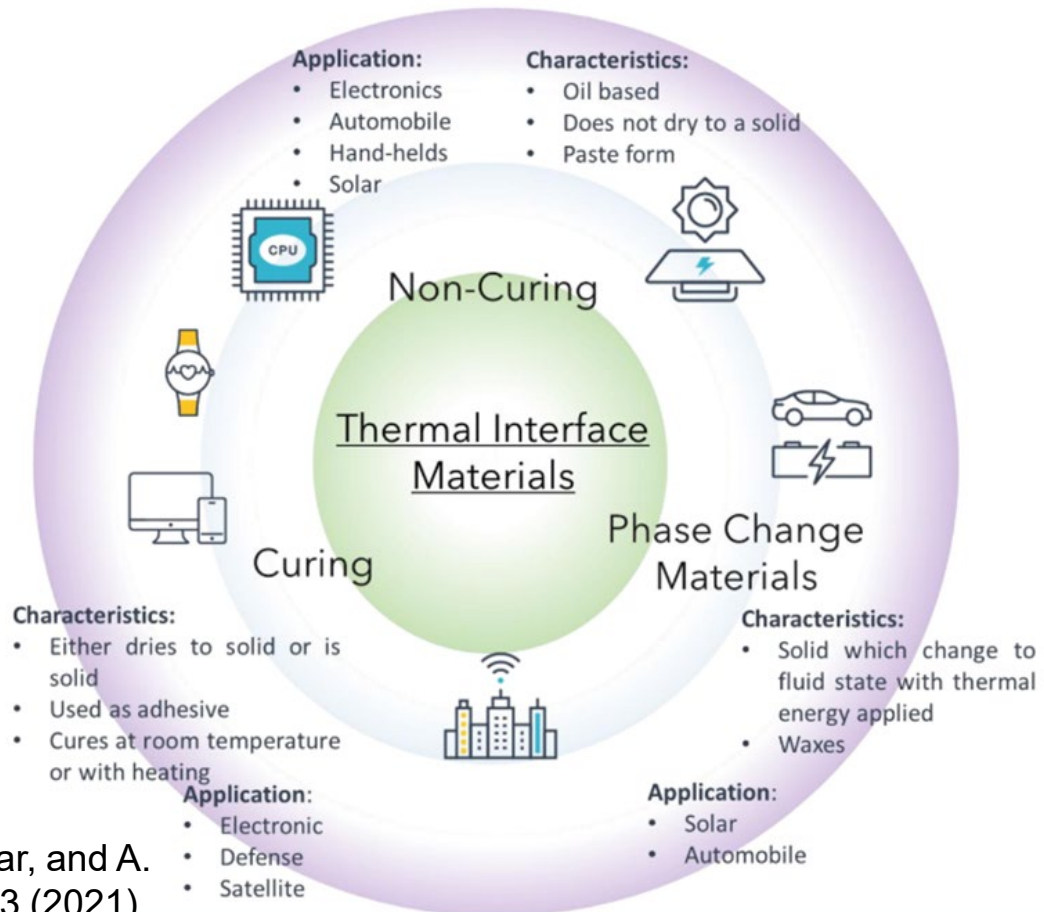
Thermal Management for Electronics – Thermal Interface Materials



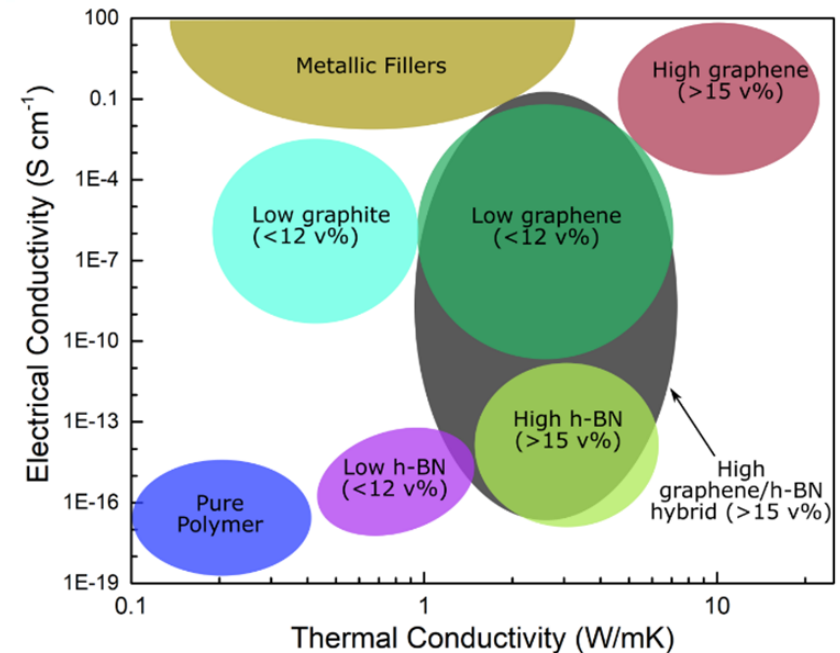
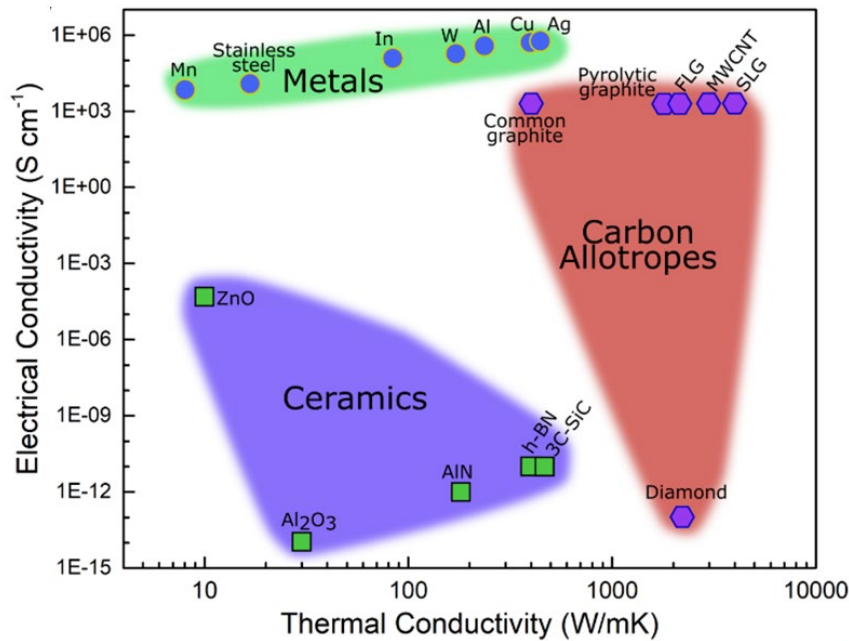
- Terminology: “Graphene” in thermal context is usually a mixture of SLG and FLG
- FLG is flexible and retain high thermal conductivity
- In electronics, “graphene” can be FLG which you can gate

J. S. Lewis, T. Perrier, Z. Barani, F. Kargar, and A. A. Balandin, *Nanotechnology*, 31, 142003 (2021).

Alexander A. Balandin -- University of California, Los Angeles



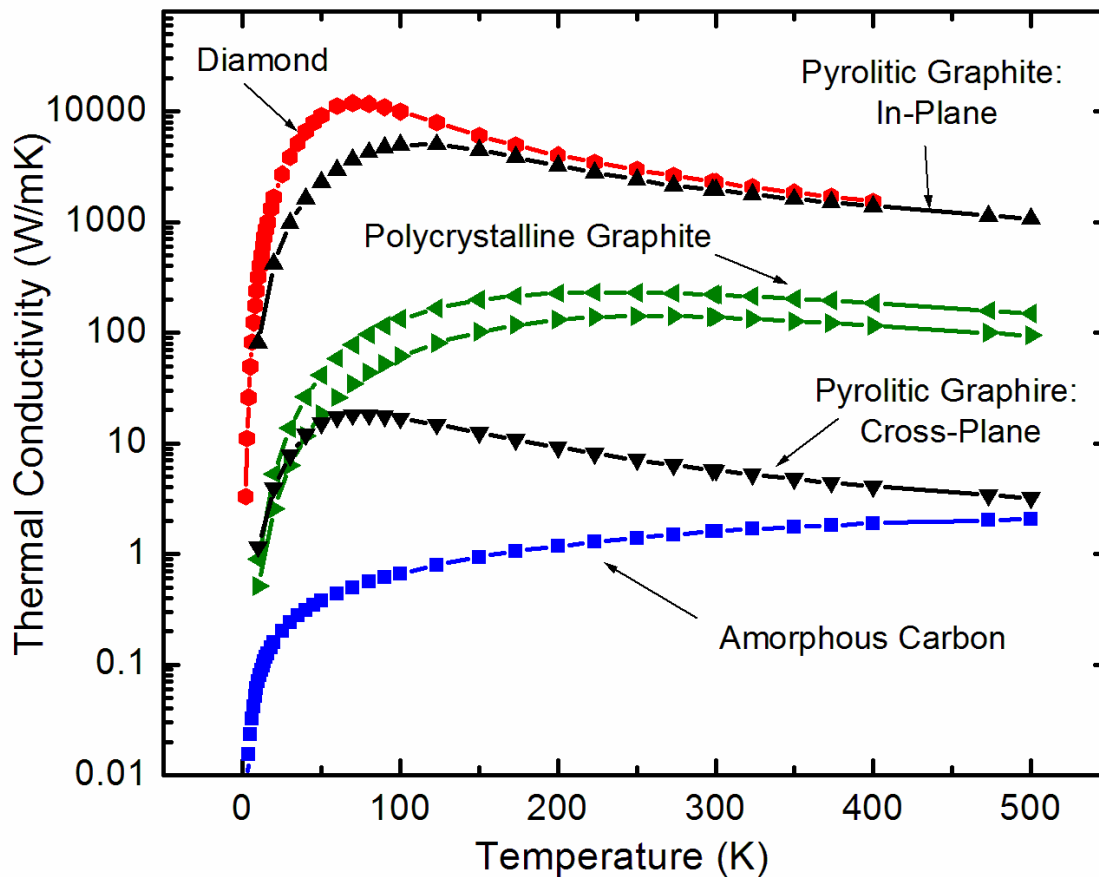
Graphene and FLG as the Fillers in Thermal Interface Materials



- Graphene for thermal applications can be mass-production using LPE and GO reduction
- The quality requirements for thermal graphene are much less strict that for electronic graphene
- Graphene and FLG mixtures can be byproducts of other mass production technologies

J. S. Lewis, T. Perrier, Z. Barani, F. Kargar, and A. A. Balandin, Nanotechnology, 31, 142003 (2021).

Thermal Conductivity of Bulk Carbon Materials



← Bulk graphite: 2000 W/mK at RT

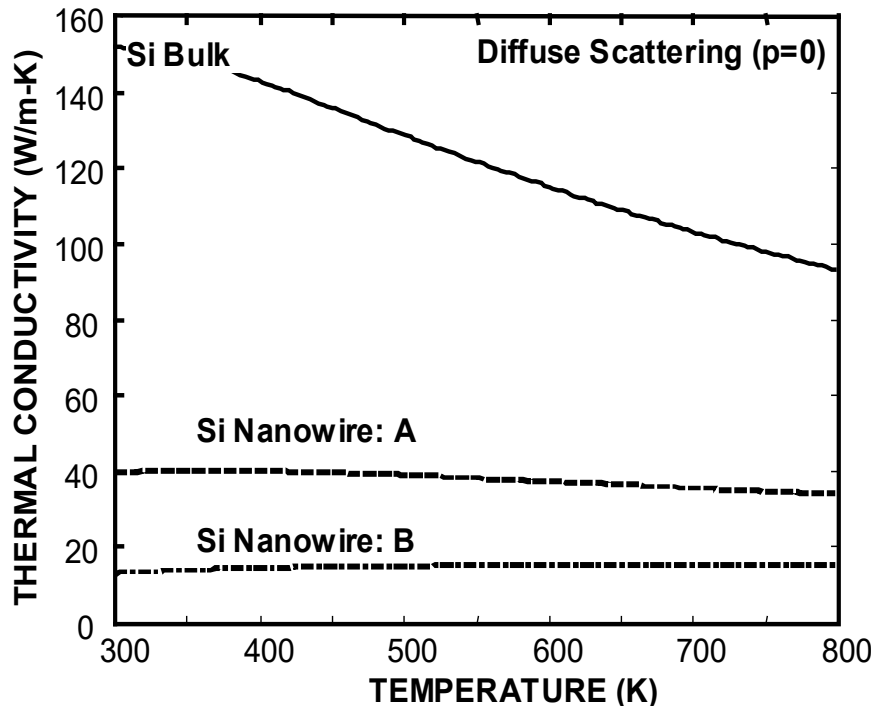
← Order of magnitude difference in high-quality graphite depending on the method and polycrystallinity

A.A. Balandin, "Thermal properties of graphene and nanostructured carbon materials," Nature Materials, 10, 569 (2011).

What happens with thermal conductivity of 3D crystal if we thin it down to 2D crystal?

Thermal Conductivity at Nanoscale: Extrinsic Phonon Transport Regime

Thermal conductivity usually decreases as one goes from bulk material to thin films and nanostructure due to phonon – boundary scattering and possible phonon spectrum change



← Thermal conductivity of bulk Si at room temperature: $K = 148 \text{ W/m-K}$

← Thermal conductivity of Si nanowire with cross section of 20 nm x 20 nm: $K = 13 \text{ W/mK}$

→ Phonon thermal conductivity:

$$K_p = (1/3)C_p v \Lambda$$

→ Boundary-limited MFP ($\Lambda = v\tau$):

$$\frac{1}{\tau_B} = \frac{v}{D} \frac{1-p}{1+p}$$

J. Zou and A.A. Balandin, J. Appl. Phys., 89, 2932 (2001). $K_p \sim C_p v \Lambda \sim C_p v^2 \tau_B \sim C_p v D$ 8

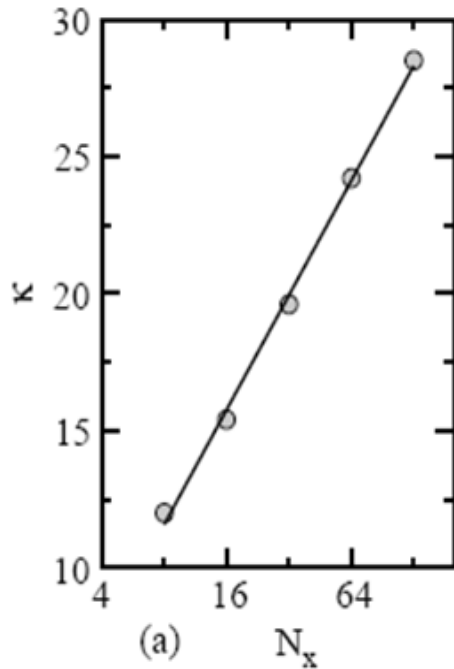
Thermal Conductivity of 2D Crystals in Intrinsic Phonon Transport Regime



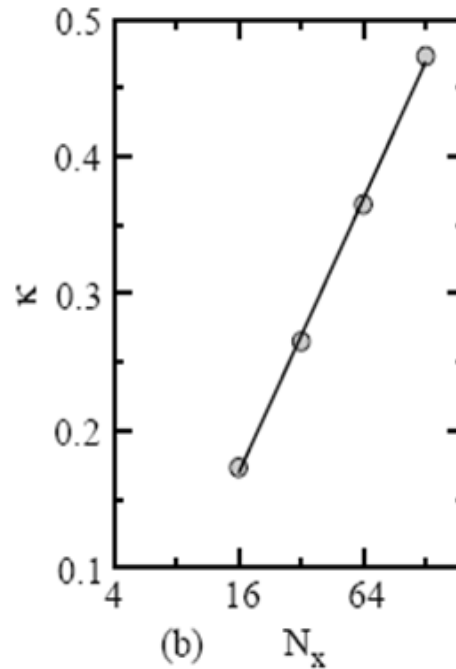
- The momentum conservation in 1D and 2D systems with anharmonicity leads to the divergence of the intrinsic thermal conductivity K with the system size
- Thermal conductivity remains finite and does not depend on the system size in 3D

Divergence of the Lattice Thermal Conductivity in 2D and 1D Crystal Lattices

The intrinsic thermal conductivity of 2-D or 1-D anharmonic crystals is anomalous



Thermal conductivity in 2D lattice vs. N_x .
S. Lepri et al. *Phys. Rep.*, 377, 1 (2003).



$K \sim \log(N)$ in 2D

$K \sim N^\alpha$ in 1D, $\alpha \neq 1$

N – system size

- [1] K. Saito, et al., *Phys. Rev. Lett.* (2010).
- [2] A. Dhar. *Advances in Physics* (2008).
- [3] G. Basile et al. *Eur. Phys. J.* (2007).
- [4] L. Yang et al. *Phys. Rev. E* (2006).
- [5] L. Delfini et al. *Phys. Rev. E* (2006).
- [6] S. Lepri et al. *Chaos* (2005).
- [7] J. Wang et al., *Phys. Rev. Lett.* (2004).
- [8] S. Lepri et al. *Phys. Rep.* (2003).
- [9] R. Livi and S. Lepri. *Nature* (2003).
- [10] O. Narayan et al., *Phys. Rev. Lett.* (2002).
- [11] A. Dhar. *Phys. Rev. Lett.* (2001).
- [12] T. Pozen et al., *Phys. Rev. Lett.* (2000).

Prior Knowledge from Carbon Nanotubes – Theory and Experiment

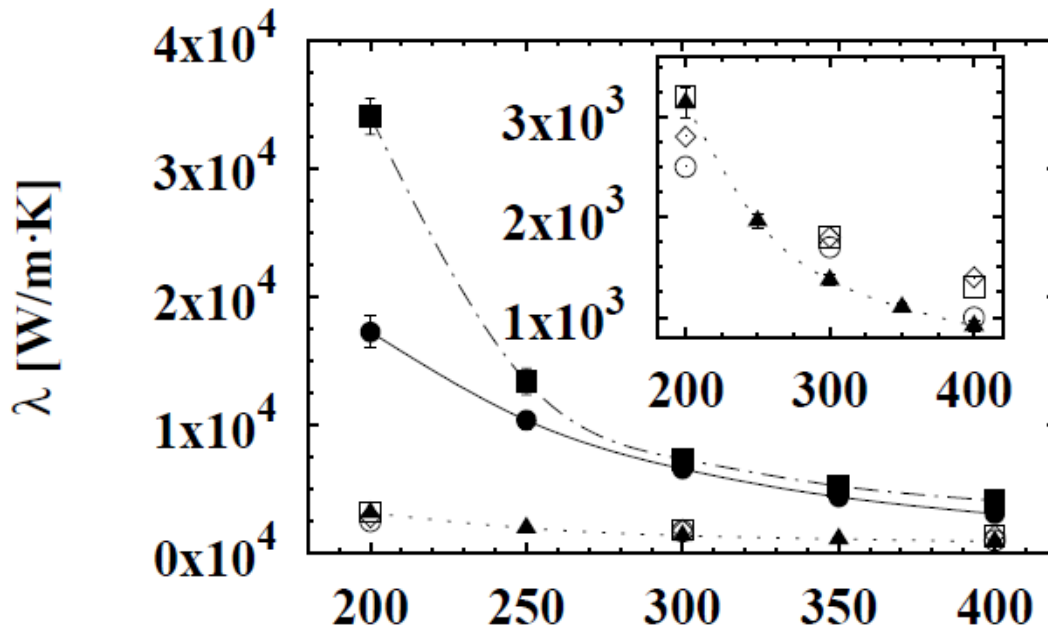


FIG. 3. Thermal conductivity λ for a (10, 10) carbon nanotube (solid line), in comparison to a constrained graphite monolayer (dash-dotted line), and the basal plane of AA graphite (dotted line) at temperatures between 200 and 400 K. The inset reproduces the graphite data on an expanded scale.

