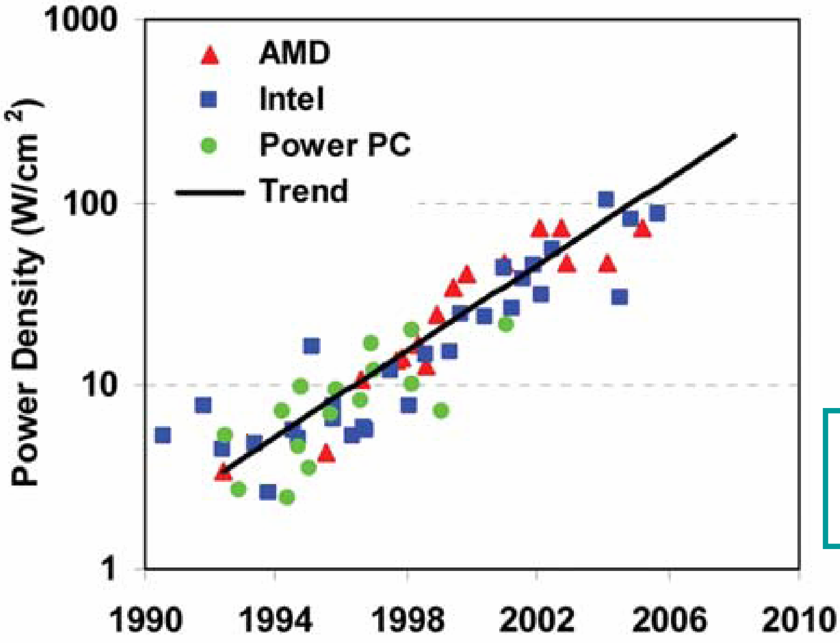
Outline

- ❖ Motivation
- ❖ Method for nanoscale thermal transport: NEGF
- ❖ Applications
  - * Nanotube phonon waveguide
  - * Graphene nanoribbons: anisotropy
  - * Graphene-based nanostructures
- ❖ Summary

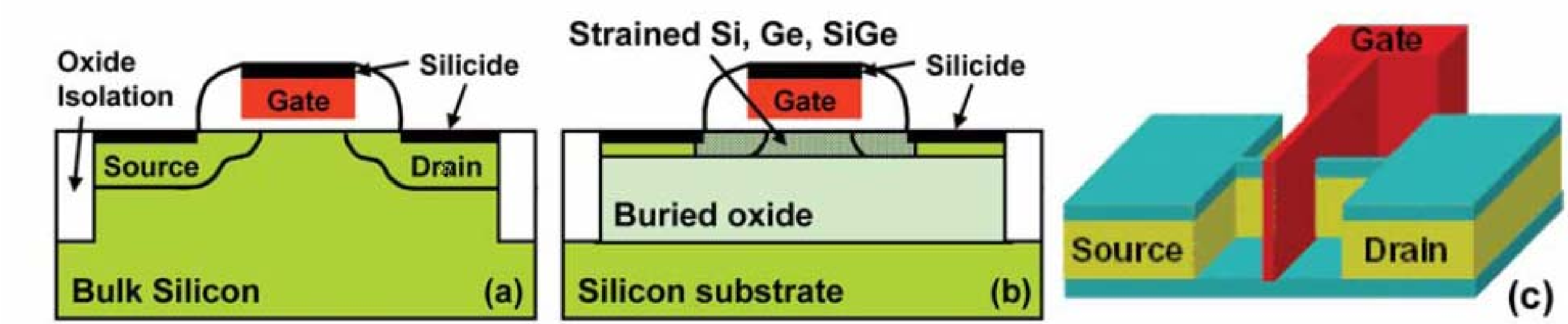
Serious heat dissipation



Material	Thermal Conductivity ($Wm^{-1}K^{-1}$)
Si (bulk)	148
Ge (bulk)	60
Silicides	40
Si (10 nm)	13
$Si_{0.7}Ge_{0.3}$	8
SiO_2	1.4

- ◆Power density increases ***exponentially***.
- ◆Thermal conductivity reduces remarkably.

Thermal Design!

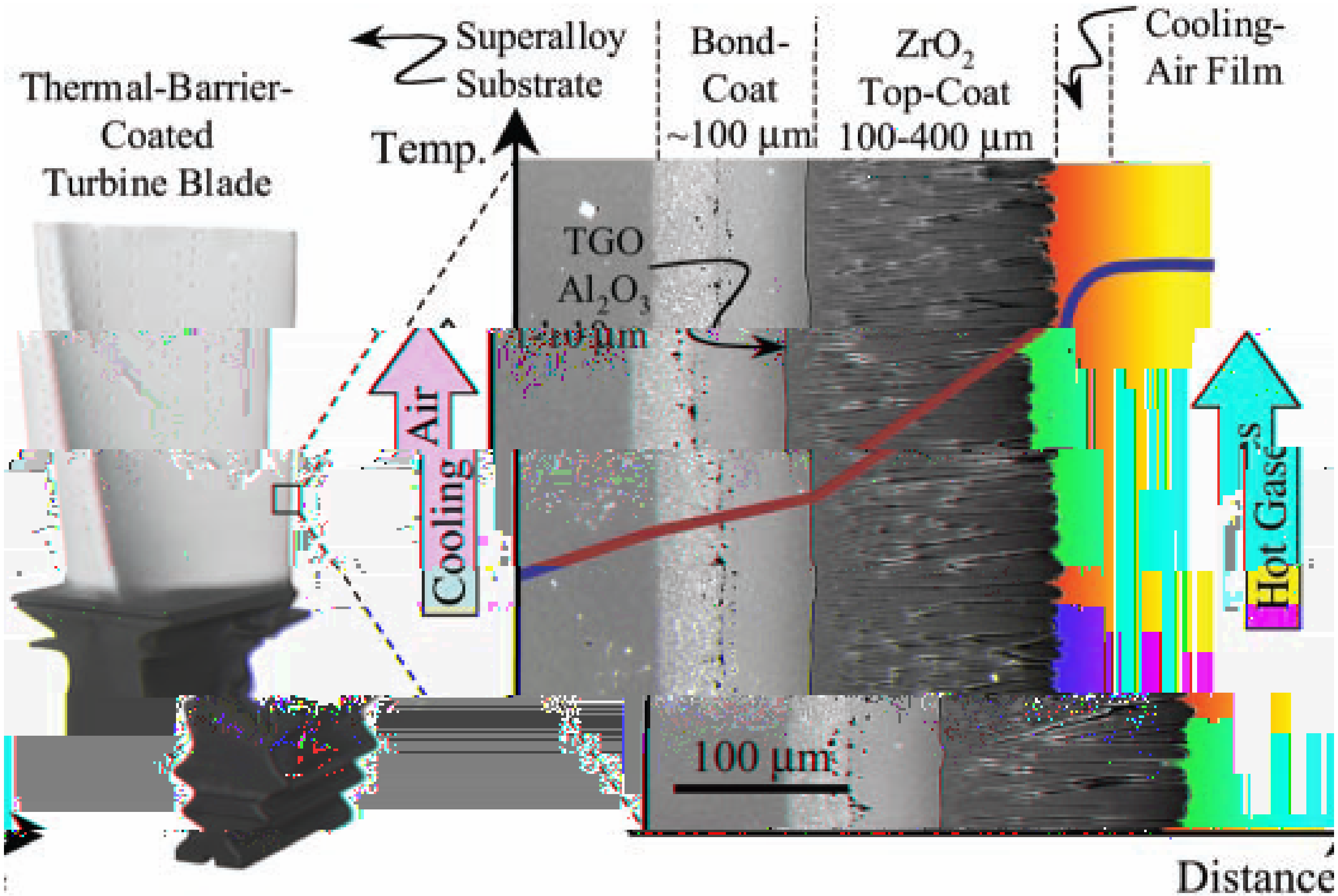


Evolution of transistor designs

E. Pop, *et al*. Proceedings of the IEEE **94,** 1587 (2006).

Thermal insulation

Thermal Barrier Coatings for Gas-Turbine Engine Applications

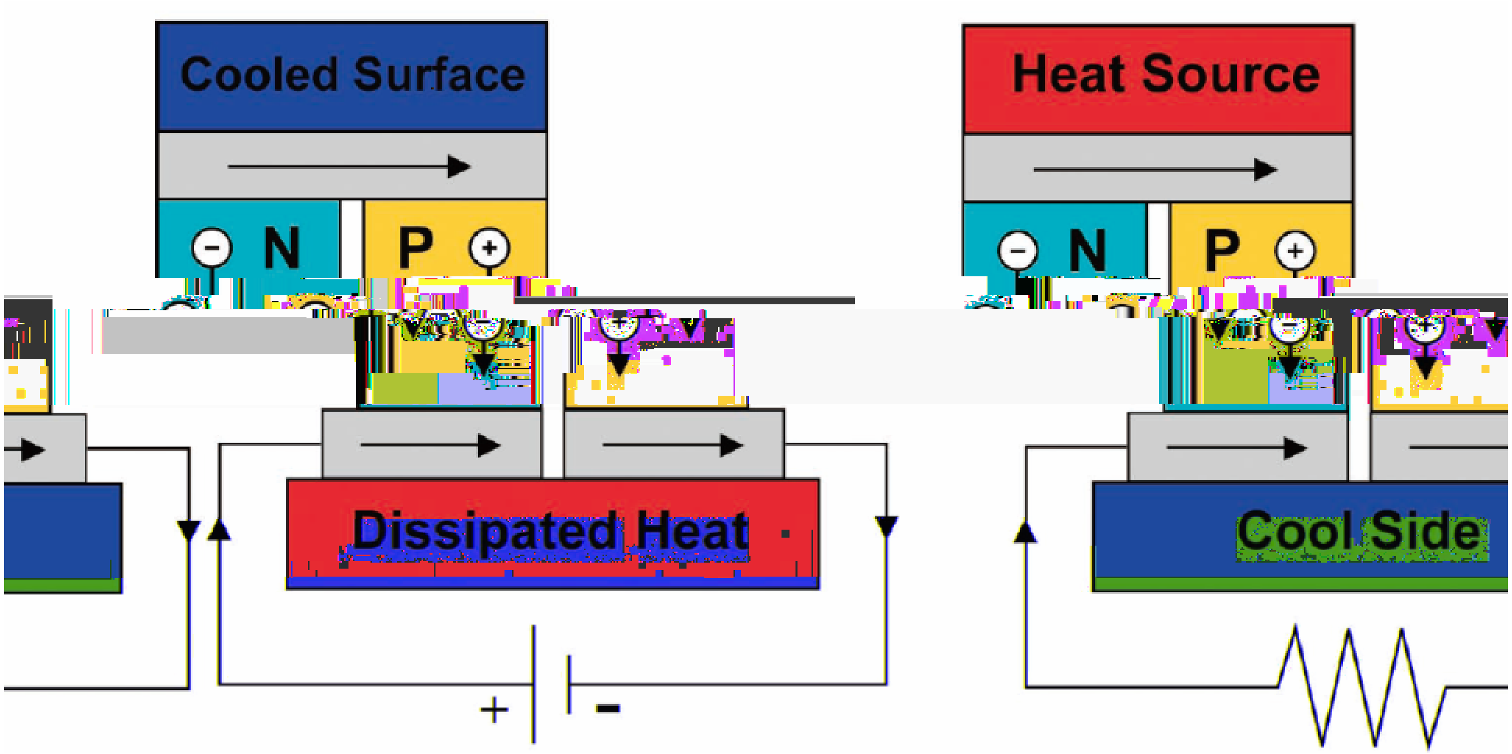


- ◆**Low thermal conductivity**
- ◆**Stable at high *T*.**

N. P. Padture, *et al.*, Science **296,** 280 (2002).

Thermoelectric applications

90% (10 TW) of effective energy is generated by heat engines, accompanying with 15 TW wasted heat.