Thermal conductivity of graphene should be larger than that of CNT

S. Berber, Y.-K. Kwon, and D. Tománek, Phys. Rev. Lett. 84, 4613 (2000).

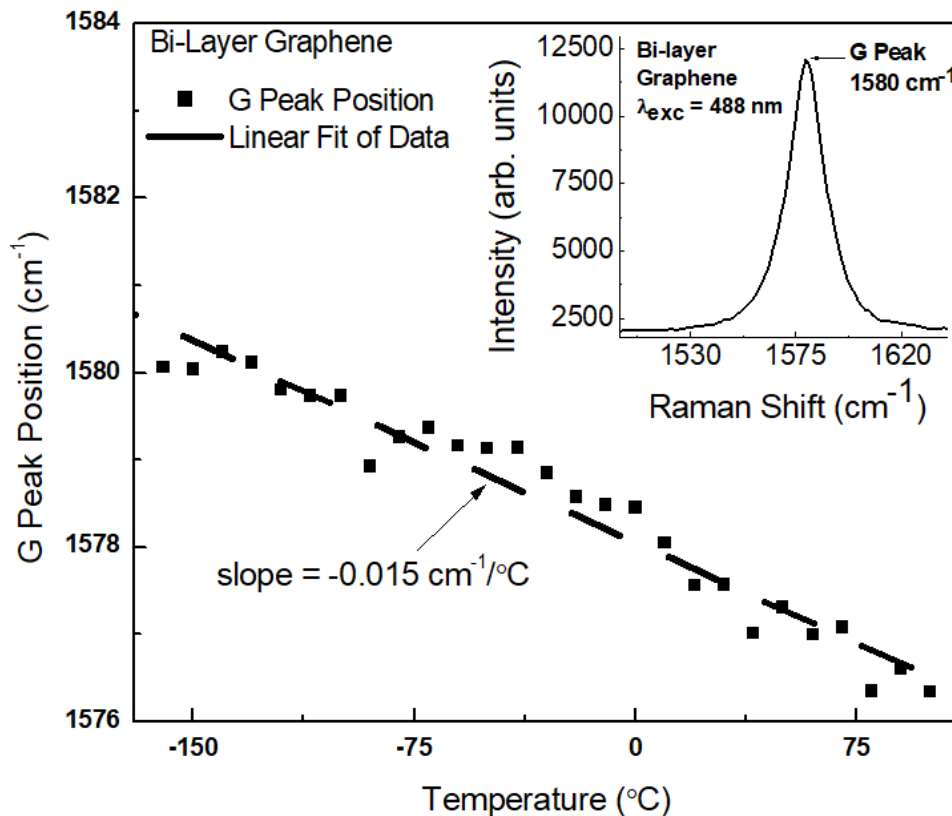
CNT: ~1758 W/mK – 5800 W/mK

According to J. Hone, M. Whitney, C. Piskoti, A. Zettl, A. Phys. Rev. B 1999, R2514 (1999)

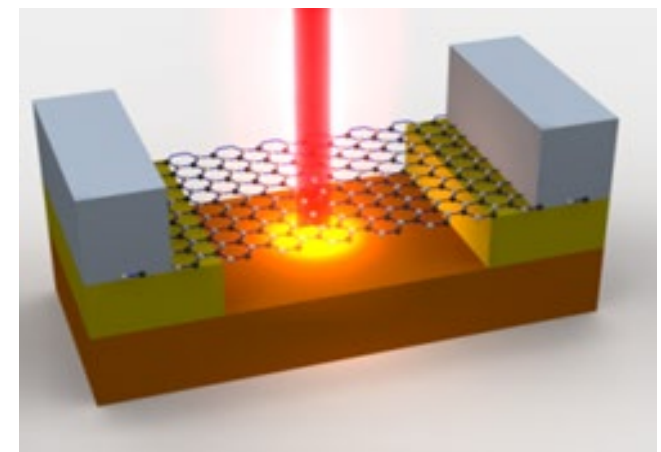
How to measure thermal transport in 2D crystal?

Electrical and optical trade-offs: More accurate techniques result in more defects to the samples 11

From Raman Spectroscopy of Graphene to Thermal Measurement

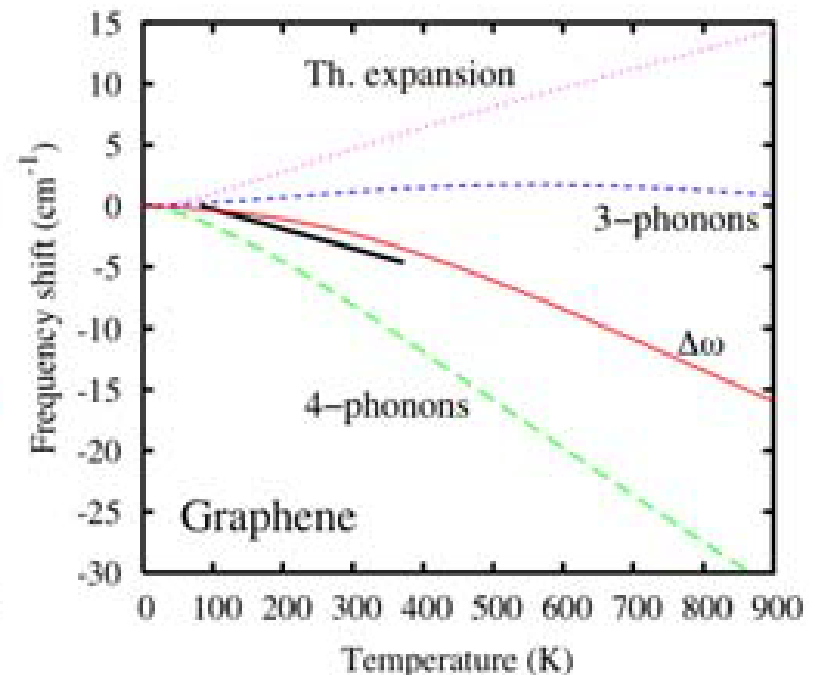
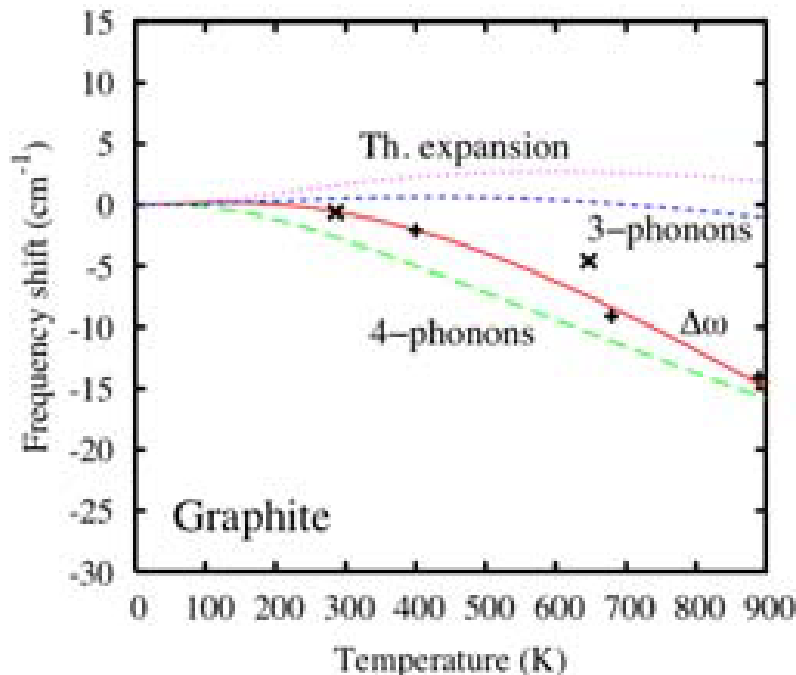


I. Calizo, et al., Nano Lett., 7, 2645 (2007).



- Raman spectrometer can act as a thermometer if one has a calibration curve
- Graphene thermal conductivity is expected to be high but the cross-section is small allowing for local heating
- Issues: sign of the slope, power absorbed

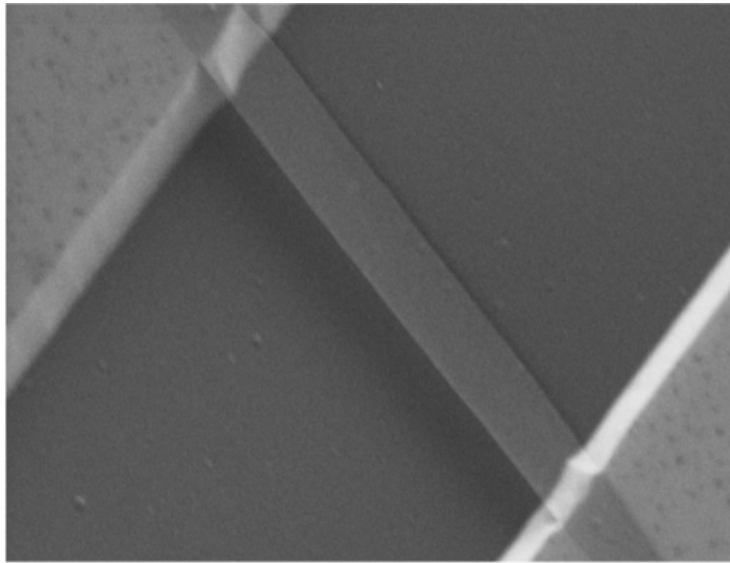
Temperature Shift of the Raman G Peak in Graphene



N. Bonini et al., Phys. Rev. Lett., 99, 176802 (2007); phys. stat. sol. (b), 245, 2149 (2008)

→ Even if the thermal expansion coefficient is negative and the in-plane lattice constant decreases with increasing T due to membrane effect, our phonon peak shift is correct

Optothermal Measurement of Graphene Thermal Conductivity

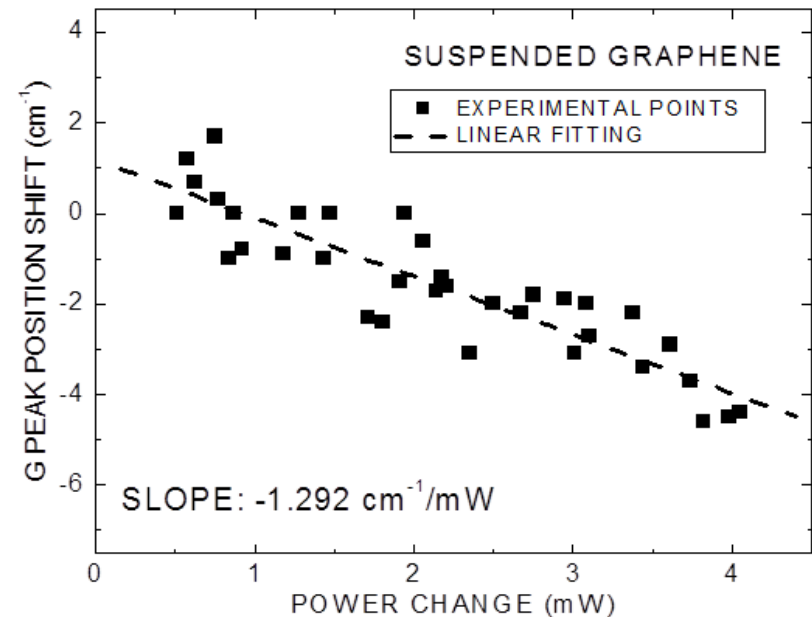


Bilayer graphene ribbon bridging 3- μm trench in Si/SiO₂ wafer

$$K = (L / 2a_G W) \chi_G (\Delta\omega / \Delta P_G)^{-1}.$$

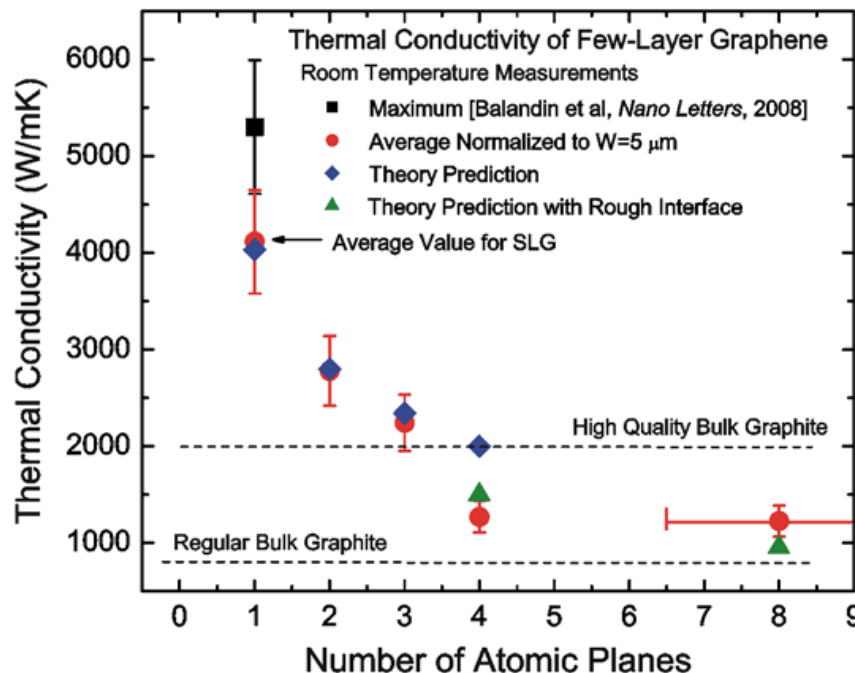
Connect $\Delta P_D \leftrightarrow \Delta P_G$ through calibration

- Laser acts as a heater: ΔP_G
- Raman “thermometer”: $\Delta T_G = \Delta\omega / \chi_G$
- Thermal conductivity: $K = (L / 2a_G W) (\Delta P_G / \Delta T_G)$

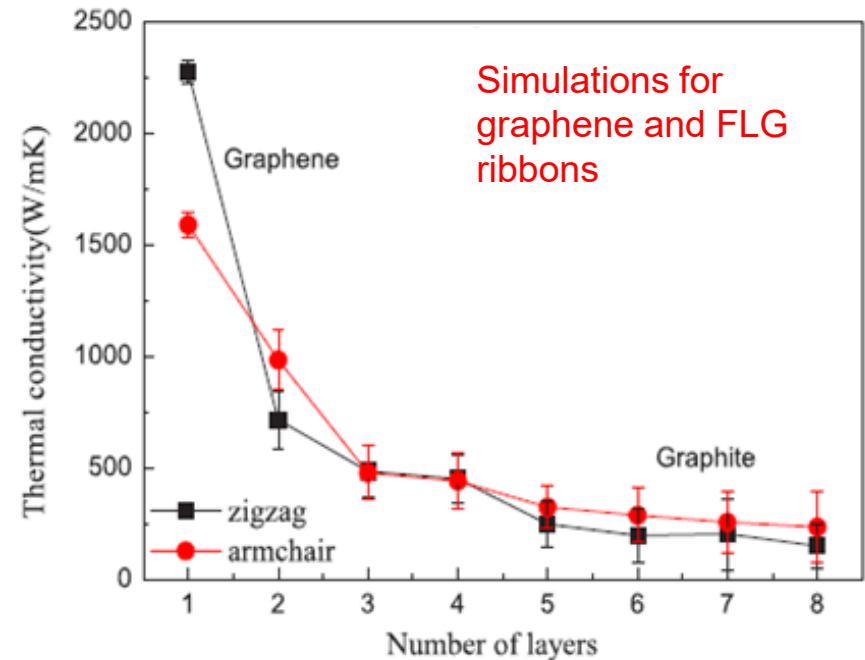


Evolution of the Intrinsic Thermal Conductivity in Low-Dimensional Systems

Our Experiments in 2008 – 2010



Molecular Dynamics Simulations



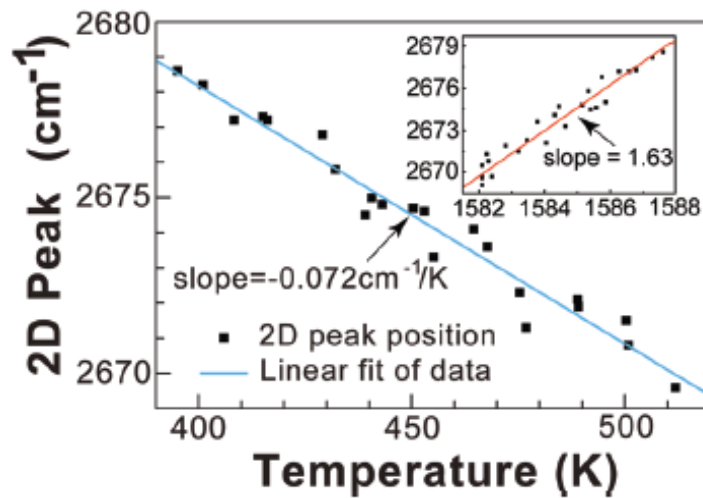
W.-R. Zhong et al., *Appl. Phys. Lett.*, 98, 113107 (2011).

Numerous publications that recover this type of reverse dependence with the thickness

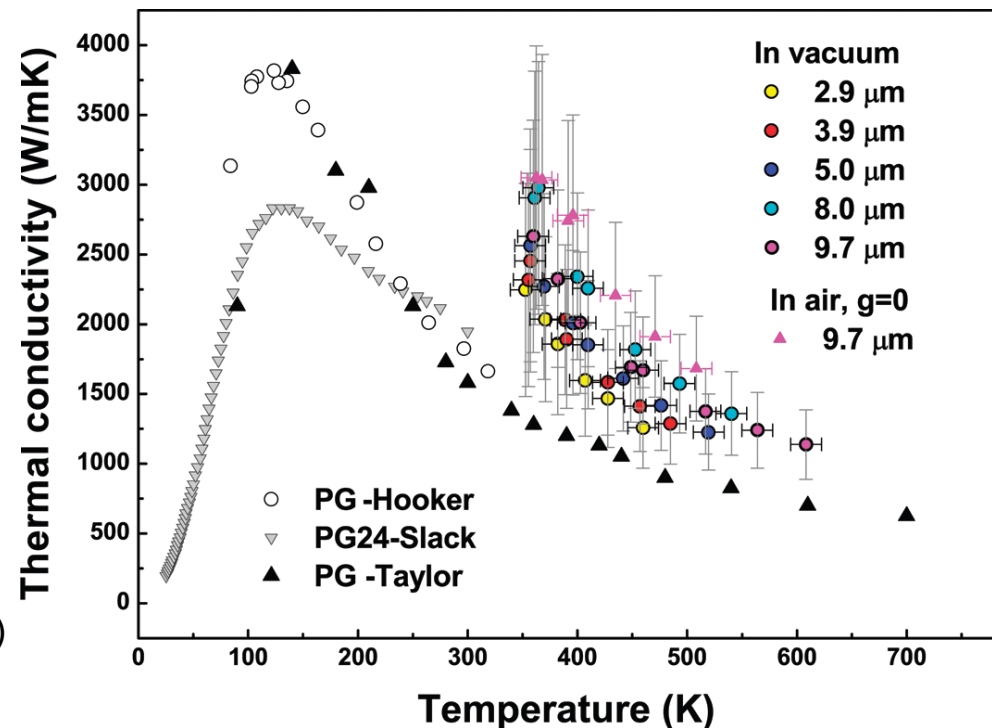
S. Ghosh, W. Bao, D. L. Nika, S. Subrina, E. P. Pokatilov, C. N. Lau, and A. A. Balandin, *Nature Mater.*, 9, 555 (2010).

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Independent Confirmations and Refinements of the Optothermal Technique



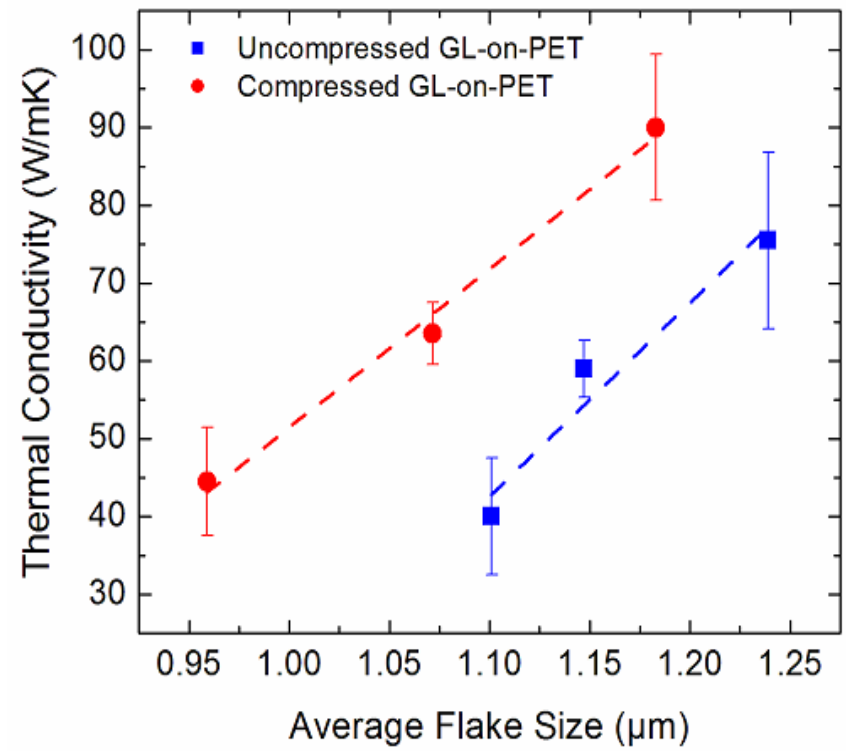
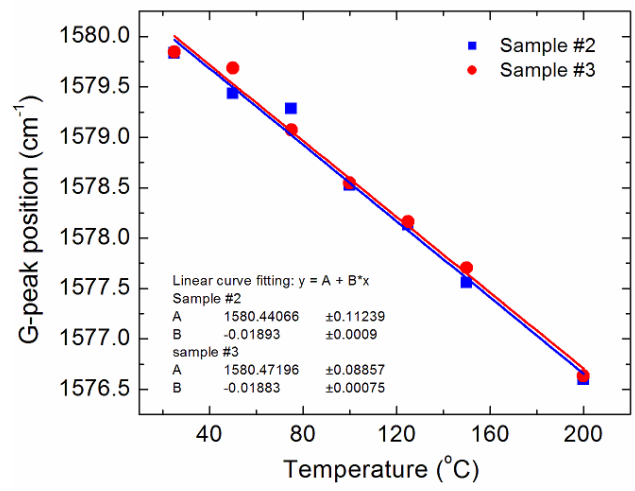
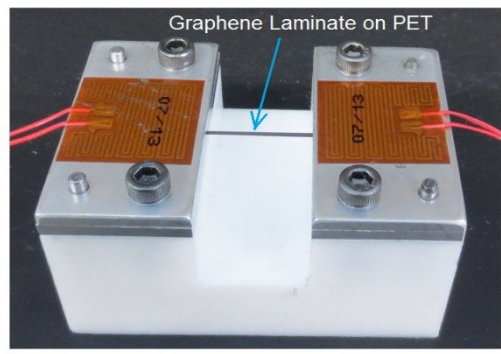
S. Chen, et al., ACS Nano, 5, 321 (2011)



- Detector can be placed under the sample
- Uncertainty assessment for the method
- The technique can be used with many 2D materials

How to explain the high values of the thermal conductivity?

Optothermal Methods can be Scaled Up - Graphene Laminate Coating



H. Malekpour, K.-H. Chang, J.-C. Chen, C.-Y. Lu, D.L. Nika, K.S. Novoselov and A.A. Balandin, Nano Lett., 14, 5155 (2014)

Theory of the Phonon Thermal Conductivity

The phonon heat flux: $\vec{W} = \sum_{s, \vec{q}} \vec{v}(s, \vec{q}) \hbar \omega_s(\vec{q}) N(\vec{q}, \omega_s(\vec{q})) = \sum_{s, \vec{q}} \vec{v}(s, \vec{q}) \hbar \omega_s(\vec{q}) n(\vec{q}, \omega_s)$

$$\vec{W} = - \sum_{\beta} (\nabla T)_{\beta} \sum_{s, \vec{q}} \tau v_{\beta}(s, \vec{q}) \frac{\partial N_0(\omega_s)}{\partial T} \vec{v}(s, \vec{q}) \hbar \omega_s(\vec{q}) \quad \text{Relaxation-time approximation}$$

Definition of the thermal conductivity:

$$W_{\alpha} = -\kappa_{\alpha\beta} (\nabla T)_{\beta}$$

The expression for thermal conductivity is obtained via Boltzmann's equation:

$$\kappa = \frac{1}{4\pi k T^2 h} \times \sum_{s=1\dots6} \int_0^{q_{\max}} \{ \hbar \omega_s(q) v_s(q) \}^2 \tau_{\text{tot}}(s, q) \frac{\text{Exp}(\hbar \omega_s(q) / kT)}{(\text{Exp}(\hbar \omega_s(q) / kT) - 1)^2} q \} dq$$

Scattering processes included into consideration:

$$\frac{1}{\tau_{\text{tot}}(s, q)} = \frac{1}{\tau_U(s, q)} + \frac{1}{\tau_B(s, q)} + \frac{1}{\tau_{Pd}(s, q)}$$

Goal: capture the specifics of the 2D material

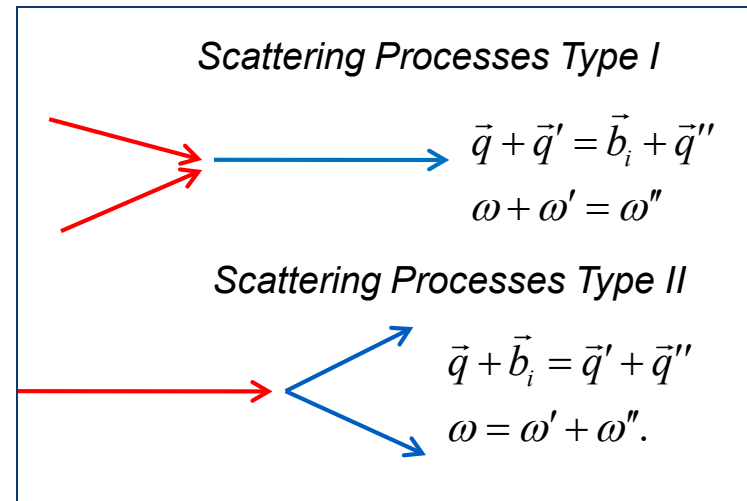
Phonon Scattering in Graphene: Three-Phonon Umklapp, Defects and Edges

Phonon Scattering Rates:

$$\frac{1}{\tau_{tot}(s, q)} = \frac{1}{\tau_U(s, q)} + \frac{1}{\tau_B(s, q)} + \frac{1}{\tau_{Pd}(s, q)}$$

$$\frac{1}{\tau_U(s, \vec{q})} = \frac{1}{\tau_U^I(s, \vec{q})} + \frac{1}{\tau_U^{II}(s, \vec{q})}$$

2D system: different ω dependence

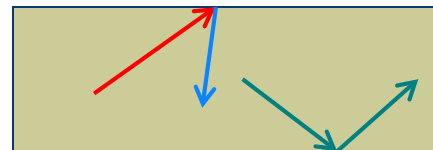


Point Defect Scattering

$$\frac{1}{\tau_{PD}(s, q)} = \frac{S_0 \Gamma}{4} \frac{q_s(\omega_s)}{v_s(\omega_s)} \omega_s^2,$$

$$\Gamma = \sum_i f_i (1 - M_i / \bar{M})^2$$

Edge Roughness Scattering



$$\frac{1}{\tau_B(s, q)} = \frac{v_s(\omega_s)}{d} \frac{1-p}{1+p}$$

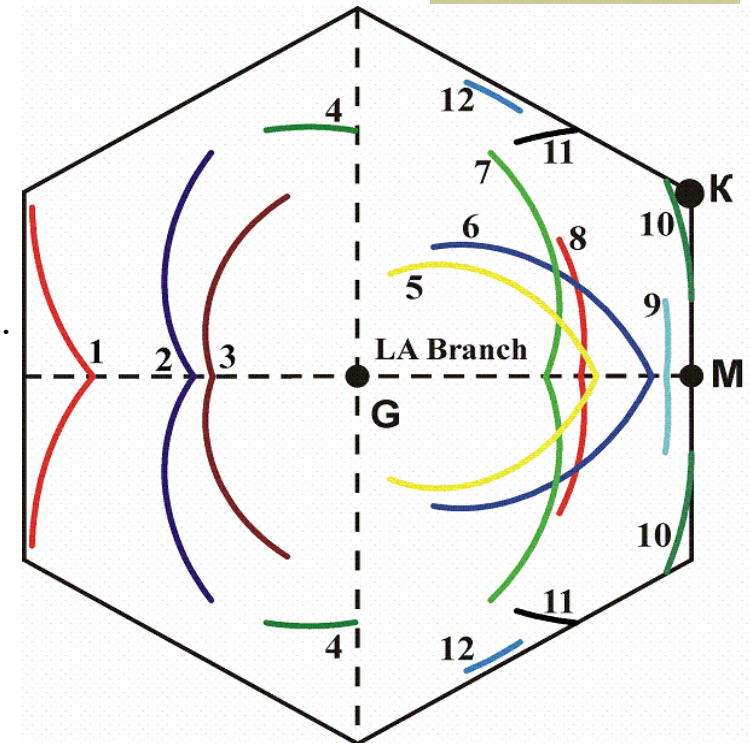
p is the specularity parameter

Three-Phonon Umklapp Scattering Diagram Technique

Accounting of all possible phonon transitions, which satisfy energy – momentum selection rules

$$\frac{1}{\tau_U^{(I)(II)}(s, \vec{q})} = \frac{\hbar \gamma_s^2(\vec{q}) \omega_s(\vec{q})}{3\pi \rho v_s^2(\vec{q})} \sum_{s's''; \vec{b}} \int_l \frac{\pm(\omega_{s''} - \omega_s) \omega_{s''}}{v_{\perp}(\omega_{s'})} (N_0' \mp N_0'' + \frac{1}{2} \mp \frac{1}{2}) dq_l'$$

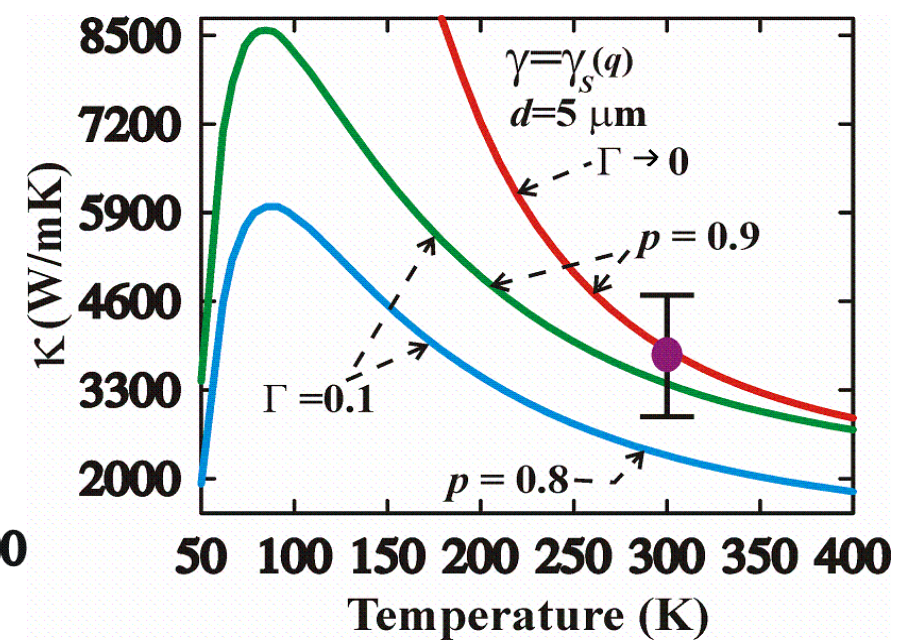
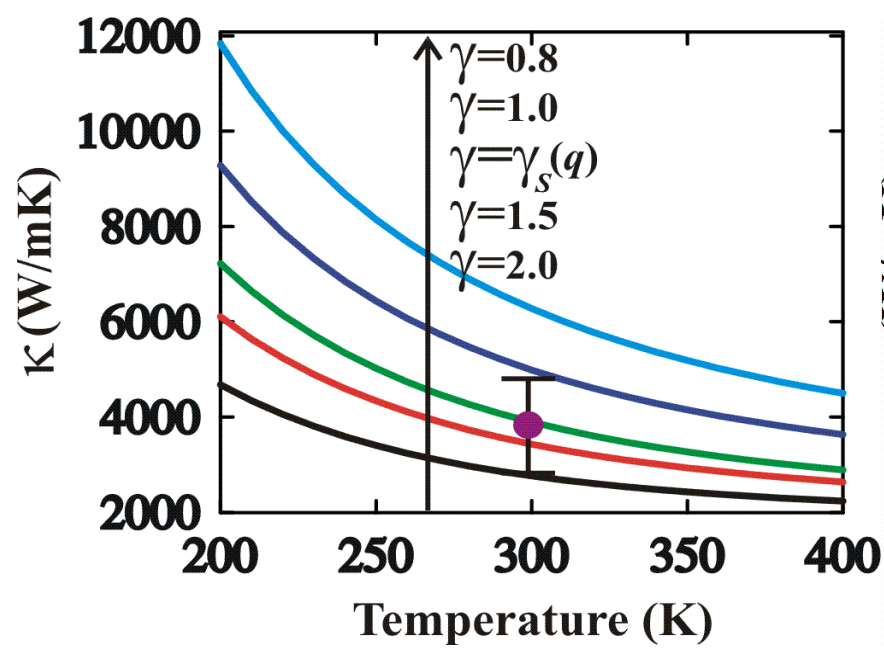
$$\kappa = \frac{1}{4\pi kT^2 h} \times \sum_{s=1...6} \int_0^{q_{\max}} \{ \hbar \omega_s(q) v_s(q) \}^2 \tau_{tot}(s, q) \frac{\text{Exp}(\hbar \omega_s(q) / kT)}{(\text{Exp}(\hbar \omega_s(q) / kT) - 1)^2} q \} dq$$



D.L. Nika, E.P. Pokatilov,
A.S. Askerov and A.A.
Balandin, Phys. Rev. B 79,
155413 (2009)

- (1) LA → ZO + TA; (2) LA → ZO + ZA; (3) LA → TA + ZO; (4) LA → ZO + ZA;
(5) LA + ZA → LO; (6) LA + ZA → TO; (7) LA + ZA → LA; (8) LA + ZO → TO; (9) LA + LA → TO;
(10) LA + TA → LA; (11) LA + ZA → TA; (12) LA + TA → TO.

Phonon Thermal Transport in Graphene – Theory vs Experiment



- Adding defects helped to make the integral well-behaved
- Ambiguity in parameters

D.L. Nika, E.P. Pokatilov, A.S. Askerov and A.A. Balandin, Phys. Rev. B 79, 155413 (2009)

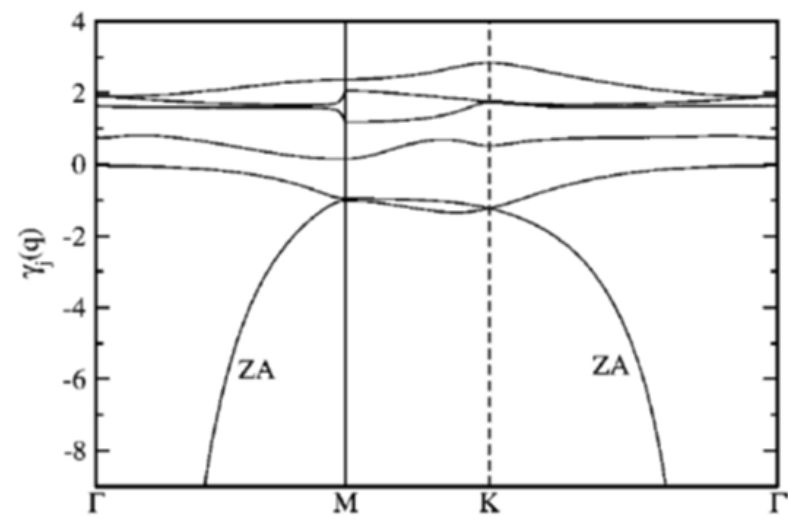
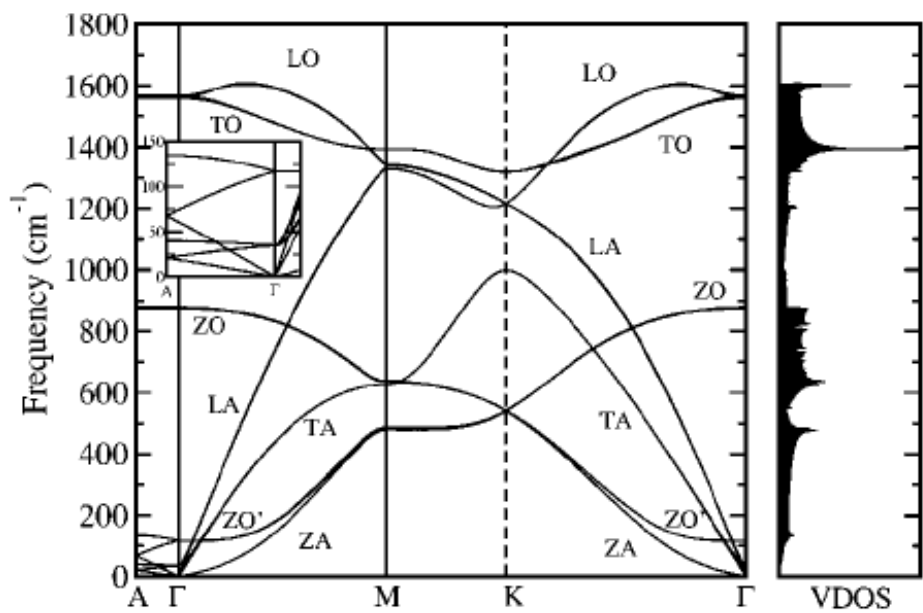
Simple intuitive model is needed for qualitative understanding

Existing Theory – Klemens Model of Heat Conduction in Bulk Graphite

Phonon Thermal Conductivity:

$$K_p = (1/3)C_p v \Lambda$$

$$K_p = \sum_j \int C_j(\omega) v_j^2(\omega) \tau_j(\omega) d\omega$$



Umklapp life-time, which defines MFP:

$$\tau_{U,s} = \frac{1}{\gamma_s^2} \frac{M v_s^2}{k_B T} \frac{\omega_{s,max}}{\omega^2}$$

P.G. Klemens, J. Wide Bandgap Materials, 7, 332 (2000).