Refrigerators and power generators

Thermal devices

Diodes, transistors	⇨	Modern electronics
Thermal devices	⇨	Future phononics

Thermal diodes, thermal transistor

Applications:

refrigerator, heat dissipation in microelectronic processor, energy saving building, thermal logical gates ……

Method: nanoscale thermal transport

- ***Traditional methods*** fail at nanoscale.

Molecular dynamics:

classical, breakdown at low T, definition of T?

Boltzmann-Peierls equation:

concept of distribution function?

- ***New methods*** are urgently needed.

Landauer formula:

simple physical picture for mesoscopic transport

Nonequilibrium Green's function (NEGF):

fully based on quantum mechanics, general.

First-principles based NEGF method

❖ Features:

- Atomic scale simulation
- Quantum effects
- No empirical parameters
- For realistic materials, nanodevices

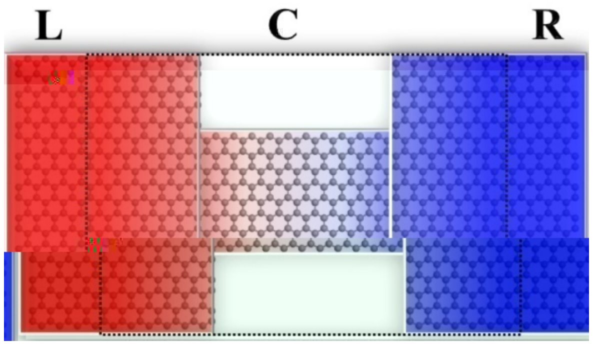
❖ Code Programming:

- FORTRAN language
- Interface with widely used softwares (VASP, SIESTA etc.)
- Semi-empirical model for faster calculations

First-principles based NEGF method

Total Hamiltonian:

$$H = \sum_{\alpha=L,C,R} H_\alpha + V + H_n$$



Harmonic Hamiltonian:

$$H_\alpha = \frac{1}{2}(\dot{u}_\alpha)^T \dot{u}_\alpha + \frac{1}{2}u_\alpha^T D_{\alpha\alpha} u_\alpha \quad (\alpha = L, C, R)$$

$$V = (u_L)^T D_{LC} u_C + (u_C)^T D_{CR} u_R$$

$$u_\alpha = (u_1^\alpha, u_2^\alpha, u_3^\alpha, \cdots)^T, \quad u_i^\alpha = \sqrt{M_i^\alpha}\, x_i^\alpha$$

Force Constants:

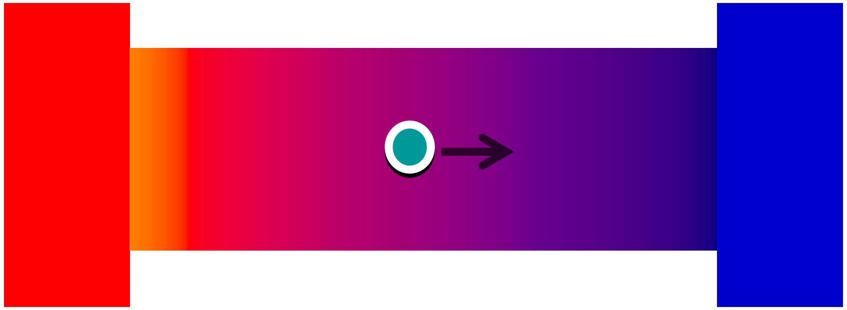
$$[D_{\alpha\beta}]_{ij} = D_{ij}^{\alpha\beta} = \frac{\partial^2 E}{\partial u_i^\alpha \partial u_j^\beta}\Big|_0$$

$$D_{i_1,i_2,\cdots,i_n} = \frac{\partial^n E}{\partial u_{i_1}\partial u_{i_2}\cdots\partial u_{i_n}}\Big|_0$$

Anharmonic Hamiltonian (center part):

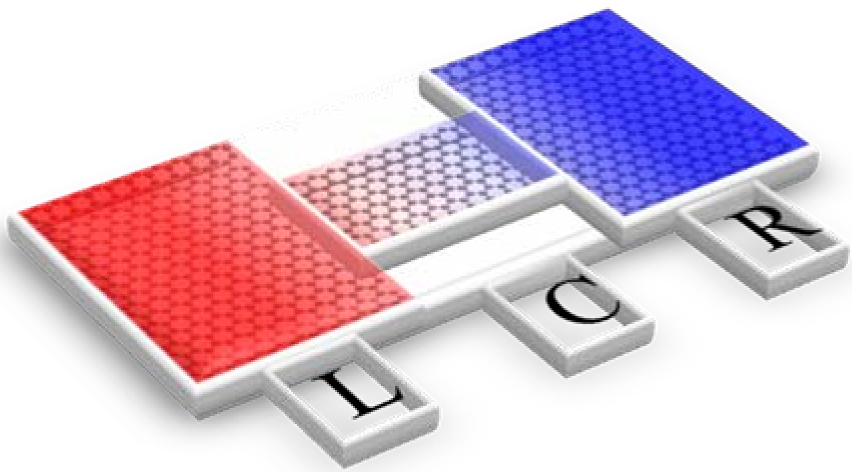
$$H_n = \sum_{n=3,4,\cdots} H^{(n)} = \sum_{n=3,4,\cdots} \frac{1}{n!} \sum_{i_1,i_2,\cdots,i_n} D_{i_1,i_2,\cdots,i_n} u_{i_1} u_{i_2} \cdots u_{i_n}$$

First-principles based NEGF method



Non-equilibrium Green's function (NEGF) Method

Thermal transport in nanodevices ⟸ First-principles based NEGF approach



Green's function:

$$G^{\mathrm{r}}_{\mathrm{CC}} = [(\omega + i\delta)^2 - D_{\mathrm{CC}} - \Sigma^{\mathrm{r}}_{\mathrm{L}} - \Sigma^{\mathrm{r}}_{\mathrm{R}}]^{-1}$$

Force constants (FCs):

$$D_{i\alpha,j\beta} = \frac{1}{\sqrt{M_i M_j}} \frac{\partial^2 E}{\partial u_{i\alpha} \partial u_{j\beta}} \Big|_0 = \frac{\Phi_{i\alpha,j\beta}}{\sqrt{M_i M_j}}$$

Phonon transmission function:

$$\Xi(\omega) = Tr[\Gamma_{\mathrm{L}}(\omega) G^{\mathrm{r}}(\omega) \Gamma_{\mathrm{R}}(\omega) G^{\mathrm{a}}(\omega)]$$

$$\Gamma_{\mathrm{L}}(\omega) = i[\Sigma^{\mathrm{r}}_{\mathrm{L}}(\omega) - \Sigma^{\mathrm{a}}_{\mathrm{L}}(\omega)] \qquad \Gamma_{\mathrm{R}}(\omega) = i[\Sigma^{\mathrm{r}}_{\mathrm{R}}(\omega) - \Sigma^{\mathrm{a}}_{\mathrm{R}}(\omega)]$$

To calculate FCs:

1. *Ab inito* methods
2. Empirical potentials
   (Brenner potential for carbon materials, …)

Thermal conductance (Landauer formula):

$$\sigma(T) = \frac{h^2}{2\pi k_B T^2} \int_0^\infty d\omega \frac{\omega^2 e^{h\omega/(k_B T)}}{(e^{h\omega/(k_B T)} - 1)^2} \Xi(\omega)$$

NEGF Method

Calculate nonlinear self energy:

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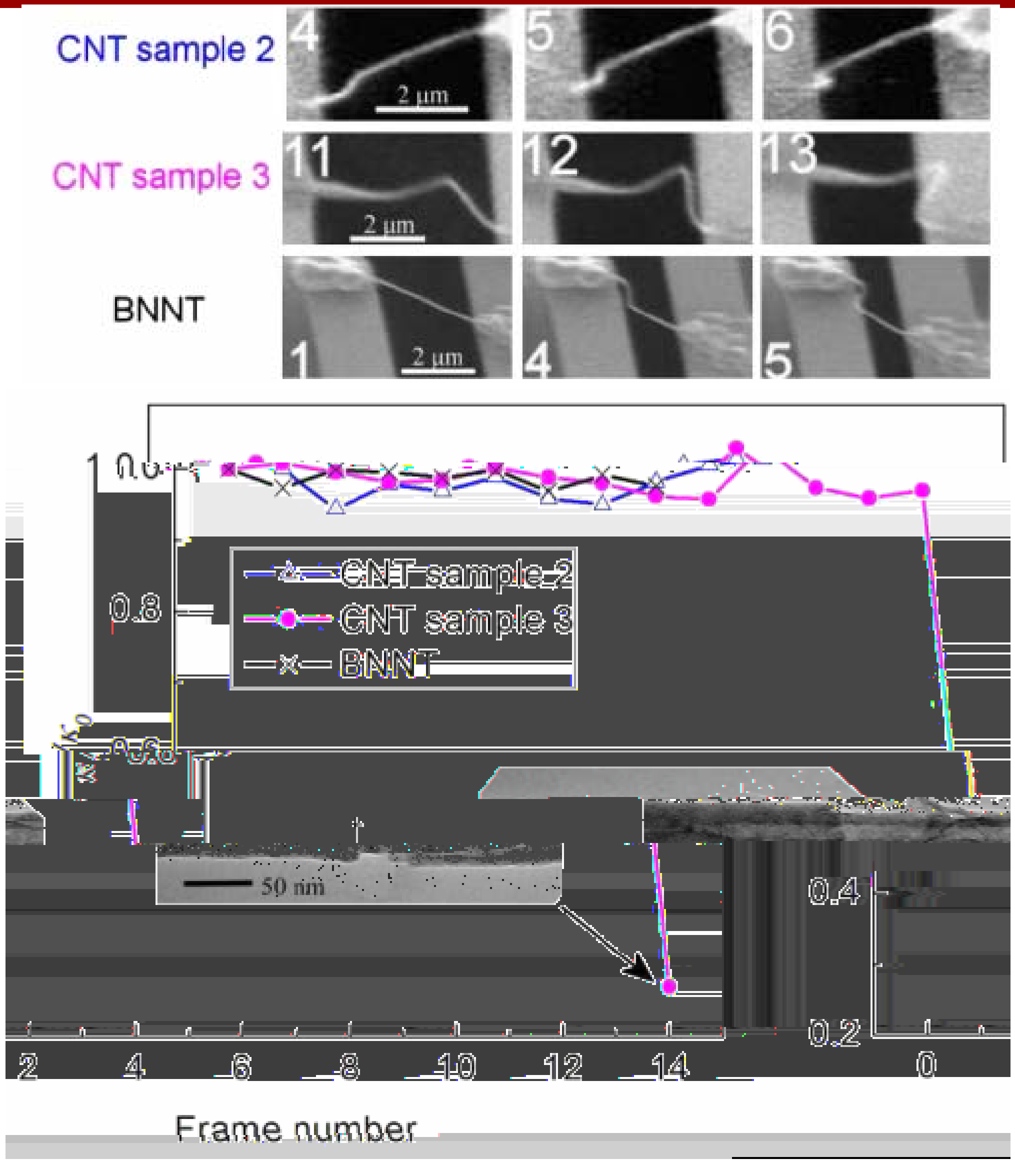
Nanotube phonon waveguide



SEM images

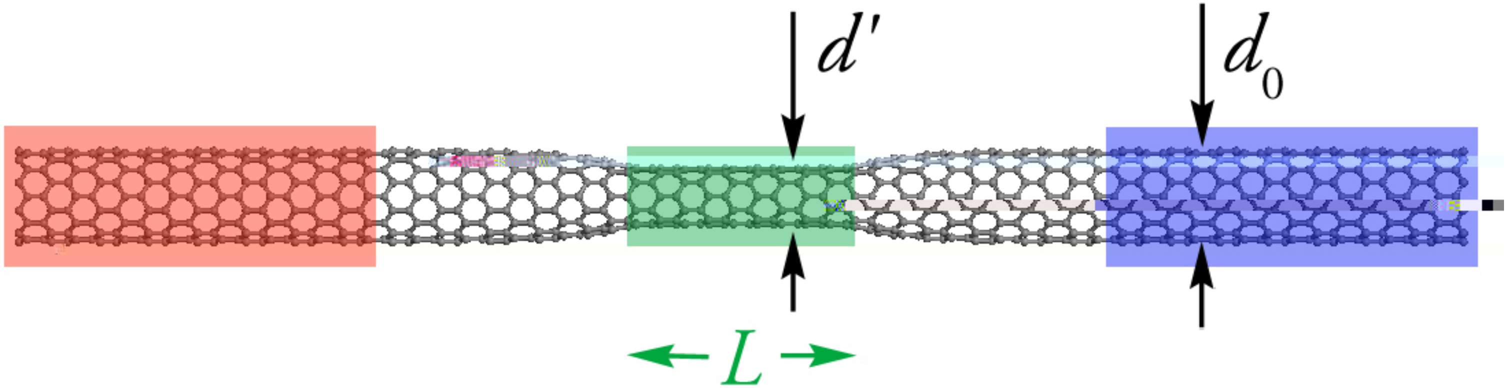
Thermal conductivity of nanotubes is insensitive to structural deformation.