2D: $C(\omega) \sim \omega \rightarrow K \sim T^{-1} \omega^1$
 3D: $C(\omega) \sim \omega^2$

P.G. Klemens Theory of Heat Conduction in Graphene

Theory of the A-Plane Thermal Conductivity of Graphite

P. G. Klemens

Journal of Wide Bandgap Materials 2000; 7; 332

DOI: 10.1106/7FP2-QBLN-TJPA-NC66

GRAPHENE

Similar considerations also apply to a single graphene sheet. Here the phonon gas is two-dimensional down to zero frequency, since there are no wave vectors outside the basal plane. However, a logarithmic divergence is also prevented, because the mean free path cannot exceed a linear dimension L , determined by the size and shape of the sheet. The factor $C(f)l(f)$ now increases with decreasing frequency only when $f > f_B$, where f_B is given by the condition that $l_i(f_B) = L$. Using Equation (3) this yields

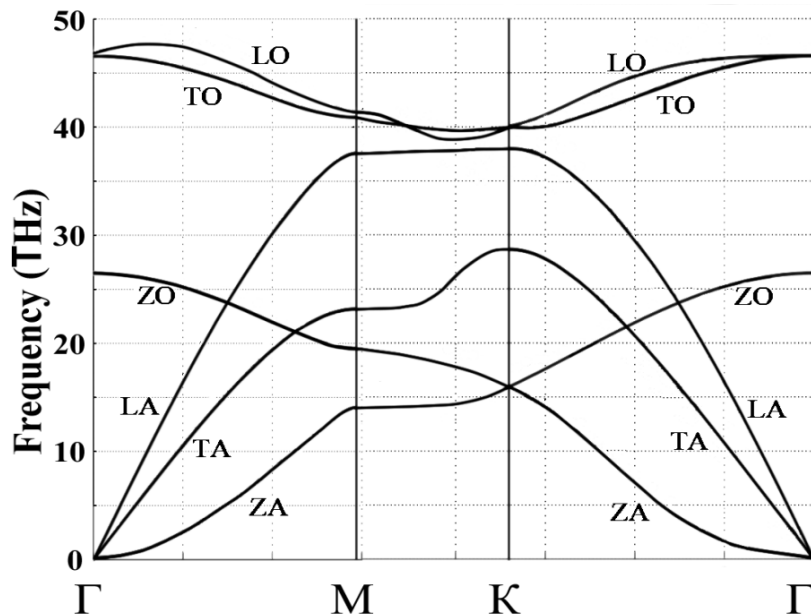
$$f_B^2 = \frac{1}{4\pi\gamma^2} \frac{Mv^2}{kT} \frac{vf_m}{L} \quad (21)$$

The Role of the Long-Wavelength Phonons in Heat Transport in Graphene

Thermal conductivity in graphene:

$$K \propto \frac{1}{\omega_m} \int_{\omega_c}^{\omega_m} \frac{d\omega}{\omega} \propto \frac{1}{\omega_m} \ln\left(\frac{\omega_m}{\omega_c}\right).$$

Graphene:



MFP = L – physical size of the system

→ Limitation on MFL: $L = \tau v_s$

$$\tau_{U,s} = \frac{1}{\gamma_s} \frac{M v_s^2}{k_B T} \frac{\omega_{s,max}}{\omega^2}$$

→ Limiting low-bound frequency:

$$\omega_{s,min} = \frac{v_s}{\gamma_s} \sqrt{\frac{M v_s}{k_B T} \frac{\omega_{s,max}}{L}}$$

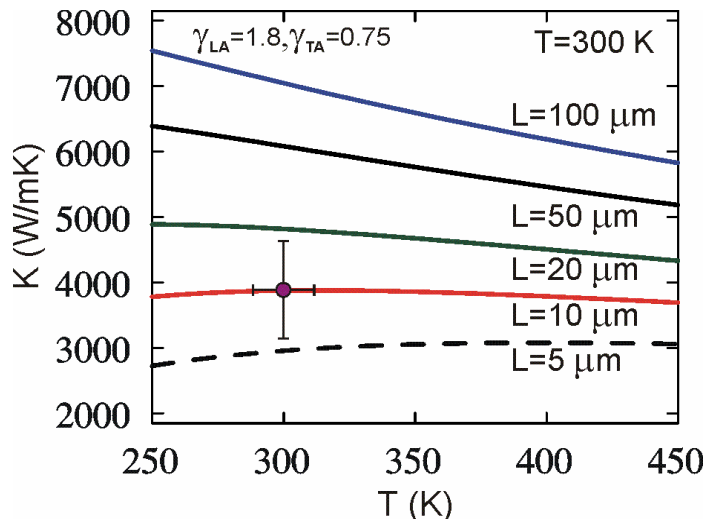
$$K = (2\pi\gamma^2)^{-1} \rho (v^4 / f_m T) \ln(f_m / f_B),$$

$$f_B = \left(M v^3 f_m / 4\pi\gamma^2 k_B T L \right)^{1/2}$$

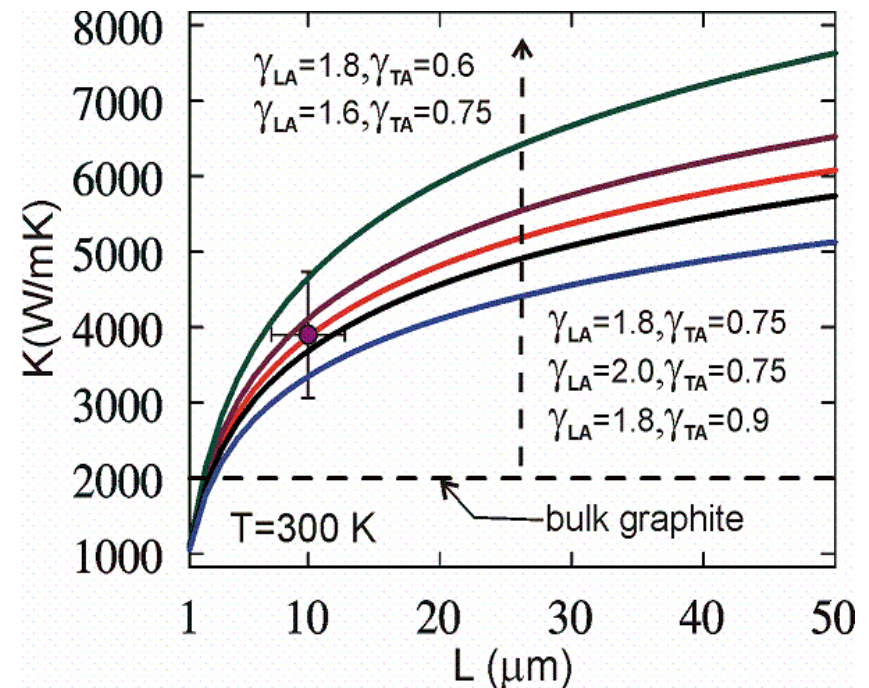
Uniqueness of Heat Conduction in Graphene – The Limits of Fourier’s Law

Phonon transport in graphene is 2D all the way down to zero frequency -- low-bound cut-off is by the condition that the phonon MFP can not exceed the physical size of the graphene flake:

$$\omega_{s,\min} = \frac{v_s}{\gamma_s} \sqrt{\frac{M v_s}{k_B T} \frac{\omega_{s,\max}}{L}}$$

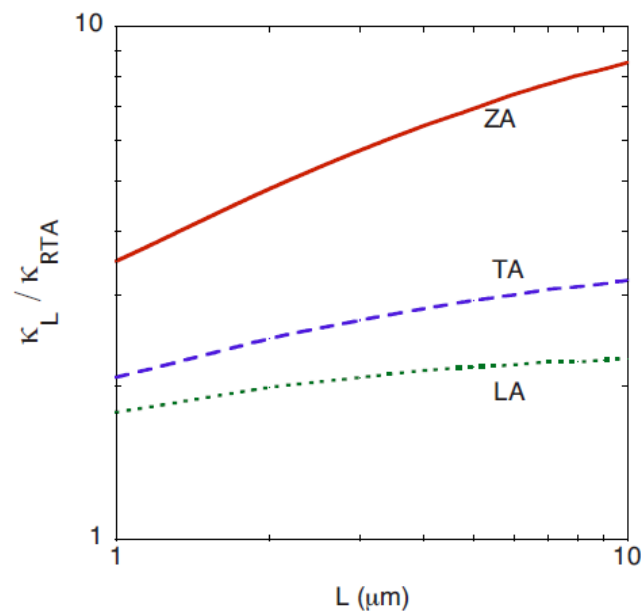
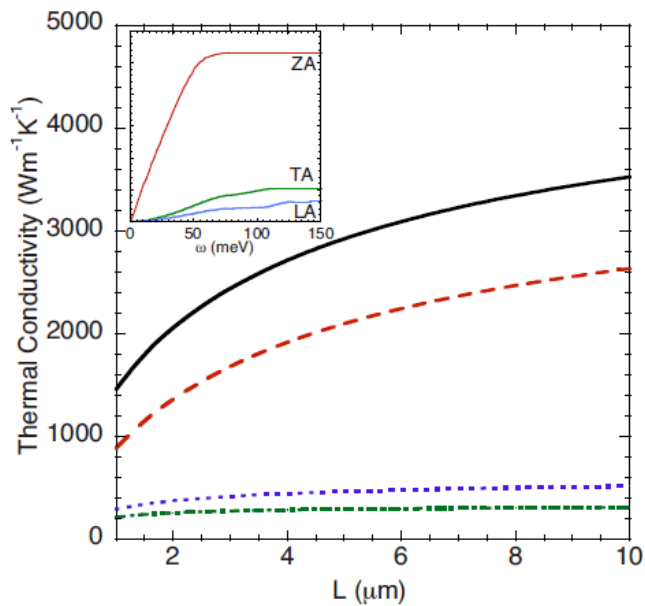


Breakdown of Fourier’s Law vs. Size-Dependent Intrinsic Thermal Conductivity



D.L. Nika, S. Ghosh, E.P. Pokatilov, A.A. Balandin, Appl. Phys. Lett., 94, 203103 (2009). 25

Contributions of the Out-of-Plane Phonons to Heat Conduction in Graphene



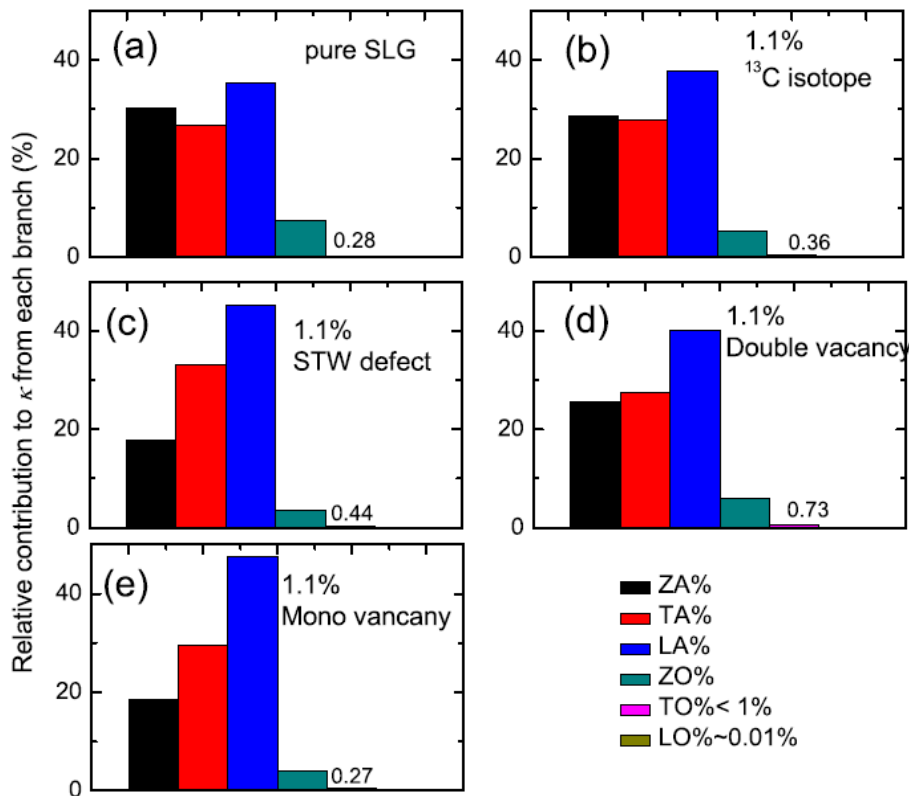
“ZA, or not ZA, that is the question”

L. Lindsay, D. A. Broido, and N. Mingo, Phys. Rev. B, 82, 115427 (2010).

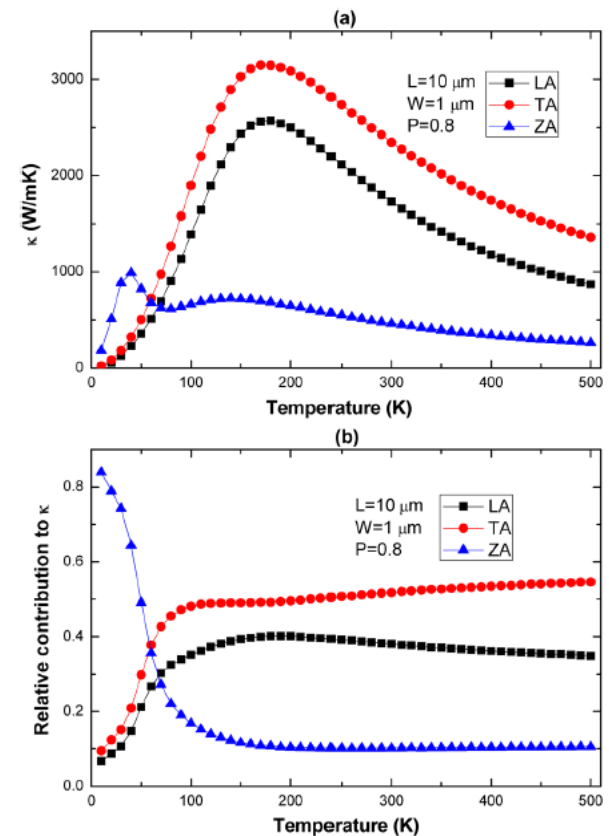
L. Lindsay, et al., Phys. Rev. B, 82, 161402R (2010).

- Large density of states of flexural phonons compared to their in-plane counterparts
- Symmetry-based selection rule that significantly restricts anharmonic phonon-phonon scattering of the flexural modes
- Theoretical thermal conductivity of graphene is size dependent: $\sim 2500 \text{ W/mK}$ at RT for $L=3 \mu\text{m}$
- The ballistic limit for graphene: $\sim 12800 \text{ W/mK}$

Relative Contribution of Different Phonon Modes to Thermal Transport

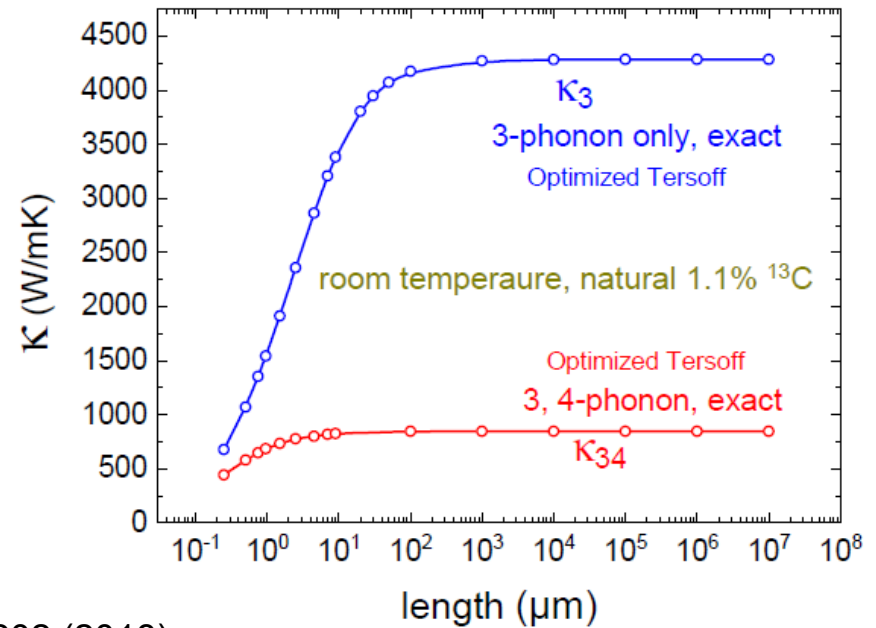
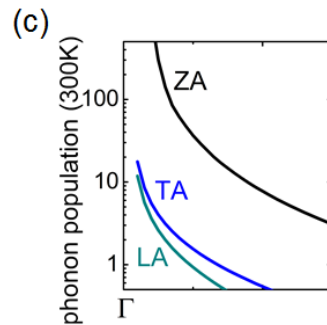
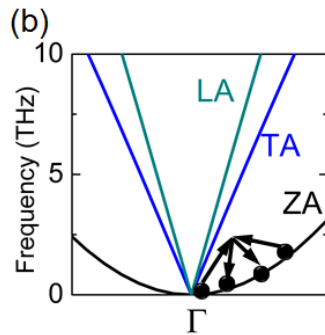
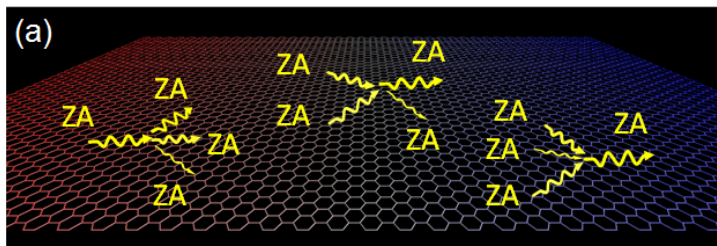


T. Feng et al., Phys. Rev. B, 91, 224301 (2015)



Y. Shen et al., J. Appl. Phys., 115, 063507 (2014) 27

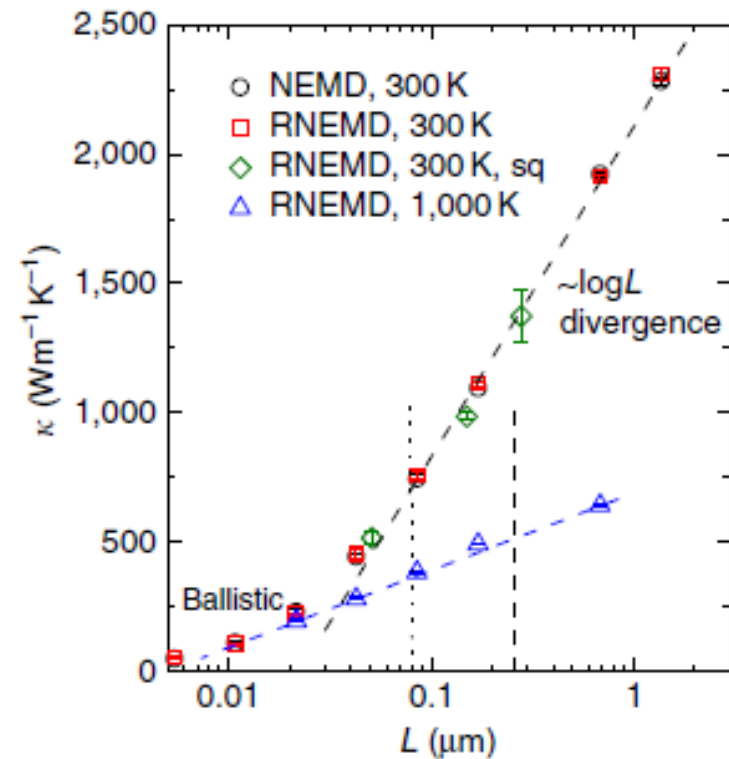
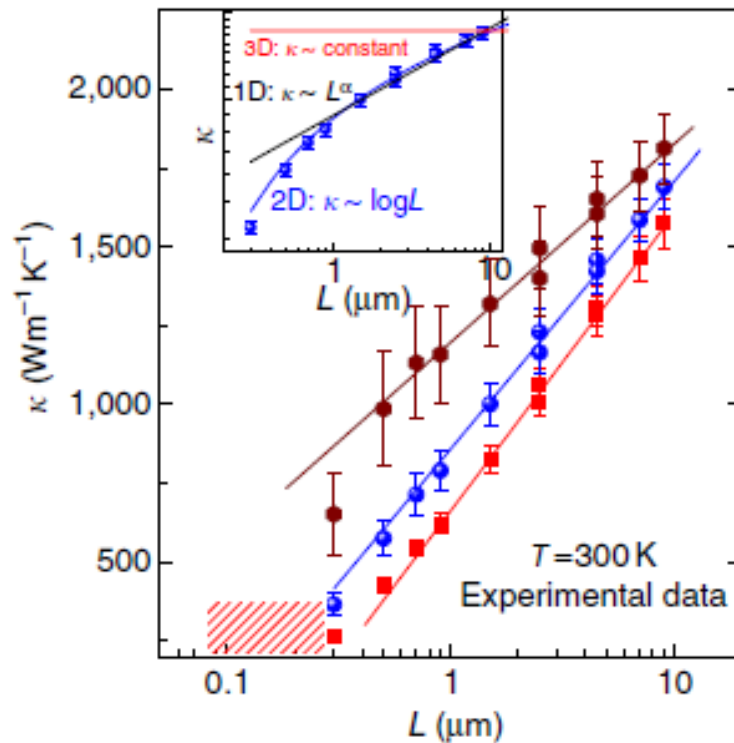
Importance of the Four-Phonon Umklapp Scattering Processes



T. Feng, and X. Ruan, Phys. Rev. B, 97, 045202 (2018)

- Reflection symmetry in graphene, which forbids three-ZA phonon scattering, allows four-ZA processes
- The relative contribution of the ZA phonon branch to heat conduction is correspondingly reduced to be lower than those of the LA and TA branches

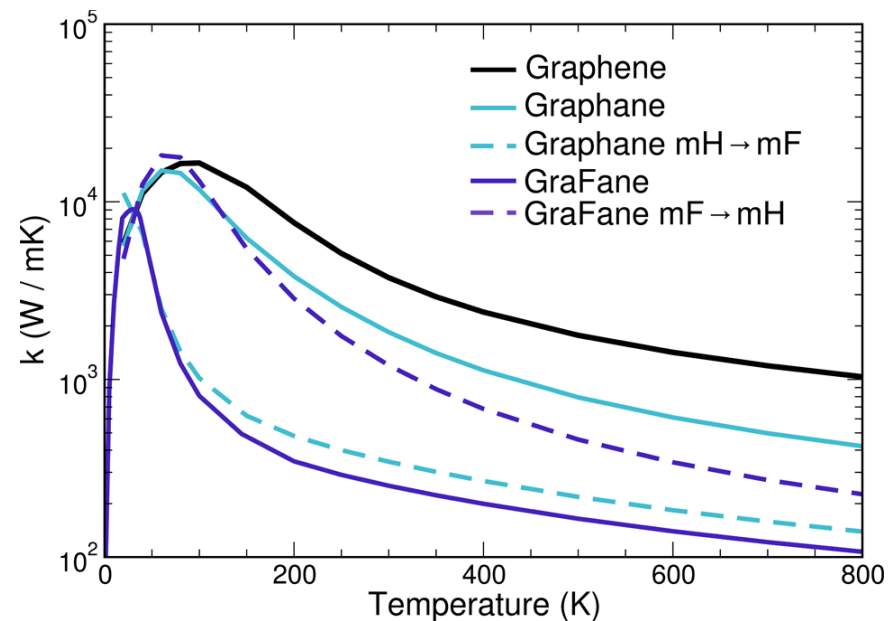
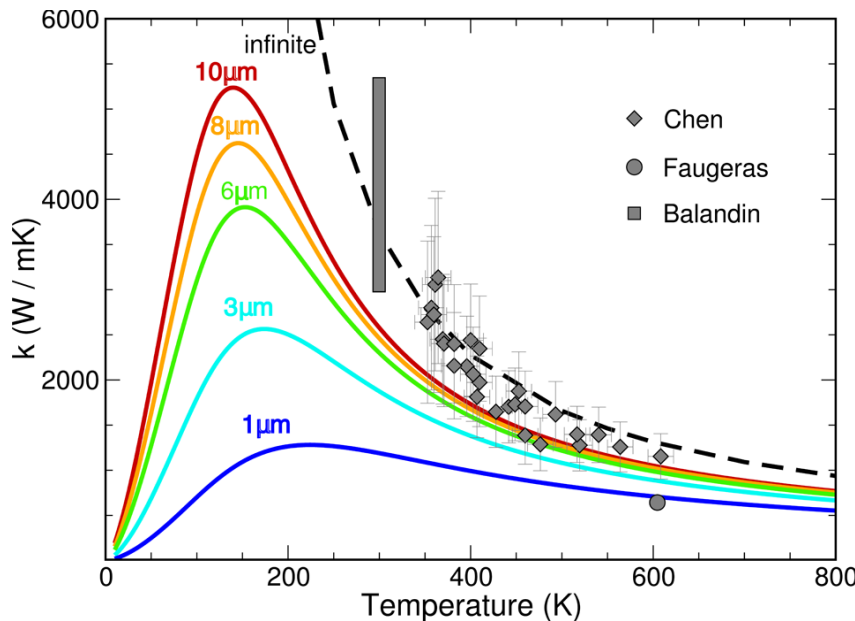
Size-Dependent Thermal Conductivity of Graphene – Experiments



X. Xu et al., Nature Com., 5, 3689 (2014)

- Size-dependence is different from the ballistic transport regime
- Graphene is not strictly 2D system because of ZA phonons

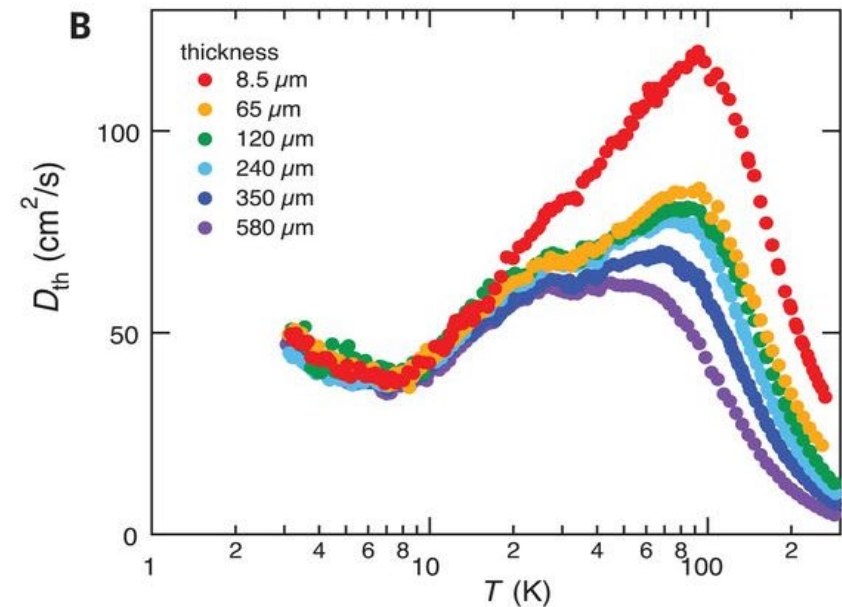
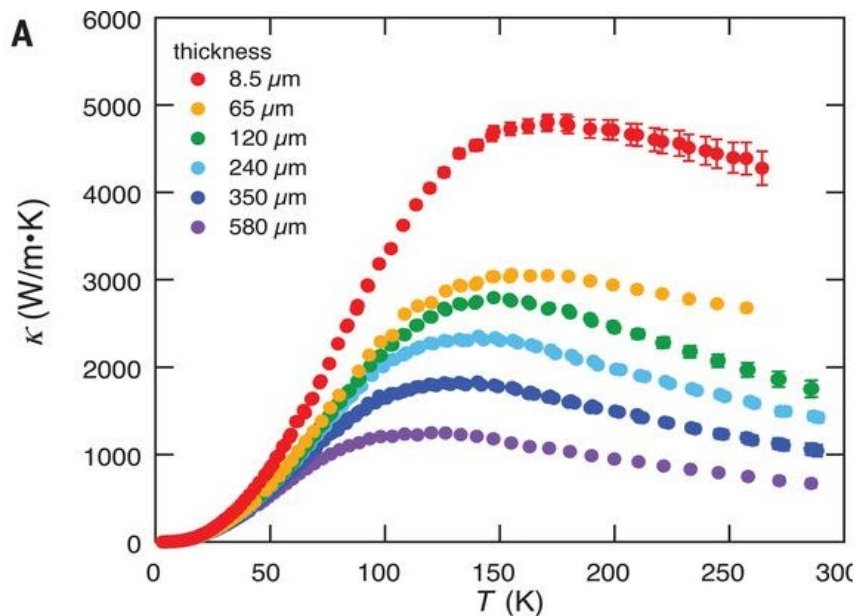
Size-Dependent Thermal Conductivity of Graphene – Theory



G. Fugallo, A. Cepellotti, L. Paulatto, M. Lazzeri, N. Marzari, and F. Mauri, Nano Lett., 14, 6109 (2014)

- The intrinsic thermal conductivity at RT can be obtained only for sample sizes of the order of 1 mm
- Collective phonon excitations, and not single phonons, are the main heat carriers in graphene
- These excitations have MFP of the order of hundreds of micrometers.

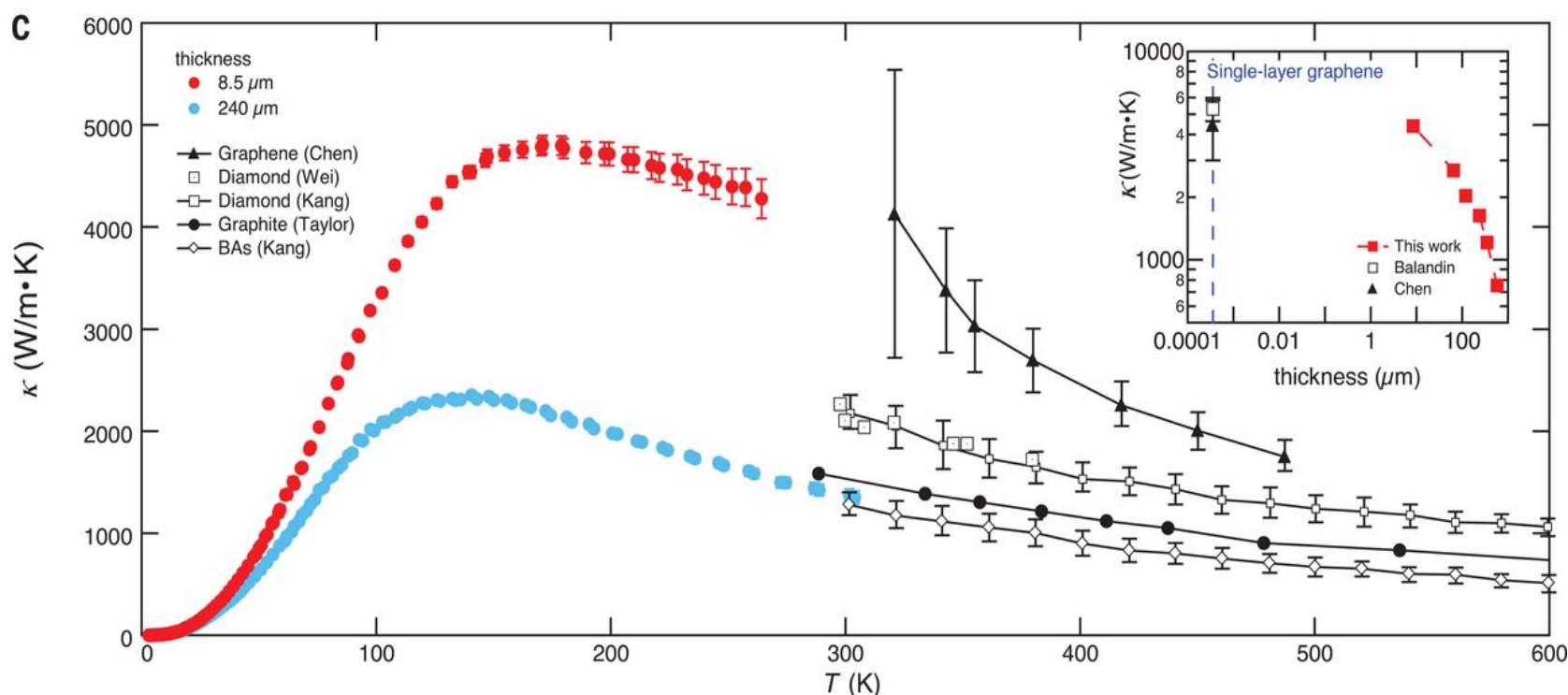
A New Turn – Phonon Hydrodynamic Transport in FLG and Graphite Thin Films



Y. Machida, et al., Science, 367, 309 (2020).

- Another possible explanation for the exceptionally high thermal conductivity of graphene and FLG – the phonon hydrodynamic transport and the closely related phenomenon of the second sound
- Phonons in the hydrodynamic regime can form packets that change the typical diffusive behavior of heat and make it propagate as a wave

Phonon Hydrodynamic Transport in FLG and Graphite Thin Films



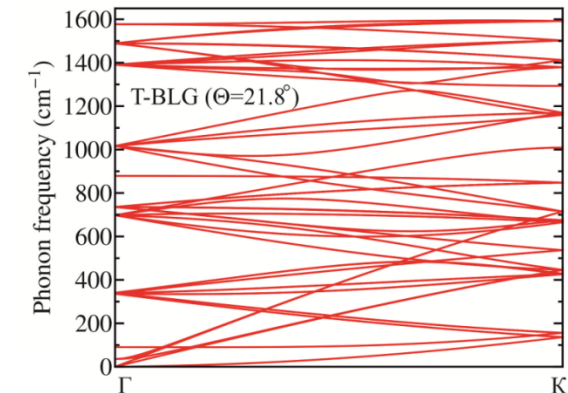
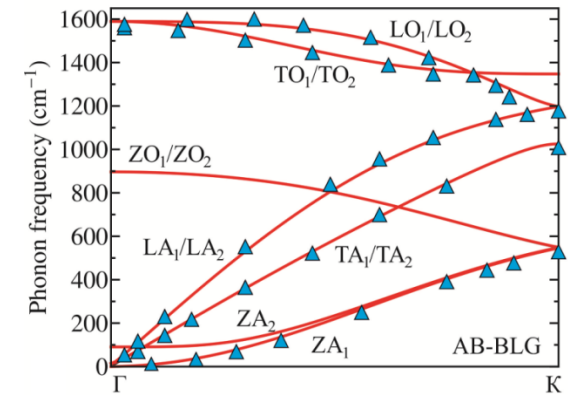
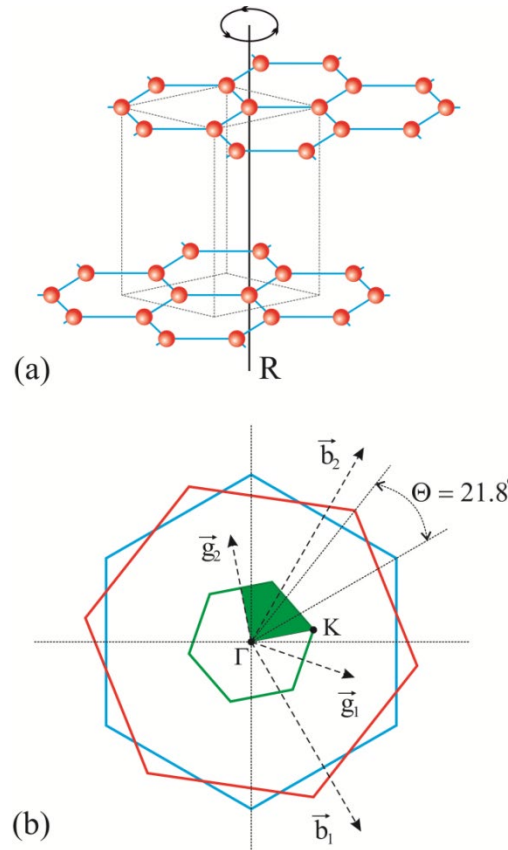
Y. Machida, et al., Science, 367, 309 (2020).