Phonon waveguide!

C. W. Chang et al., Phys. Rev. Lett. 99, 045901 (2007).

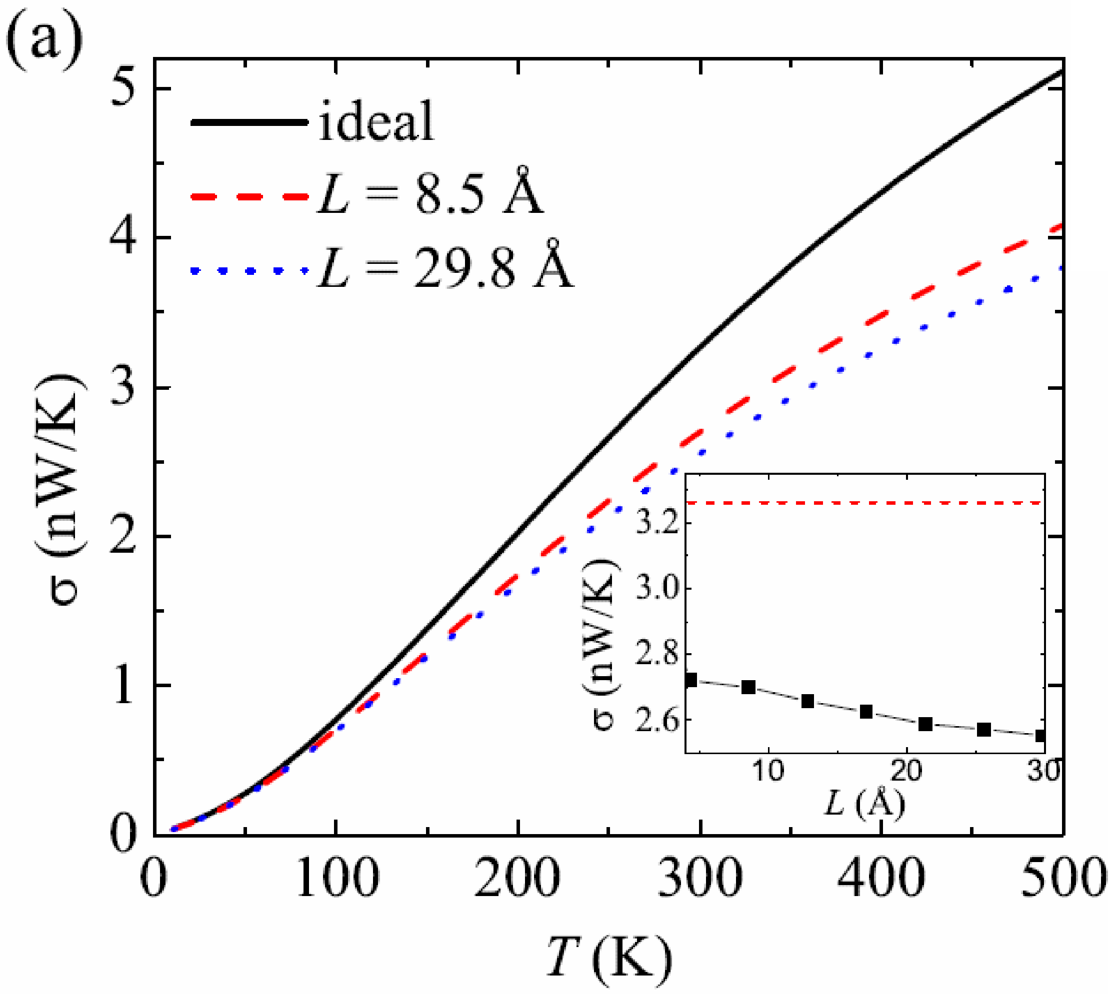
Transport system

What is the underlying physics for nanotube phonon waveguide?



Radial strain: $\eta = d'/d_0$

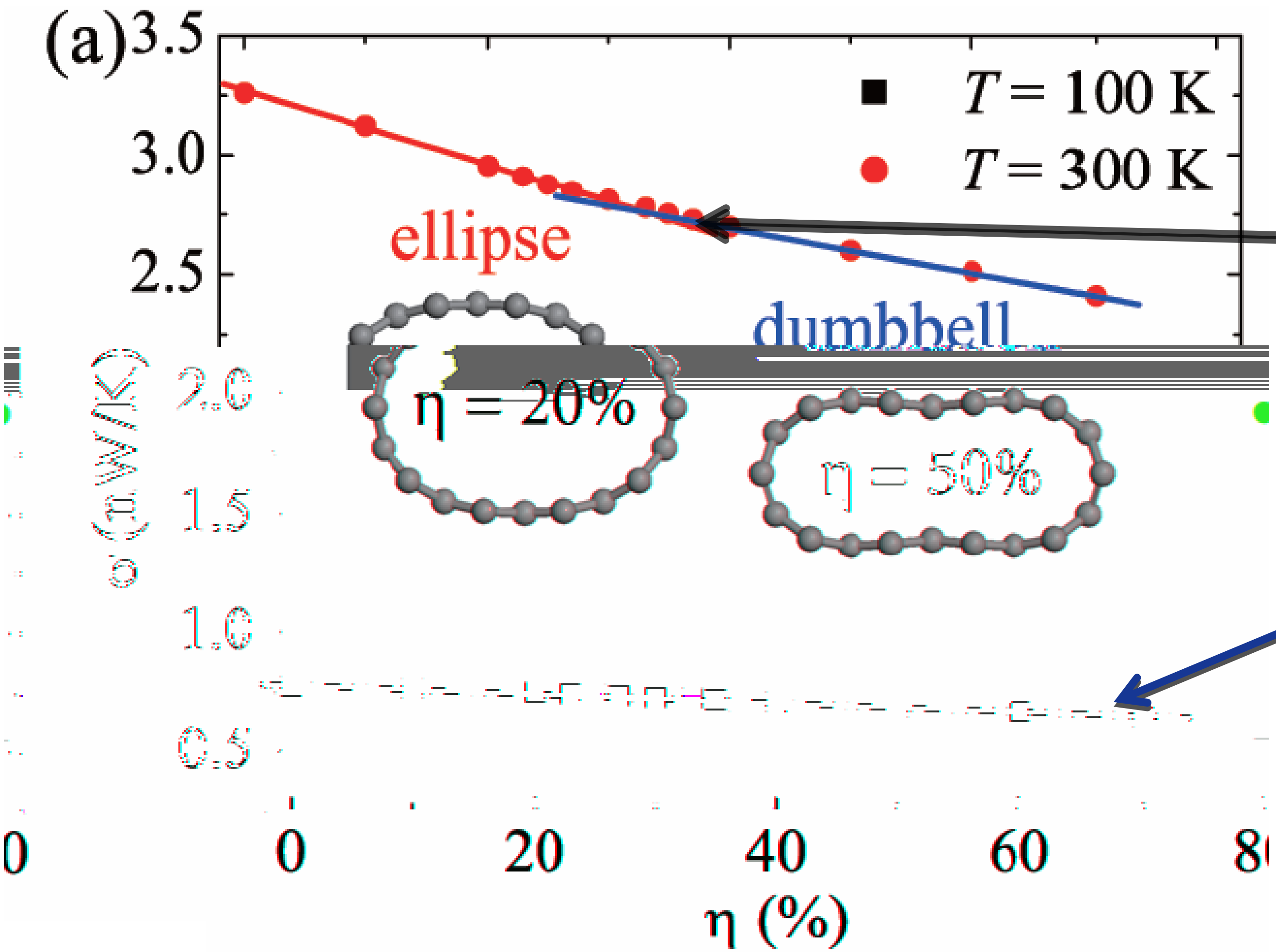
Phonon scattering occurs at interface



Thermal conductance is insensitive to the length of the confined region.

(10,0) CNT η =40%

Radial strain-dependent thermal conductance

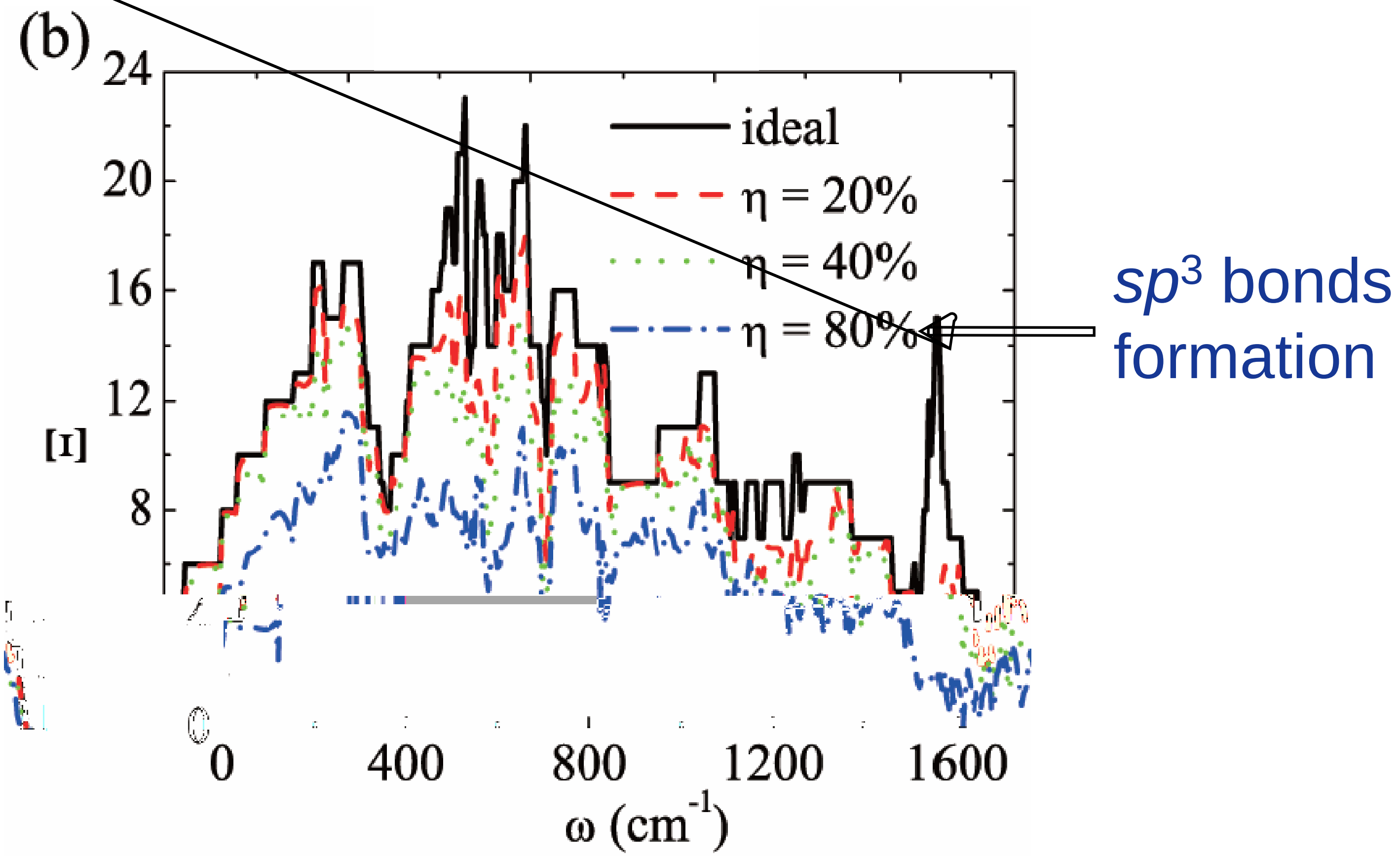


Structural transition

Linear in the whole elastic regime!