→ Further experimental work is needed to connect the thickness ranges and directly assess the hydrodynamic transport with Brillouin spectroscopy or other technique

Phonon Engineering by Twisting

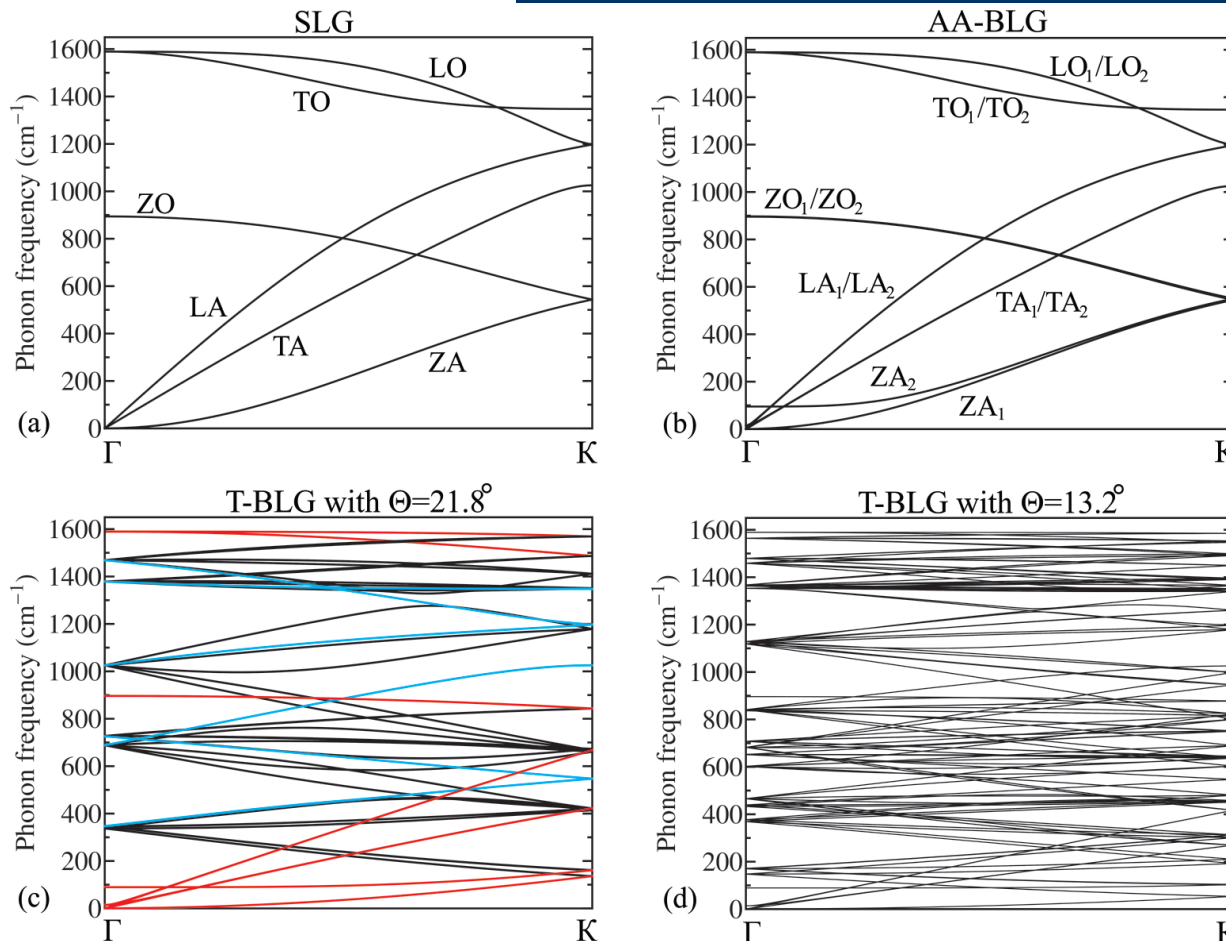


Twisting only weakly affects the interlayer interaction in van der Waals materials but it breaks the symmetry of the stacking



→ Electron properties as in single layer graphene
 → New peaks in the Raman spectra at $1100\text{-}1625 \text{ cm}^{-1}$ and $\sim 1375 \text{ cm}^{-1}$

Phonons in Twisted Bilayer Graphene

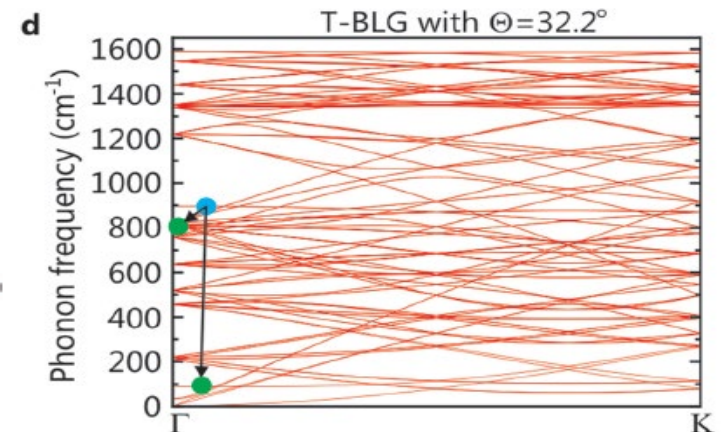
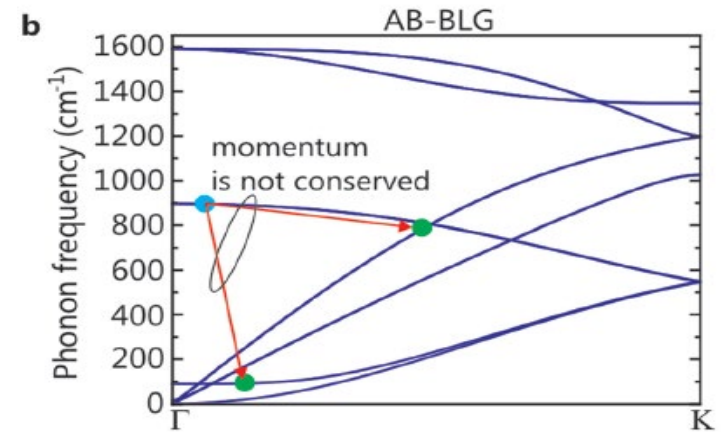
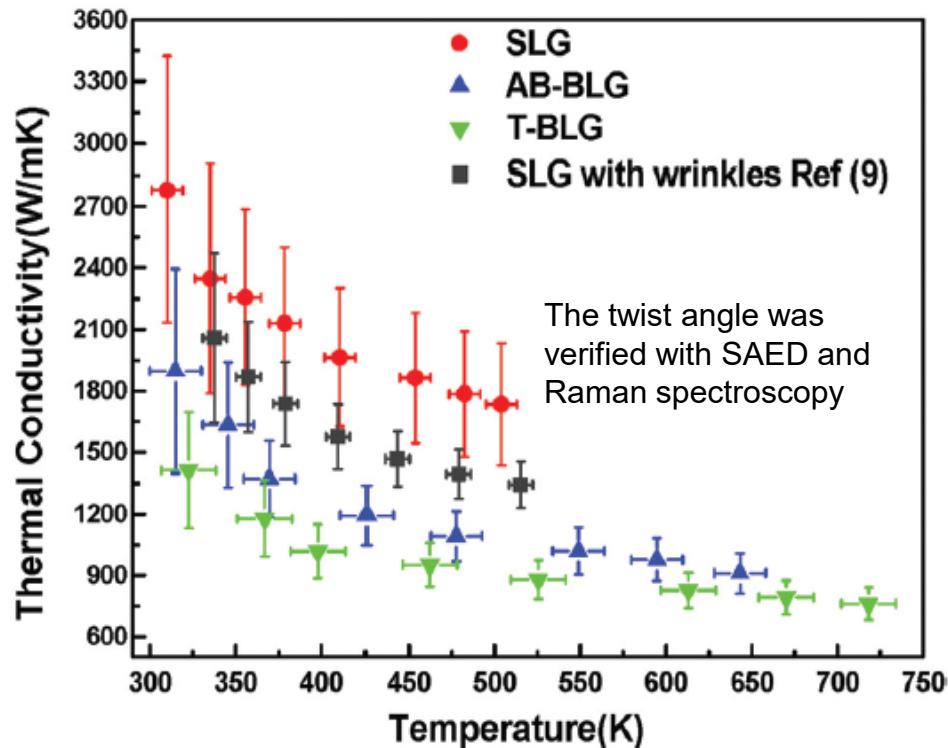


- In nanostructures one can engineer acoustic phonon modes by spatial confinement
- It is not possible to affect optical phonon modes
- Twisting of the layers in 2D materials allows one to engineer phonons from acoustic to optical energy range

A. I. Cocemasov, D. L. Nika, and A. A. Balandin, "Phonons in twisted bilayer graphene," *Phys. Rev. B*, 88, 35428 (2013).

Thermal Conductivity of Twisted BLG

Experimental Data

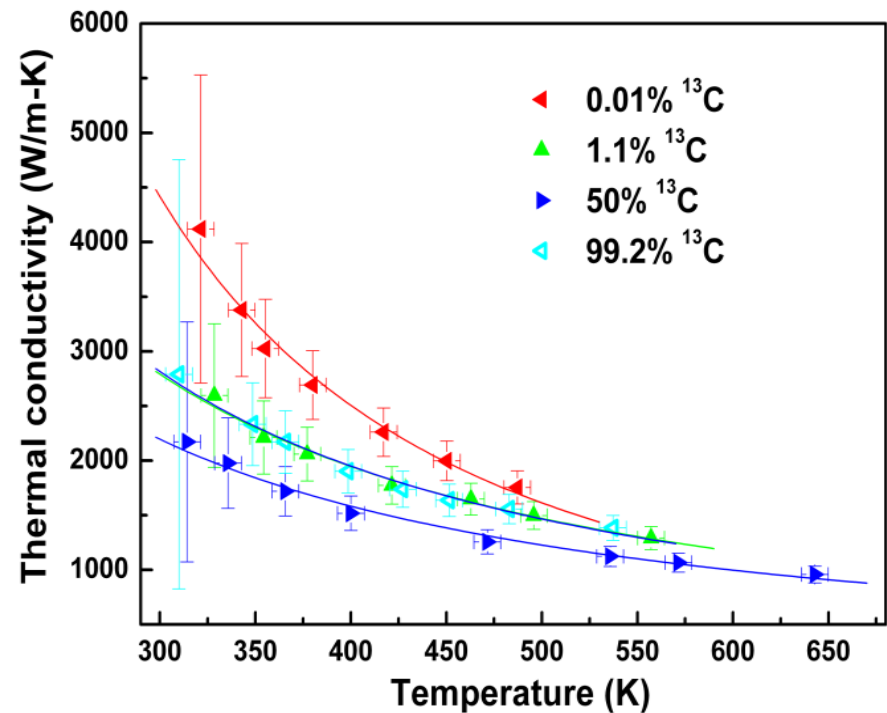
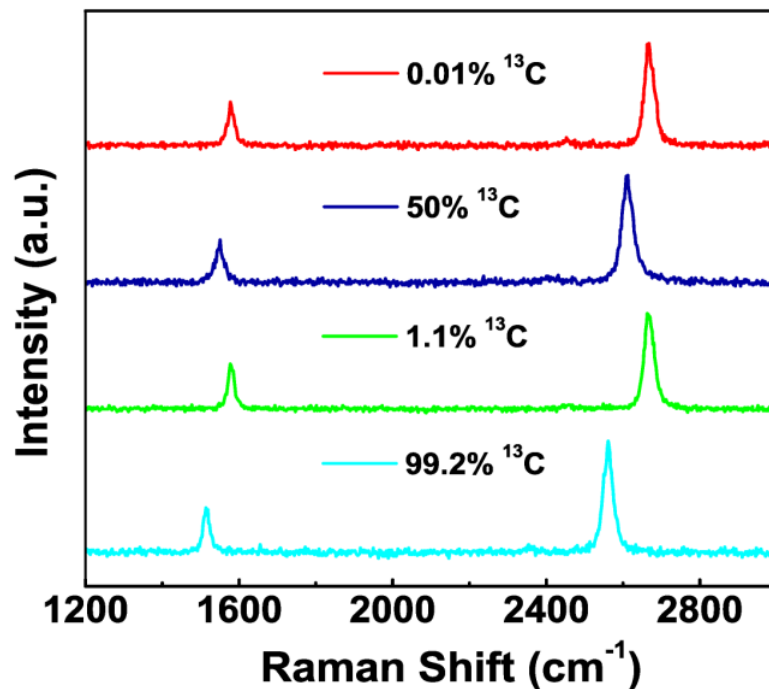


H. Li, H. Ying, X, Chen, D.L. Nika, W. Cai, A.A. Balandin and S. Chen, *Nanoscale*, 6, 13402 (2014)

Alexander A. Balandin -- University of California, Los Angeles

From graphene to other twisted van der Waals materials

Phonon Transport in Isotopically Engineered Graphene

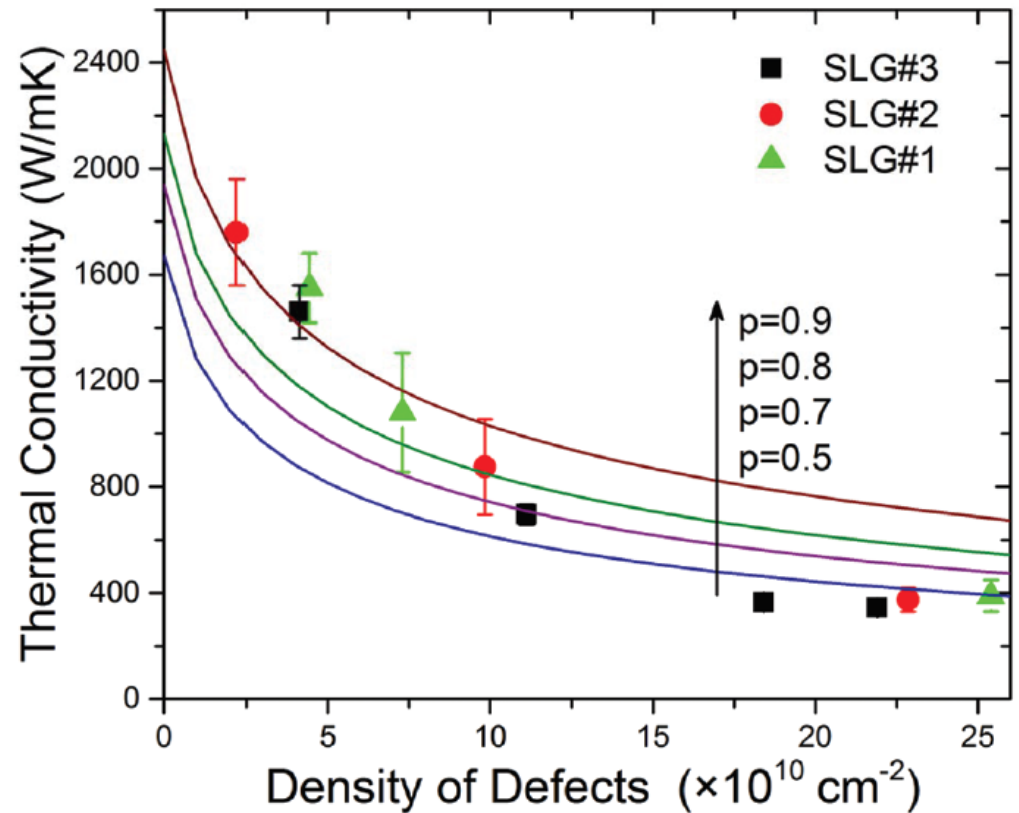
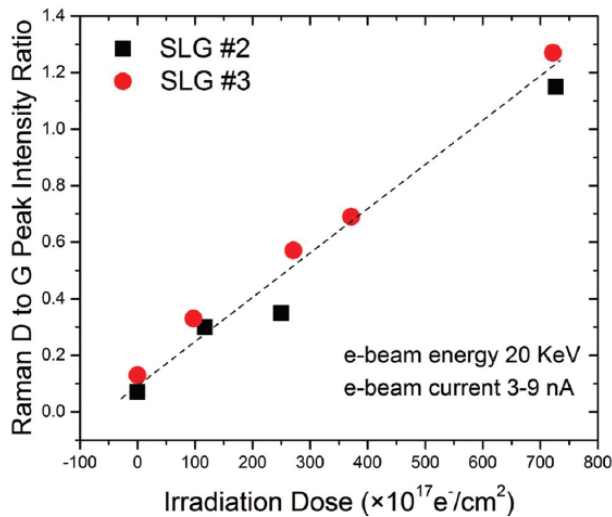
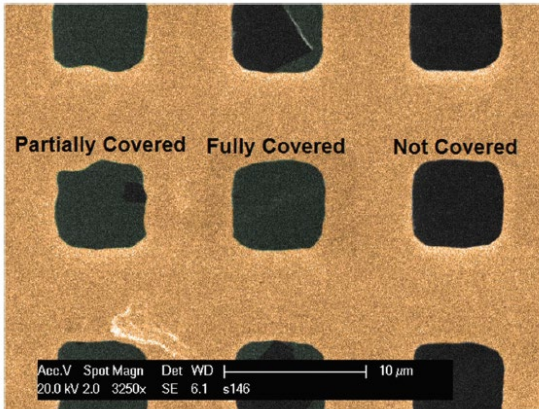


¹³C and ¹²C difference: ~ 64 cm⁻¹

$$\omega \propto M^{-1/2}$$

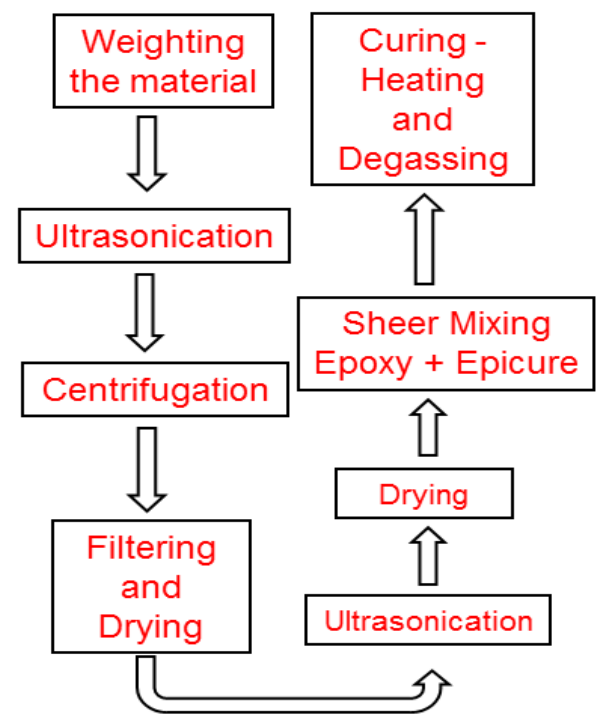
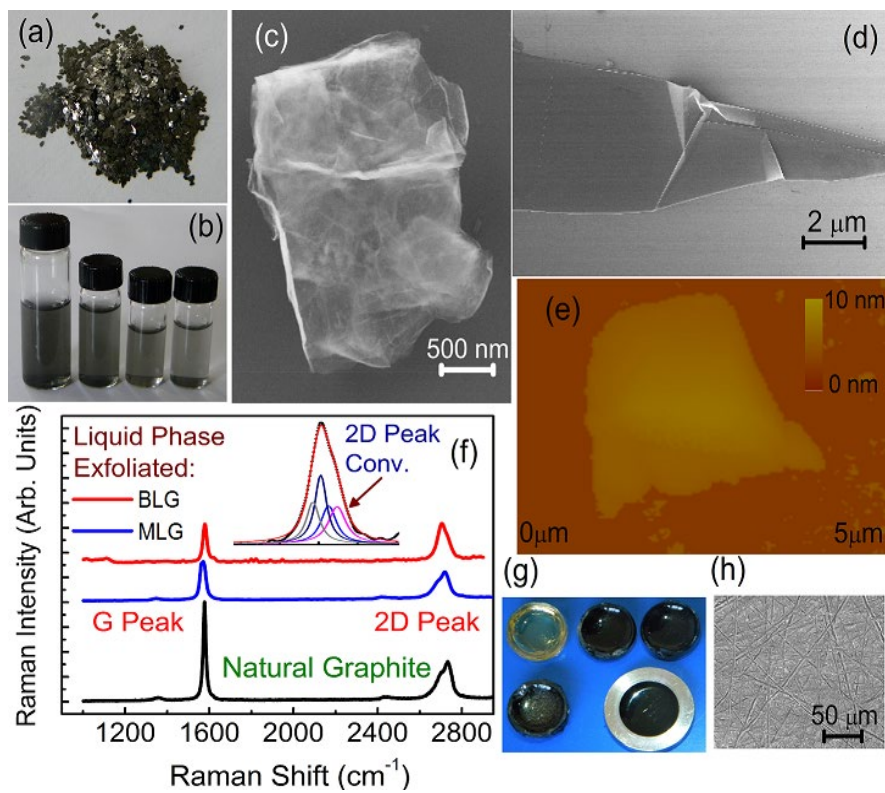
S. Chen, Q. Wu, C. Mishra, J. Kang, H. Zhang, K. Cho, W. Cai, A.A. Balandin and R.S. Ruoff, Nature Materials, 11, 203 (2012).

Effects of the Defects Introduced by Electron Beam Irradiation



H. Malekpour, et al, Nanoscale, 8, 14608 (2016). 37

Real-Life Applications: Graphene Enhanced TIMs

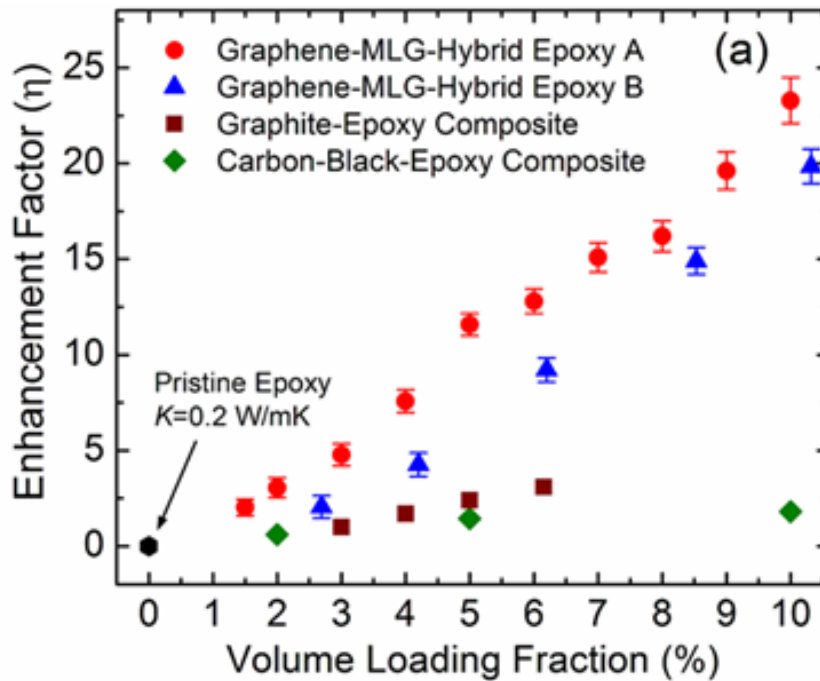


aqueous solution of sodium cholate

K.M.F. Shahil and A.A. Balandin, "Graphene - multilayer graphene nanocomposites as highly efficient thermal interface materials," Nano Letters, 12, 861 (2012).

Graphene Thermal Interface Materials

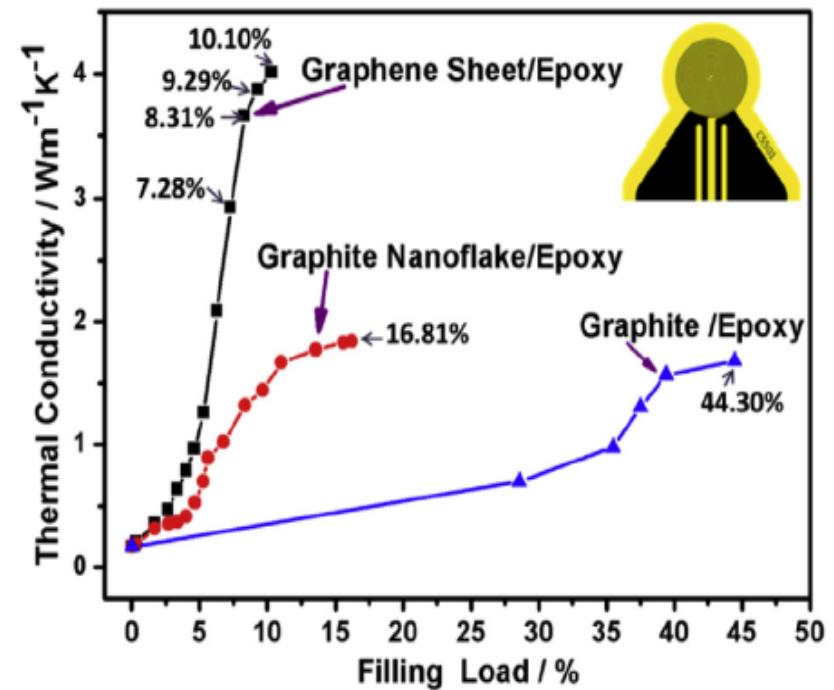
The First Report of Graphene TIMs:



K.M.F. Shahil and A.A. Balandin, Nano Letters, 12, 861 (2012).

Alexander A. Balandin -- University of California, Los Angeles

Independent Experimental Confirmation:

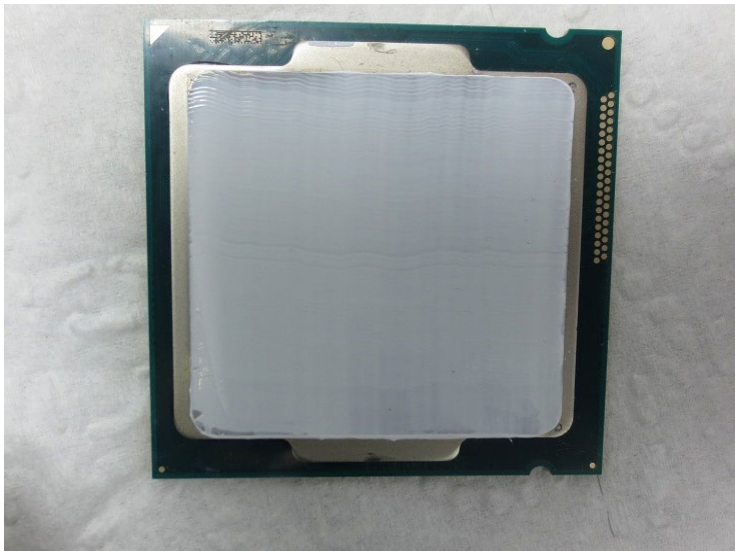


Y.-X. Fu et al., International Journal of Thermal Sciences, 86, 276 (2014).

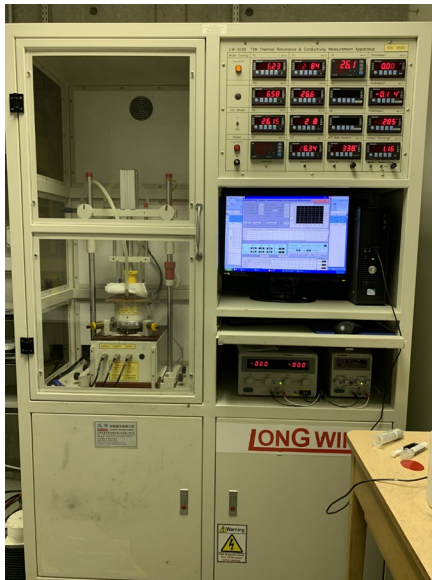
Testing Graphene TIMs in Actual Computers



TIMs with different concentration of graphene (left); graphene TIM applied to CPU (bottom left); CPU attached to the heat sink (bottom)

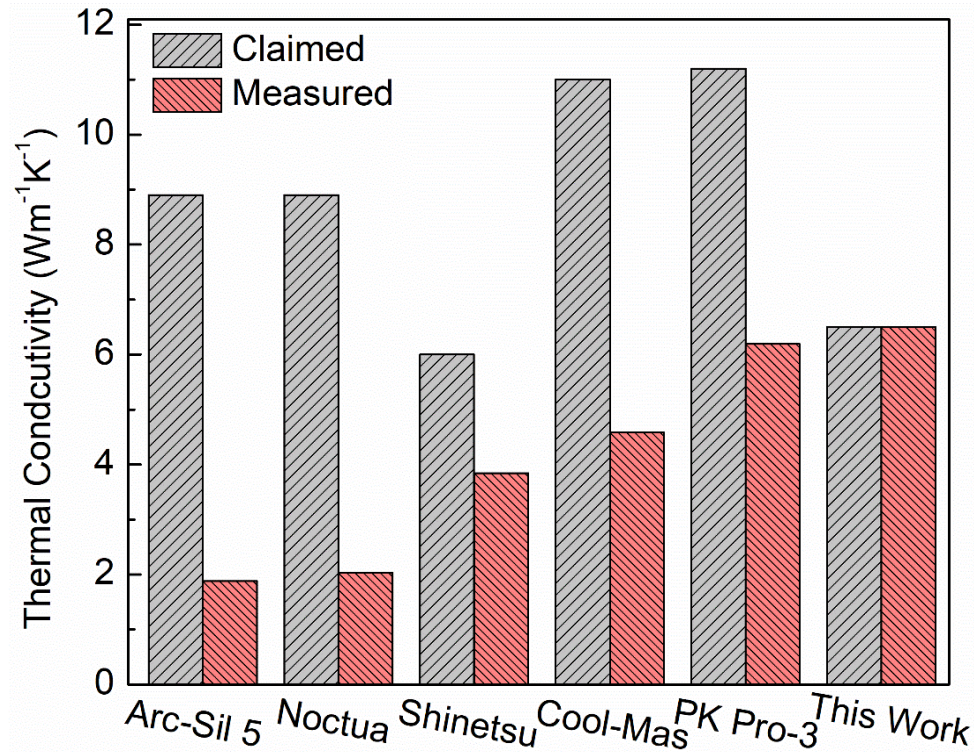


Non-Curing Graphene Composites for Thermal Management of Electronics



ASTM D5470 steady-state vs. transient laser flash approach.

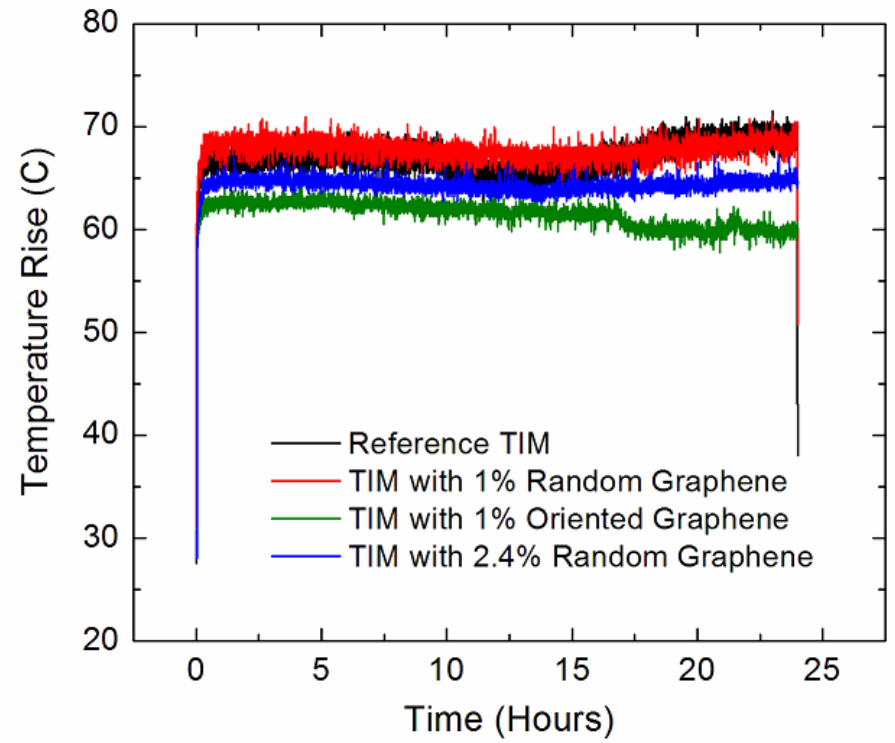
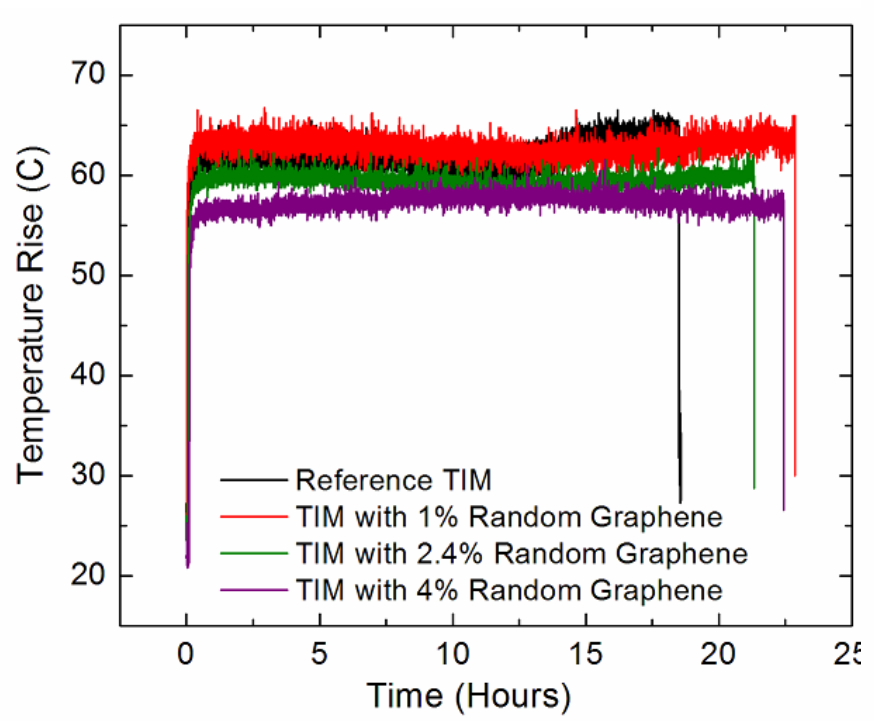
The non-curing graphene TIM has $\phi=19.8$ vol% (40 wt%) filler loading. A commercial non-curing TIM with the highest thermal conductivity (PK Pro-3) uses ~ 90 wt% of Al and ZnO filler loading.



S. Naghibi, et al., Adv. Electron. Mater., 1901303 (2020).

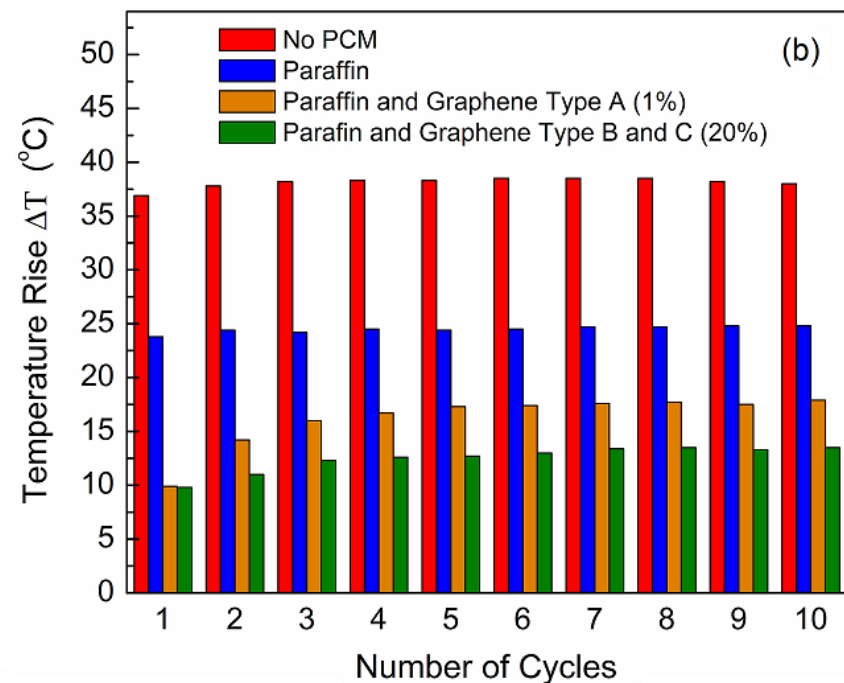
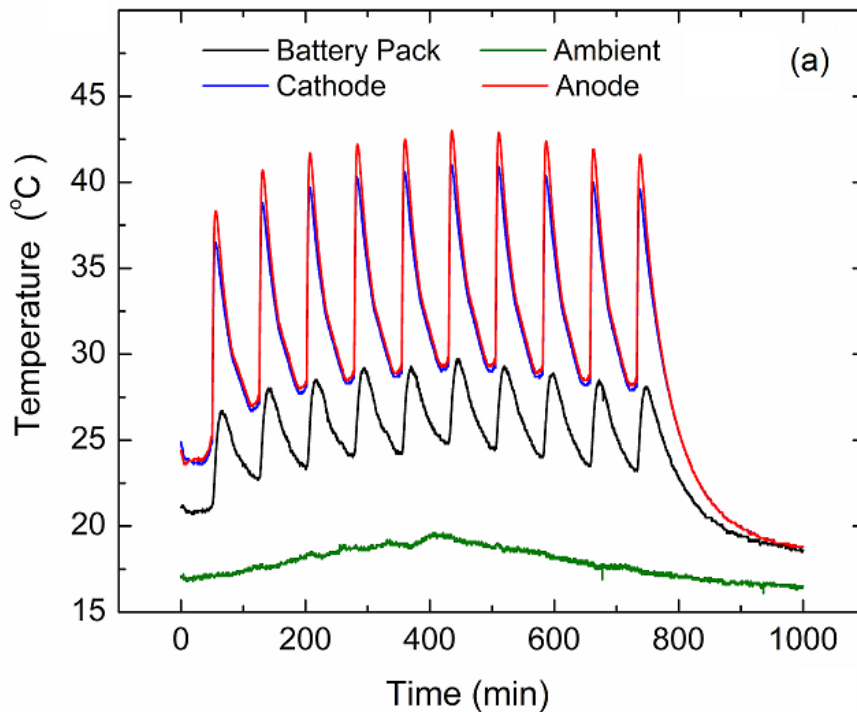
Practical Testing of Graphene Enhanced TIMs in Desktop Computers