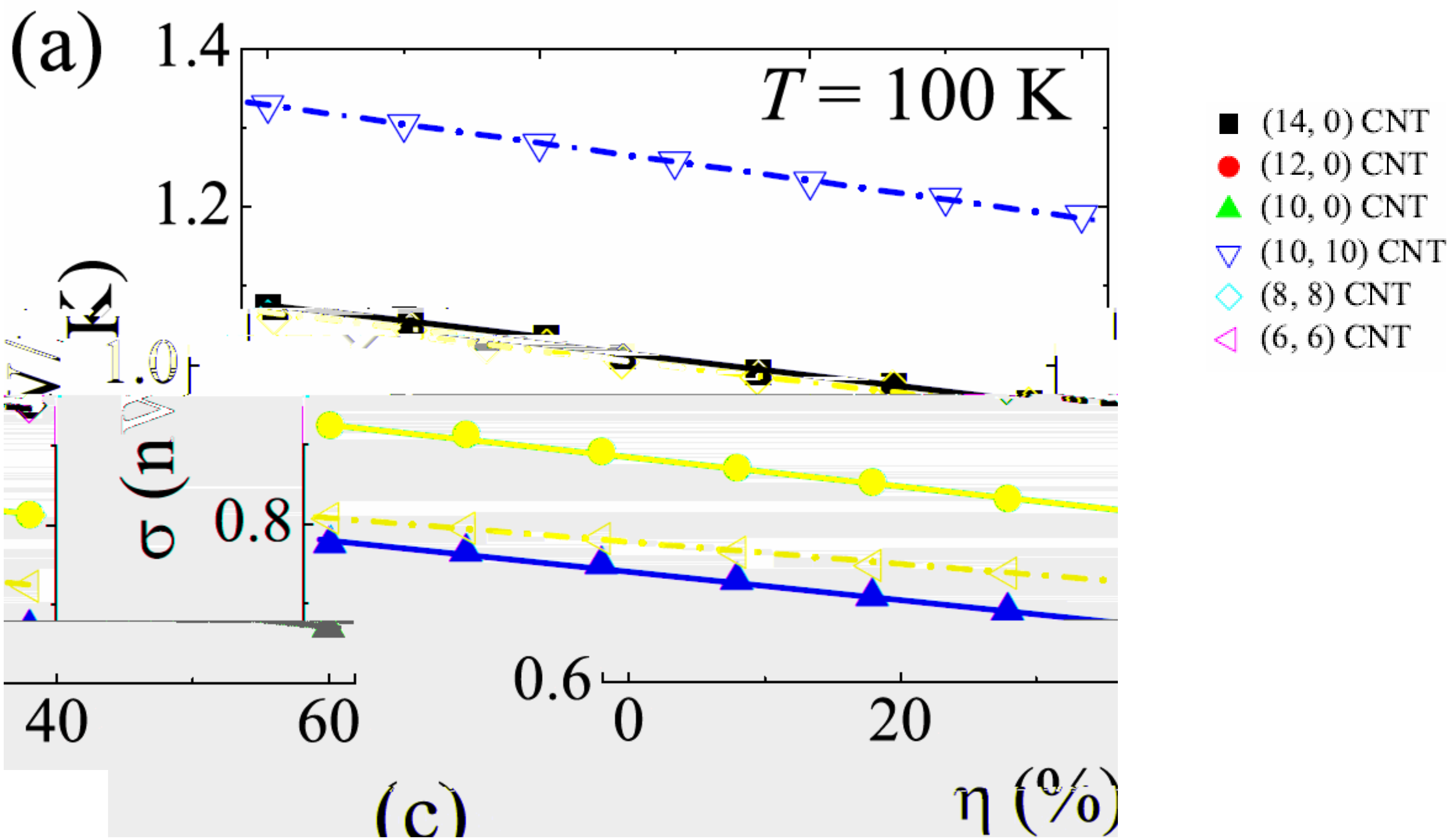
Nontrivial linear response, very robust, why?

Radial strain-dependent phonon transmission



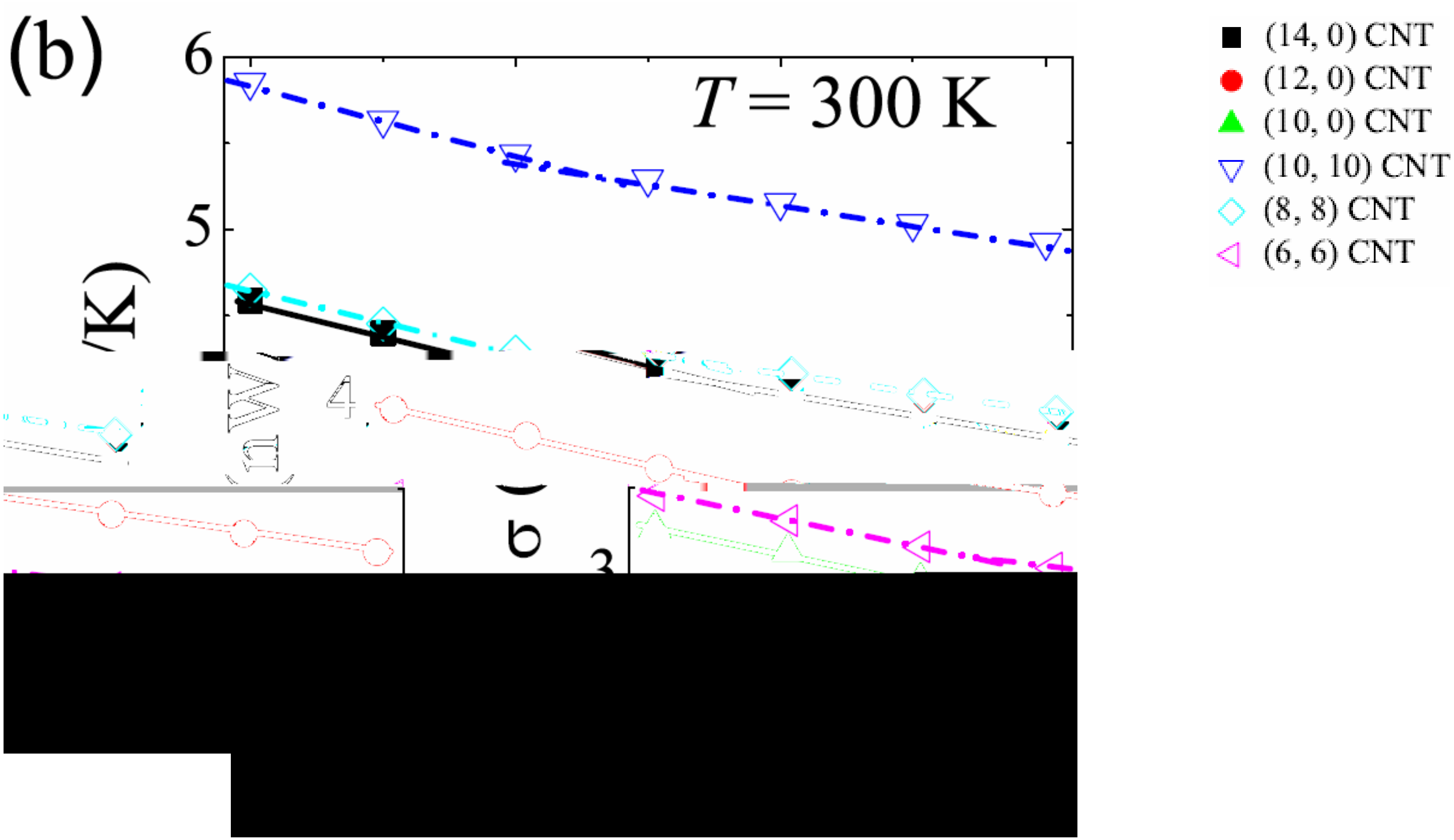
- Quasi-ballistic transport at low frequencies.
- Large strain (within elastic regime) can be viewed as a perturbation to the transport low frequency modes.

Effects of tube diameter and chirality



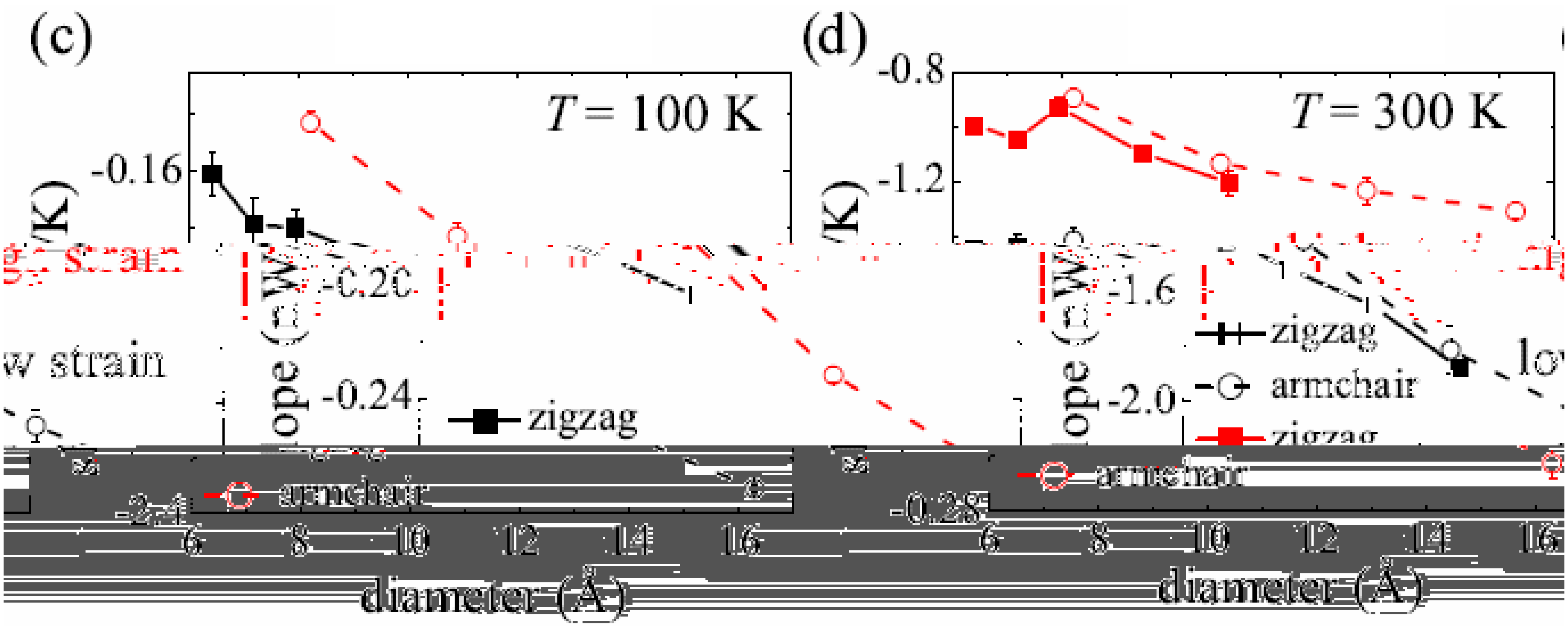
The robust linear response behavior is general in CNTs.

Effects of tube diameter and chirality



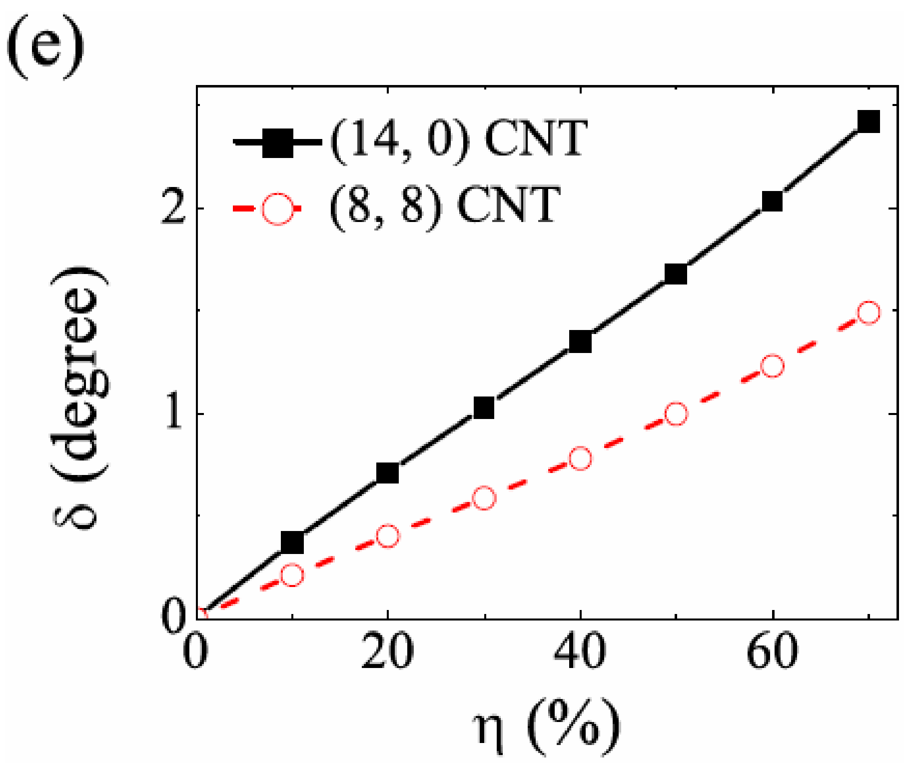
Different linear dependence at low and high strains.

Effects of tube diameter and chirality



Larger CNTs have more phonon modes; and thus the same strain causes larger thermal conductance decrease.

Radial strain induced Bond angle change



- The root mean square deviation δ is larger in zigzag CNTs than in armchair CNTs, which explains the weak chirality dependence in thermal conductance.

Conclusion

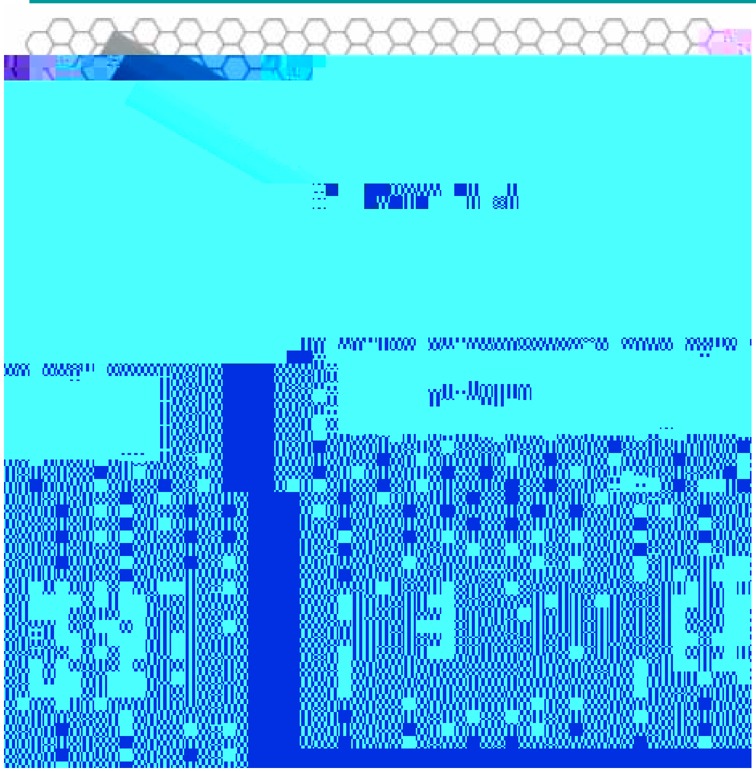
- Thermal conductance shows a universal linear dependence on radial strain over all the elastic range.

- Thermal conductance is quite robust against large radial deformation (up to the strain of 70%), indicating that CNTs are perfect phonon waveguides.

- Physical origin: under the elastic deformation, the mechanical properties don’t change much.

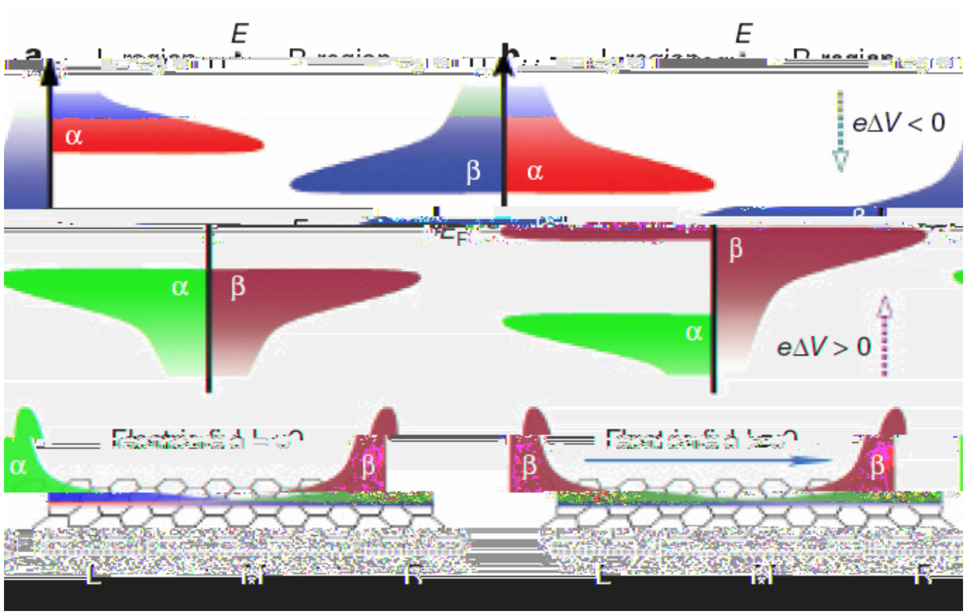
Graphene nanoribbons: Anisotropy

Nanoelectronics devices



Q. Yan, *et al*., Nano Lett. **7,** 1469 (2007).

Spintronics



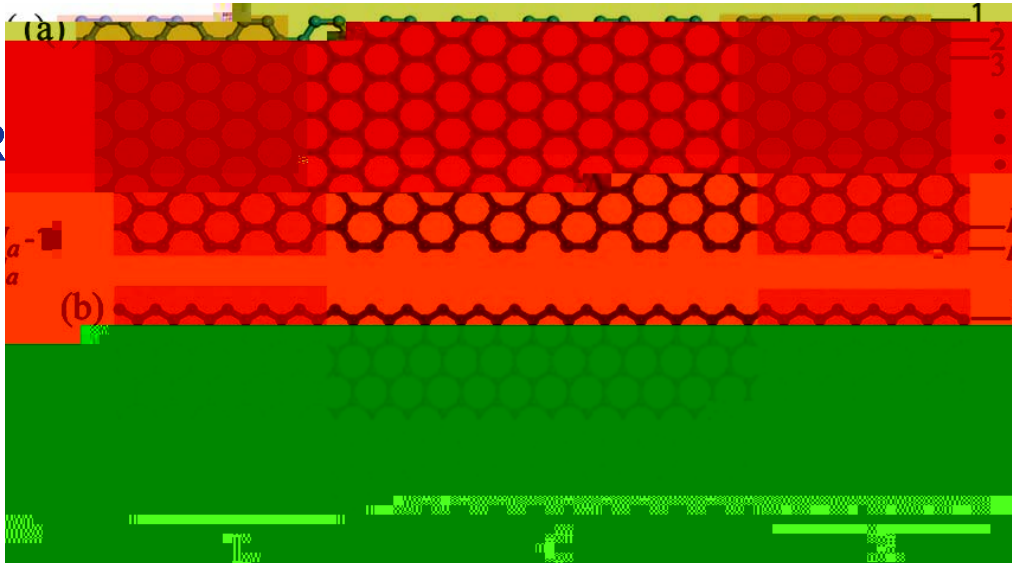
Y. W. Son, *et al*., Nature **444,** 347 (2006).

Model: edge effects

- **Width and edge shape** of GNRs significantly influence *electronic properties*.
- Effects on thermal transport properties?
- Critical for developing any practical graphene-based devices.

Armchair GNR

Zigzag GNR

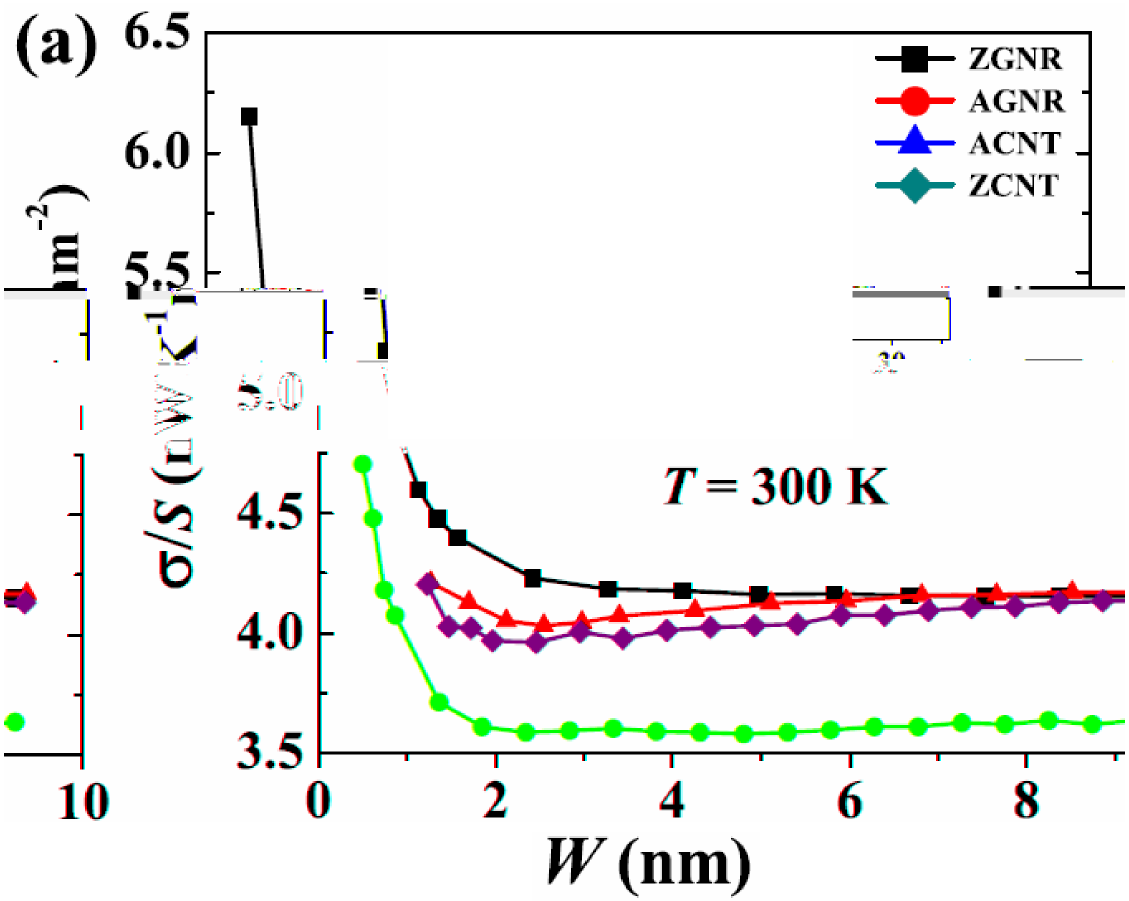


Results and Discussion

Scaled thermal conductance: σ /*S*

($S = W\delta$, $\delta = 0.335$ nm)

W refers to circumference for CNTs.