Temperature Rise Testing



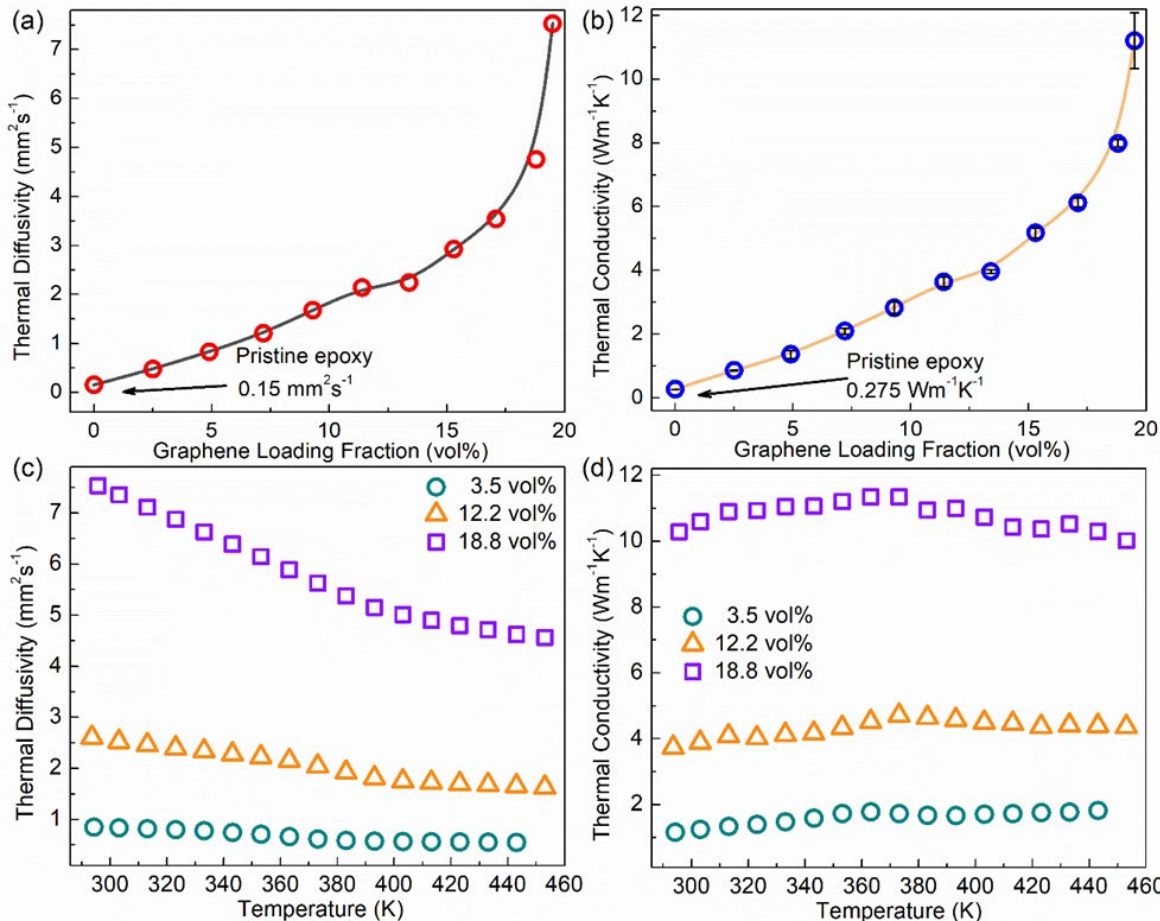
J. Renteria et al., Magnetically-functionalized self-aligning graphene fillers for high-efficiency thermal management applications, *Materials and Design* 88, 214 (2015)

Hybrid Graphene – Paraffin PCM for Li-Ion Batteries



P. Goli, et al., "Graphene-enhanced hybrid phase change materials for thermal management of Li-ion batteries," J. Power Sources, 248, 37 (2014).

Fundamental Science Questions in Graphene Composites – Thermal Percolation Threshold

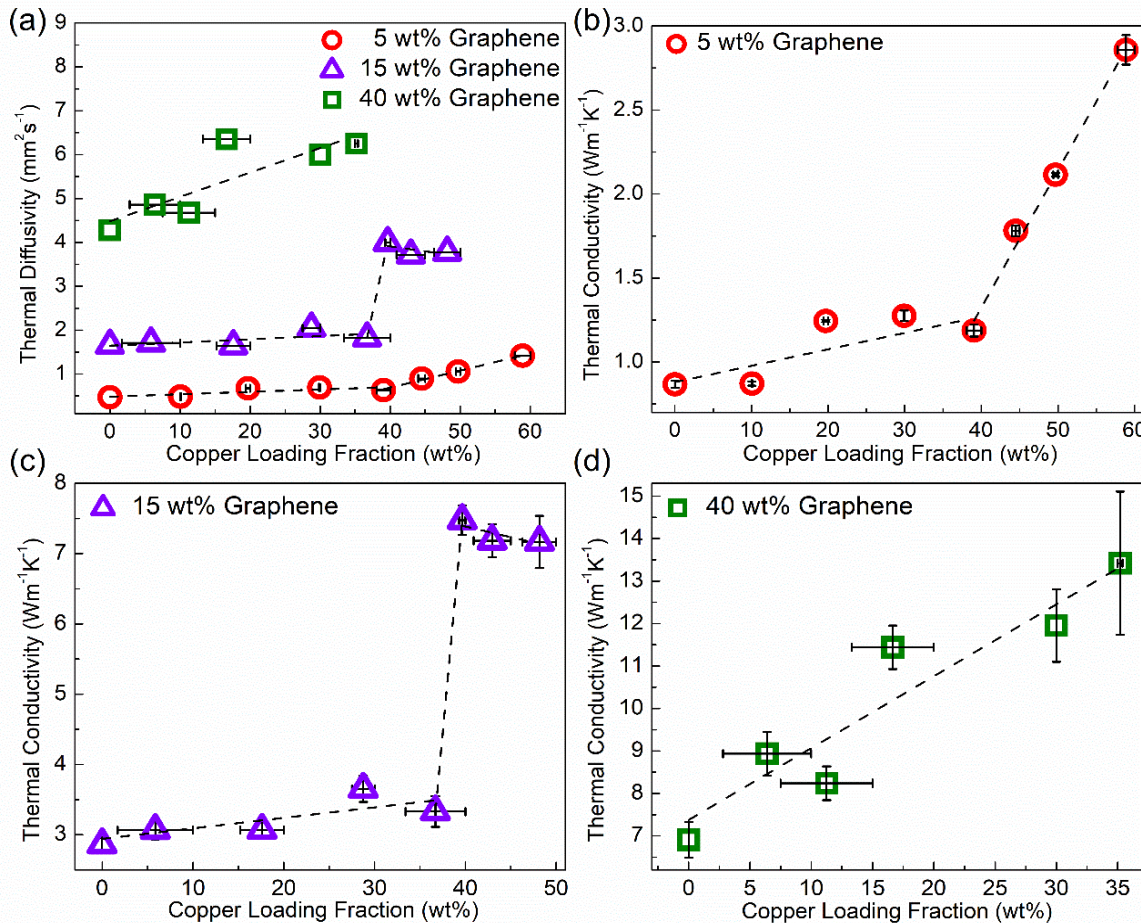


→ Thermal diffusivity and thermal conductivity of the composites as a function of graphene loading fraction at RT

→ Thermal diffusivity and thermal conductivity of three different composites with the low, medium, and high loading of graphene fillers as a function of temperature.

Z. Barani, et al., Adv. Electron. Mater., 2000520 (2020).

Binary Fillers – Effects on Thermal Percolation

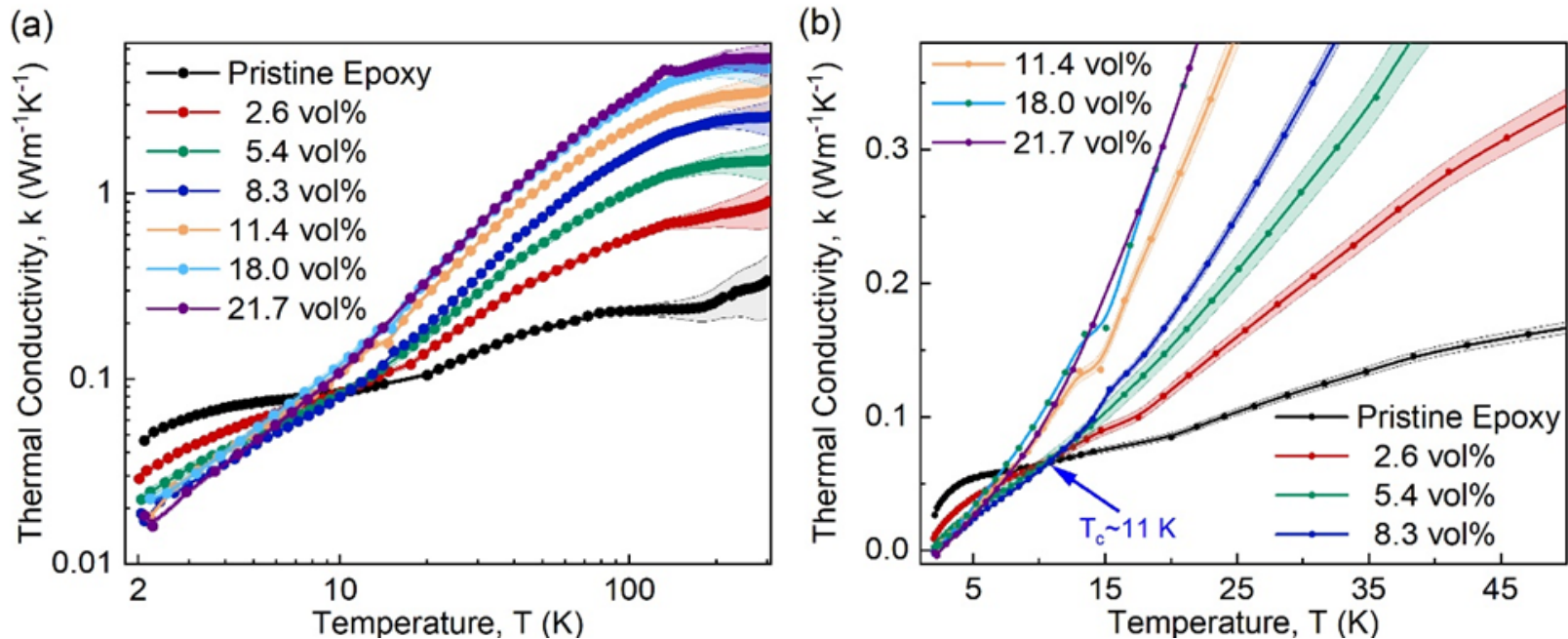


- Fillers with different type of dominant heat carriers
- The “synergistic” effect for size-dissimilar fillers has a well-defined loading threshold
- Intercalation of spherical copper nanoparticles between the large graphene flakes, resulting in formation of the percolation network

Z. Barani, et al., Adv. Functional Mater., 1904008 (2019). 45

Cryogenic Characteristics of Graphene Composites

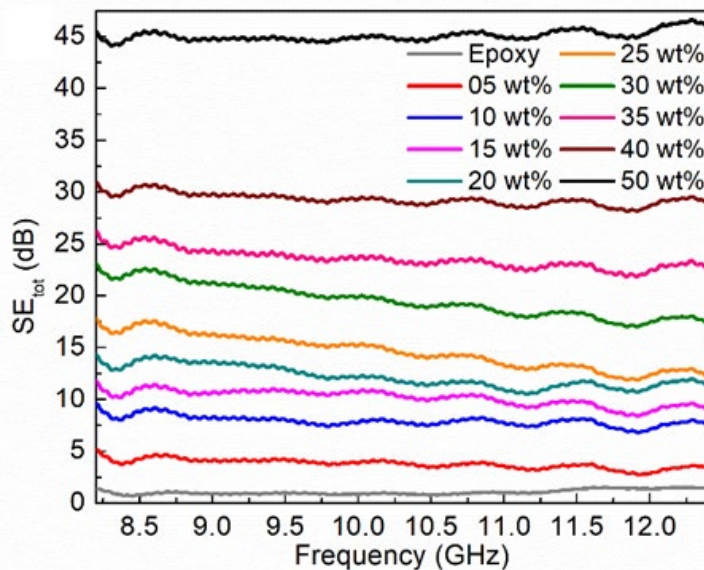
Evolution from Thermal Conductors to Thermal Insulators



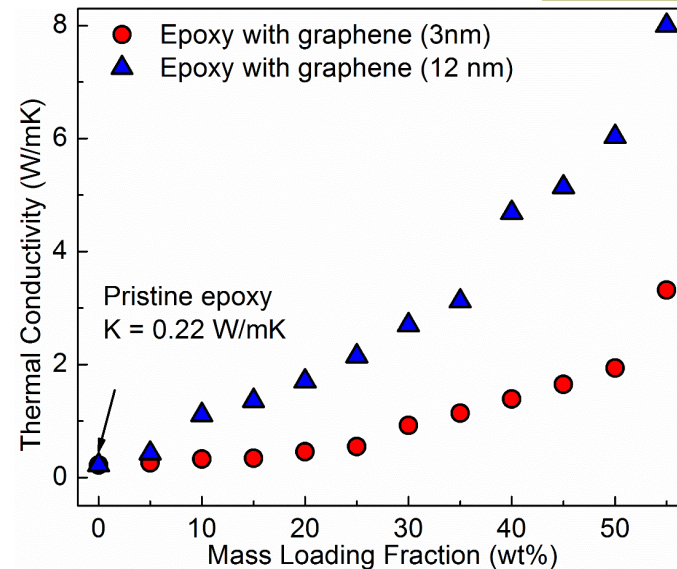
- Thermal conductivity of graphene composites in the temperature range of $2 \text{ K} \leq T \leq 300 \text{ K}$
- The cross-over temperature, T_c

Z. Ebrahim Nataj, Y. Xu, D. Wright, J. O. Brown, J. Garg, X. Chen, F. Kargar, and A. A. Balandin, *Nature Com.*, 14, 3190 (2023).

Dual Functionality of Graphene Composites – EMI Shielding and Thermal Management



Efficient total EMI shielding, $SE_{tot} \sim 45$ dB, in the X-band frequency range, $f = 8.2$ GHz – 12.4 GHz, while simultaneously providing the high thermal conductivity, $K \approx 8 \text{ Wm}^{-1}\text{K}^{-1}$, which is a factor of $\times 35$ larger than that of the base matrix material.



- The composite works even below the electrical percolation threshold – local coupling of EM wave to the filler
- Electrically insulating films can be efficient EMI shield and dissipate the heat

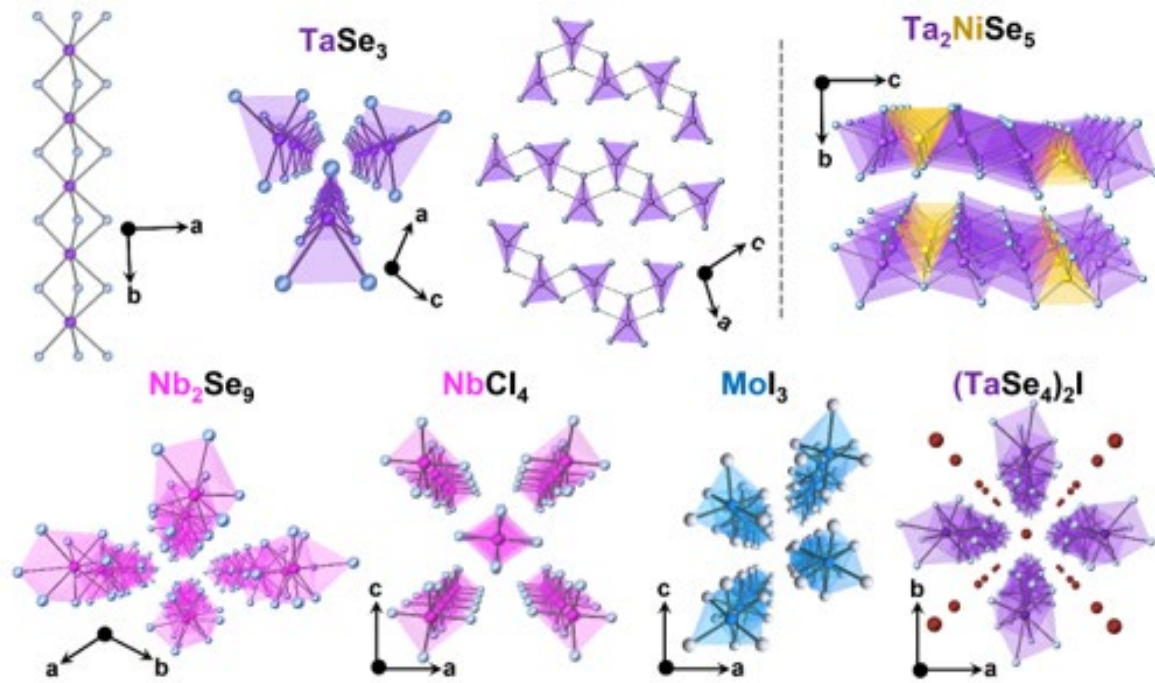
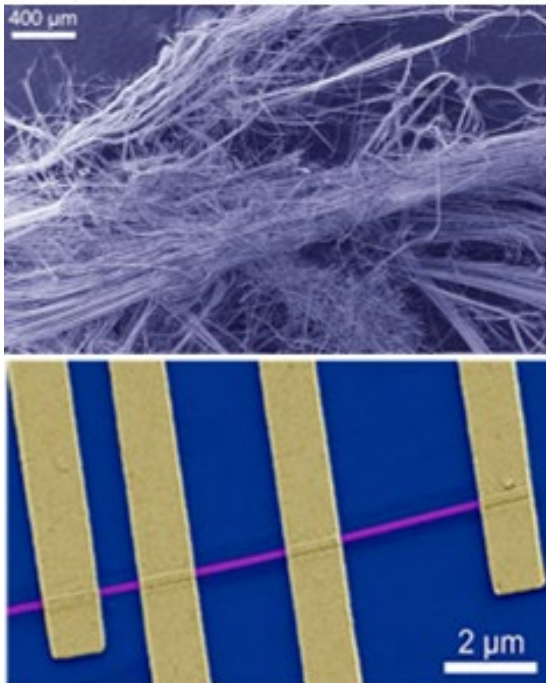
F. Kargar, et al., Adv. Electron. Mater., 5, 1800558 (2019).

Outlook for Graphene Thermal Field

- Thermal conductivity of graphene reveals unusual size-dependent phenomenon – different from ballistic transport
- Few-layer graphene is an excellent system to study phonons in low-dimensions
- Theory development continues – four-phonon processes; phonon hydrodynamic transport
- Few-layer graphene can be used as fillers in thermal composites
- Optimization of graphene composites, length scales, aspect ratios, orientation, composition, can bring revolutionary changes to TIMs
- Thermal applications of graphene and FLG are already the reality

Where to Go Next to Study Phonons ?

One-Dimensional van der Waals Materials



A. A. Balandin, et al., "One-dimensional van der Waals quantum materials", Mater. Today, 55, 74 (2022).

Acknowledgements



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Dr. Ruben Salgado
Dr. Zhong Yan
Dr. Ece Aytan
Dr. Hoda Malekpour

My research group emerging from the lab, after the end of the COVID regulations, to enjoy the Californian sun in UC Riverside. We are getting ready to move 70 miles north to UCLA and continue to rock-and-roll.