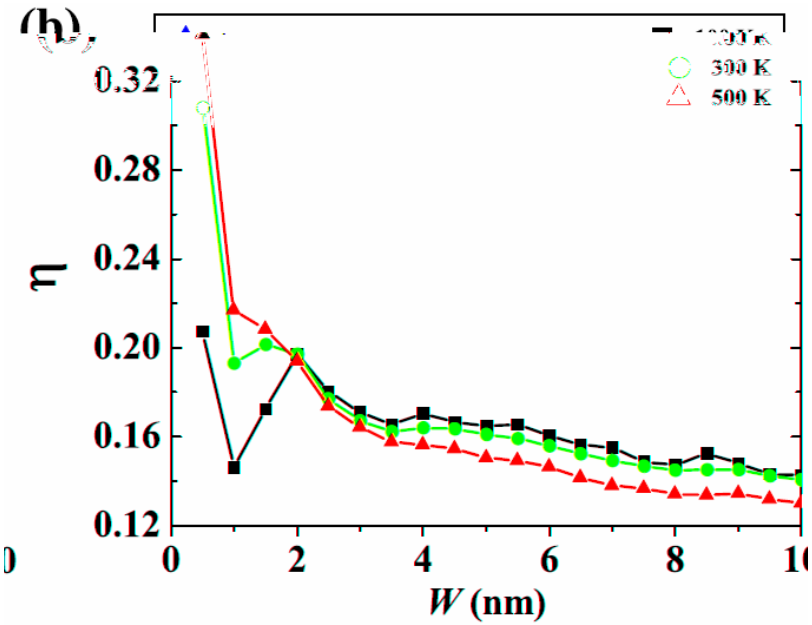


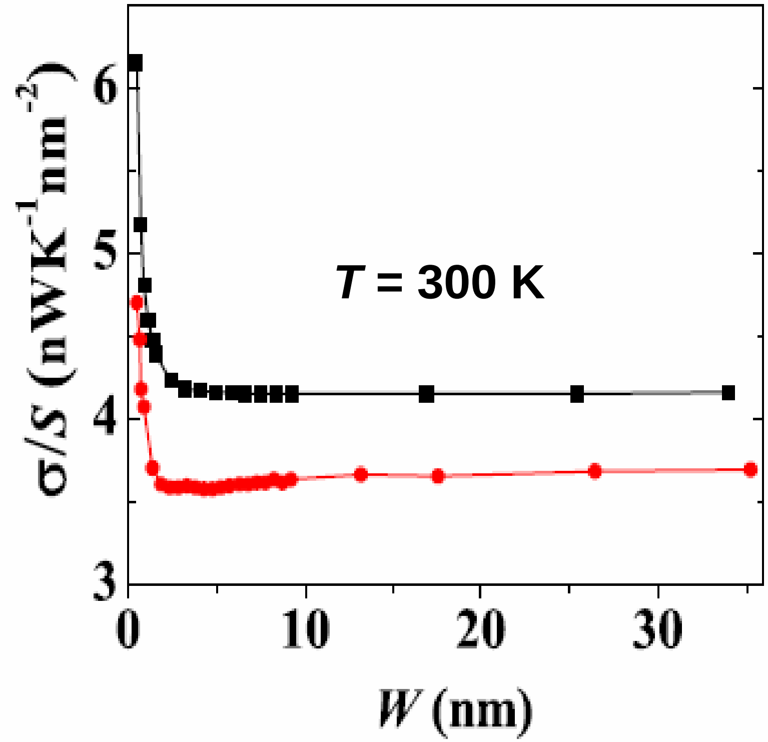
Thermal conductance:

- Isotropic in CNTs
  σ /*S*(ZCNT) ~ σ /*S*(ACNT)
- Anisotropic in GNRs
  σ /*S*(ZGNR) > σ /*S*(AGNR)

Results and Discussion

Anisotropy factor: $\eta = \frac{(\sigma / S)_{ZGNR}}{(\sigma / S)_{AGNR}} - 1$





The anisotropy (up to ~ 30%) may ***disappear*** when W ~ 140 nm.

Thermal conductance is isotropic in graphene.

K. Saito, *et al.*, Phys. Rev. B **76,** 115409 (2007).

Results and Discussion

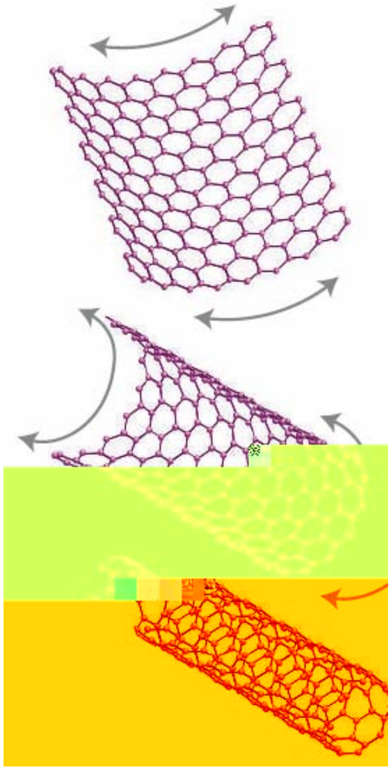
<110> SiNW exhibits $\sigma/S(300\text{ K})$ 50% and 75% larger than <100> and <111> SiNWs. T. Markussen, *et al*., Nano Lett. **8,** 3771 (2008).

Anisotropic phonon structure of bulk Si.

What is the origin of anisotropic thermal conductance in GNRs?

Isotropic in graphene and CNTs,
but anisotropic in GNRs.

Different boundary condition at edges.



Most phonon modes are influenced by the boundary.
This makes the analysis complicated.

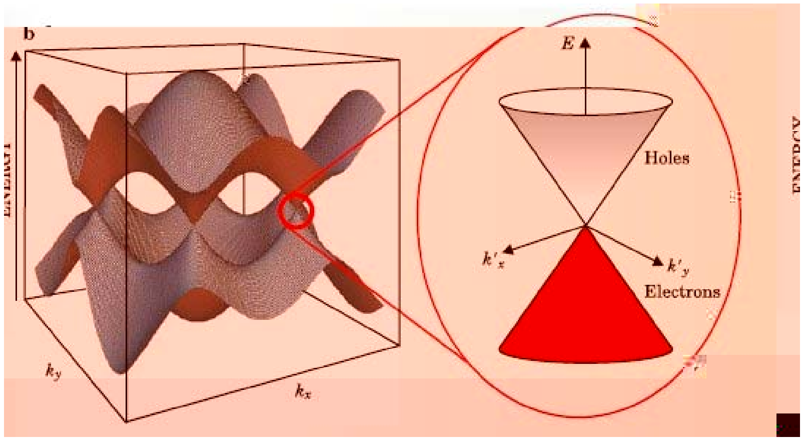
Conclusion

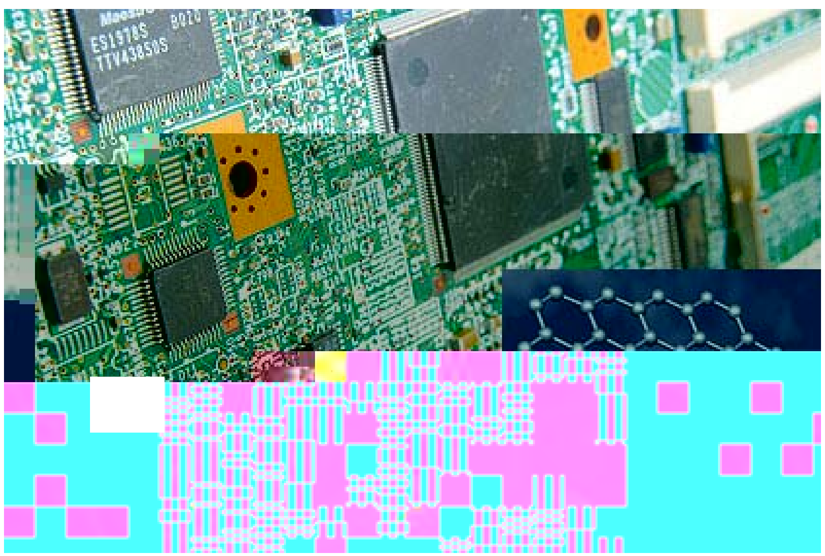
- **Anisotropy** room temperature thermal conductance of ZGNRs is up to ~ 30% larger than that of AGNRs. The anisotropy disappears when $W > 100$ nm.

- This intrinsic anisotropy originate from different boundary condition at ribbon edges.

- Important implications for the applications of GNRs in nanoelectronics and thermoelectricity.

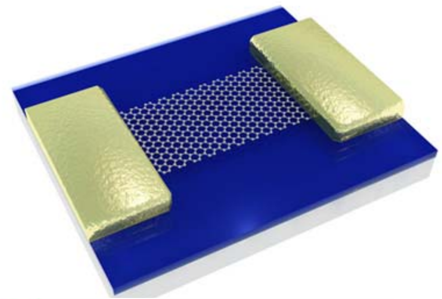
Graphene-based nanostructures

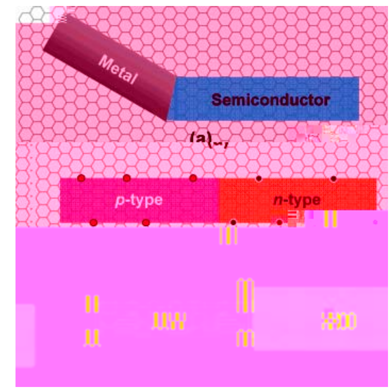


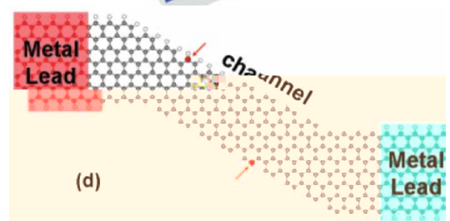


Two major building blocks:

Graphene junction

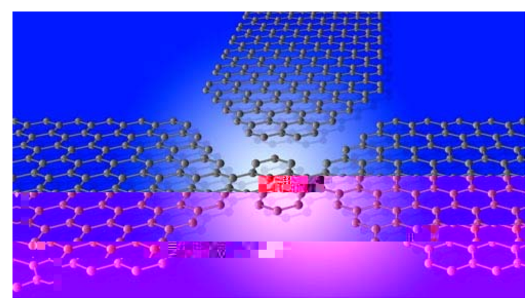






Q. M. Yan, *et al*. Nano Lett. **7**, 1469 (2007).

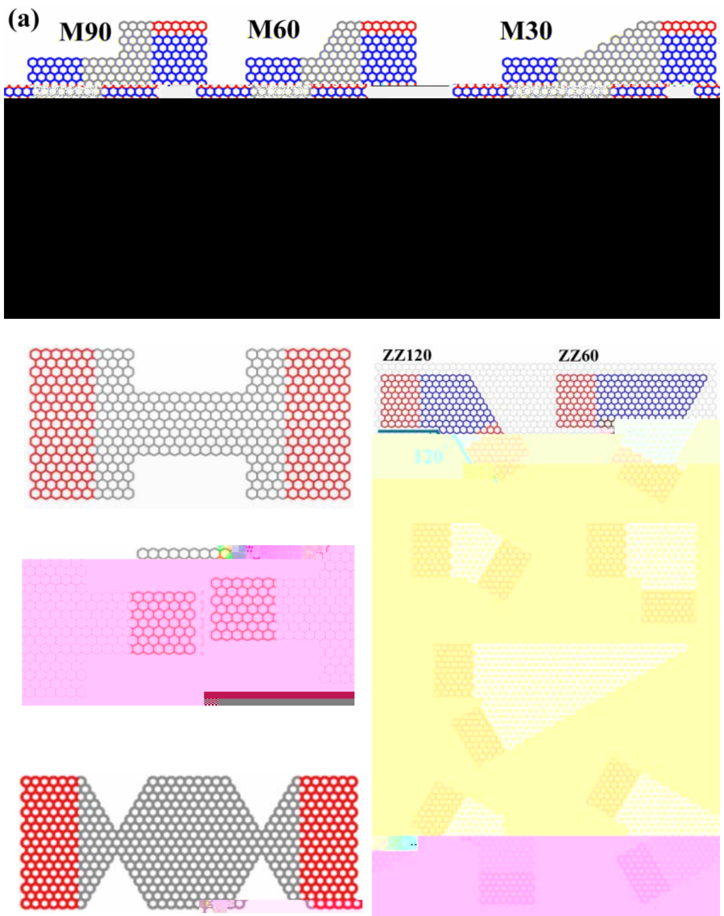
Graphene quantum dot



Richard Van Noorden Nature 442, 228 (2009).

Structure & thermal transport

Graphene-based nanodevices

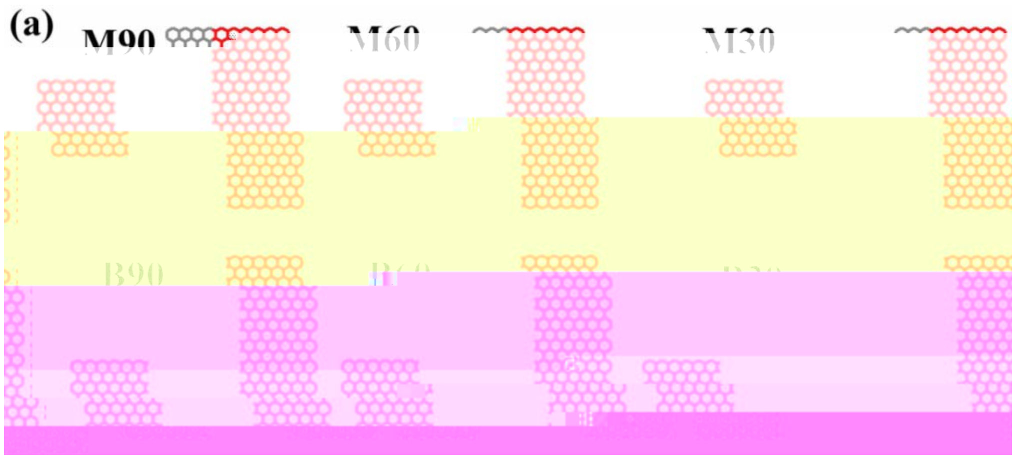


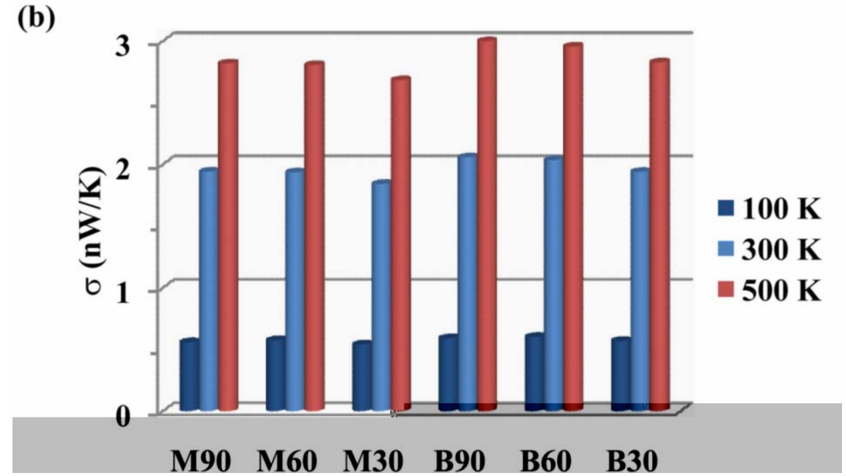
Red parts: thermal leads

Structural characteristics: contact geometries, widths, edge shapes, connection angles …

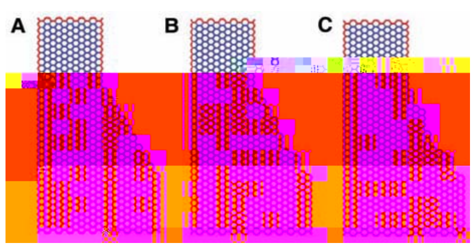
How do they affect thermal transport?

Contact geometry

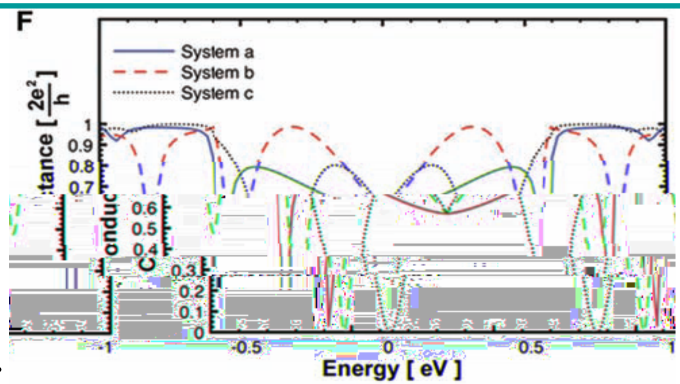




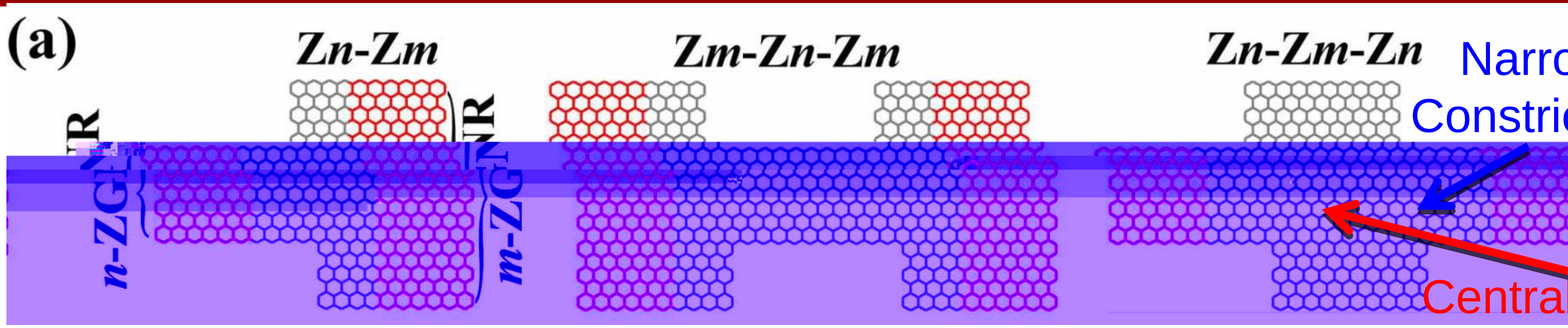
Thermal transport is ***insensitive*** to ***the detailed structure in the contact region***, quite different from electronic transport.



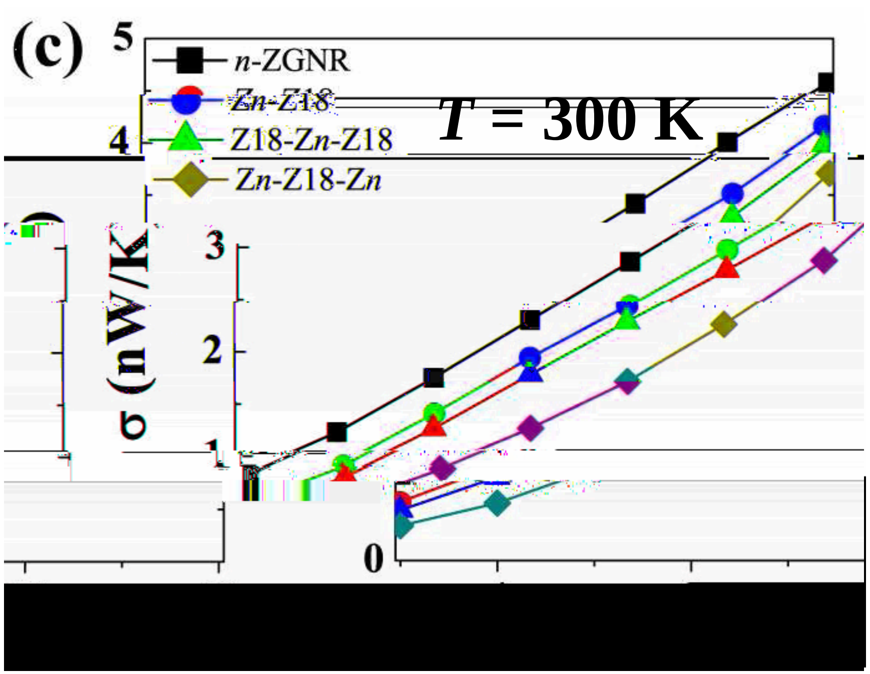
X. Jia, *et al.*
Science 323, 1701 (2009).



Width effects



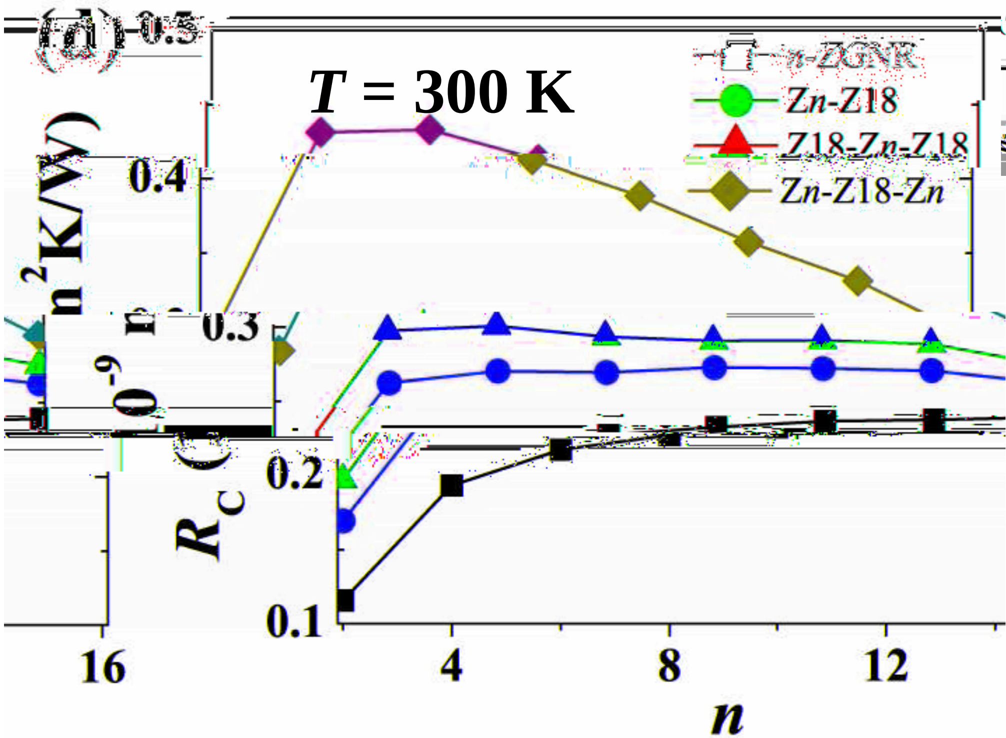
Single-Interface Junction   Double-Interface Junction   Quantum Dot (QD)

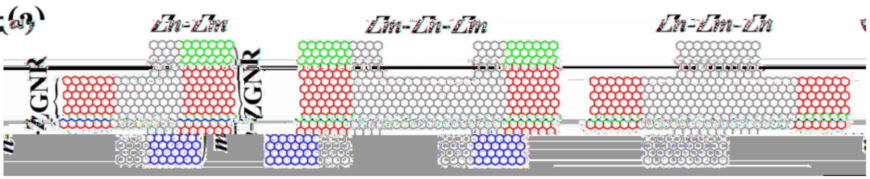


The width variation of the *wide* part has little influence on thermal conductance of the junctions

The width variation of the *narrow* part strongly affects thermal conductance.

Thermal contact resistance





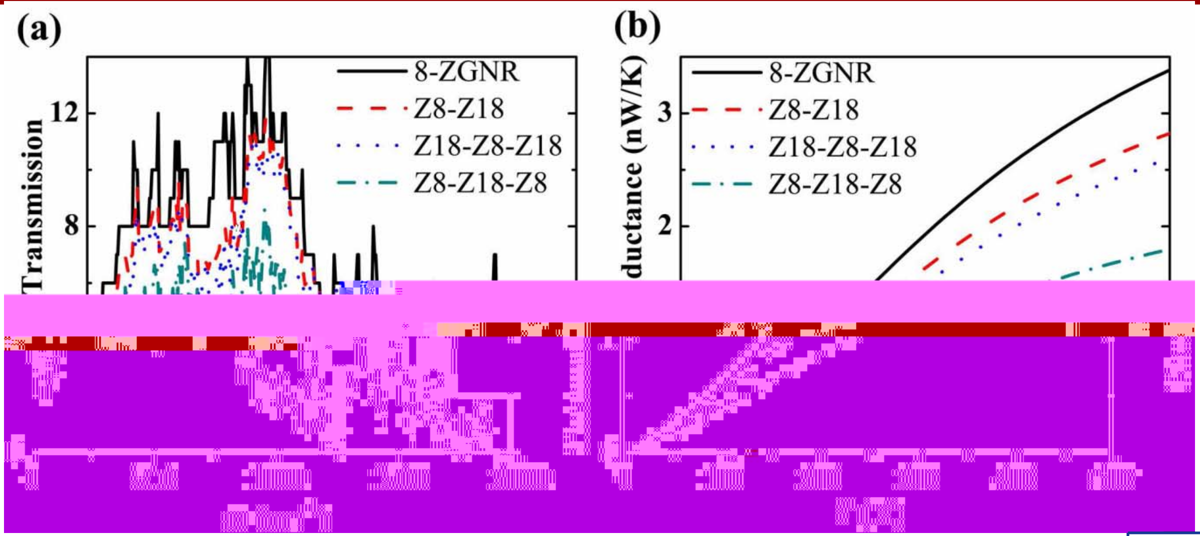
$R_C = \Delta T / j = S / \sigma$
($S = W \times \delta$, $\delta = 0.335$ nm)

R_C of graphene nano-junctions is ~ 3.0×10^{-10} m^2K/W at 300 K.

R_C between graphene and SiO_2
($5.6 \times 10^{-9} \sim 1.2 \times 10^{-8}$ m^2K/W at 300 K)
Z. Chen *et al*., APL 95, 161910 (2009).

Interestingly, R_C of double-interface junctions is just slightly higher than that of single-interface junctions.

Phonon LDOS

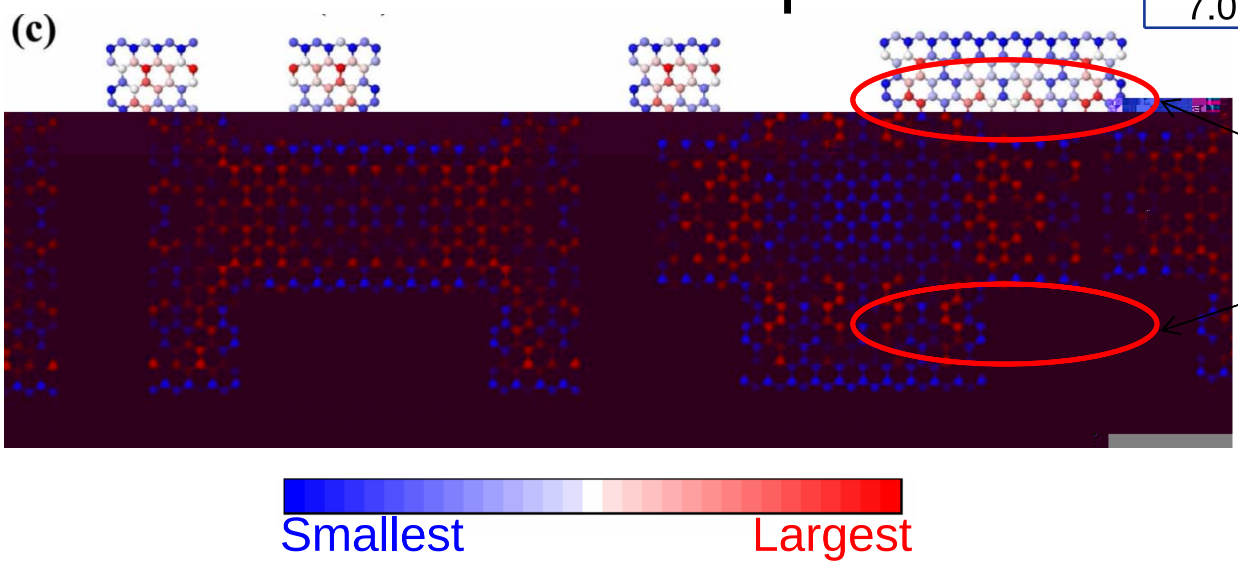


Local Density of States (LDOS) for phonons

$$\rho_i(\omega) = -\frac{2\omega}{\pi} \sum_{\alpha=x,y,z} \mathrm{Im}[G^r(\omega)]_{i\alpha,i\alpha}$$

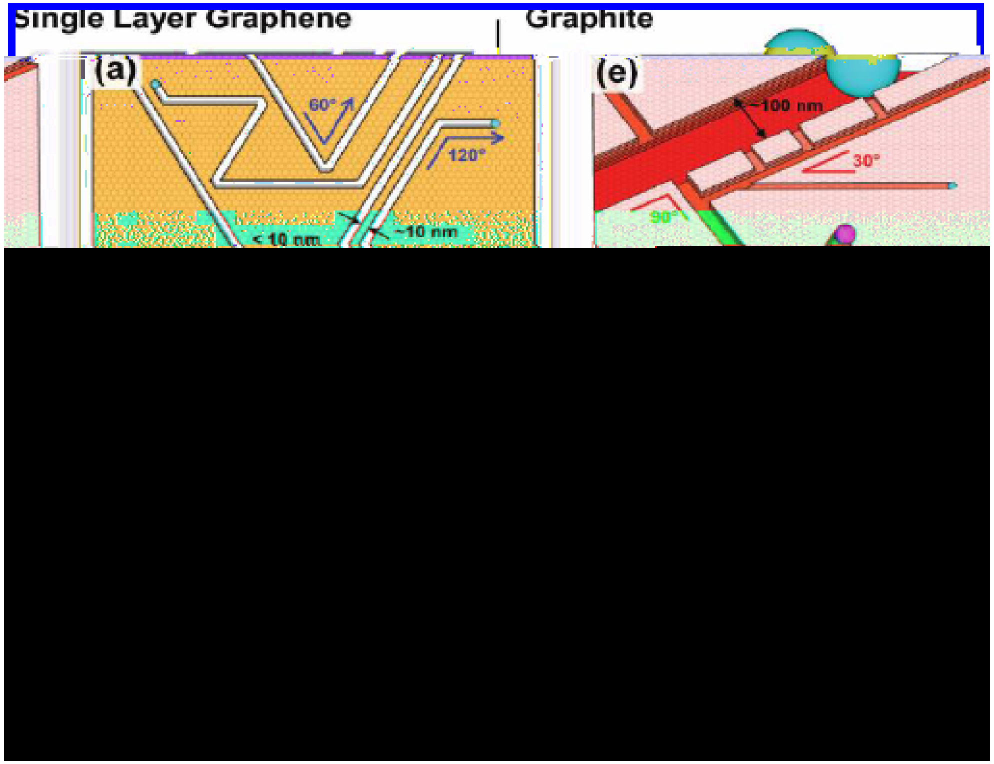
Transmission at 900 cm^{-1}			
8-ZGNR	Z8-Z18	Z18-Z8-Z18	Z8-Z18-Z8
7.00	6.05	5.27	2.62

LDOS for 900 cm^{-1} phonons

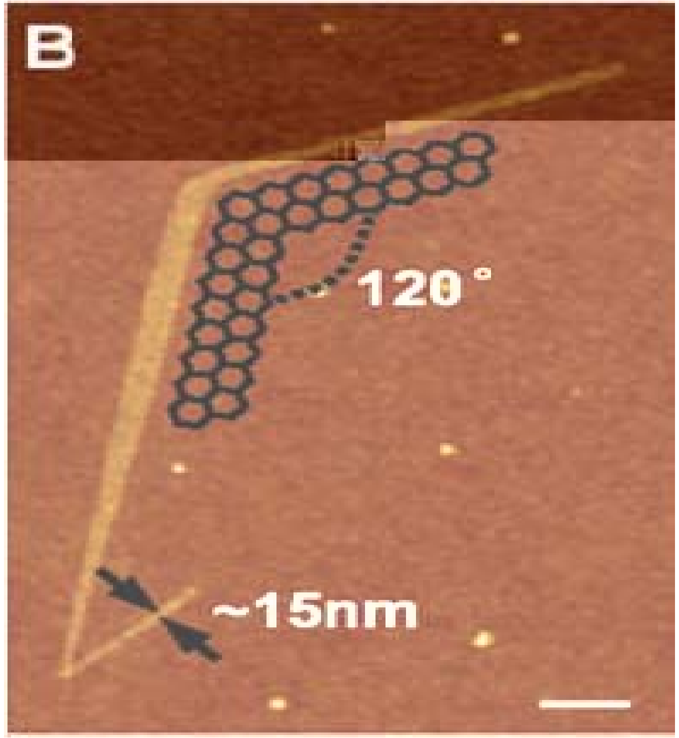


Easily scattered into the protruding part

Graphene junctions in experiments



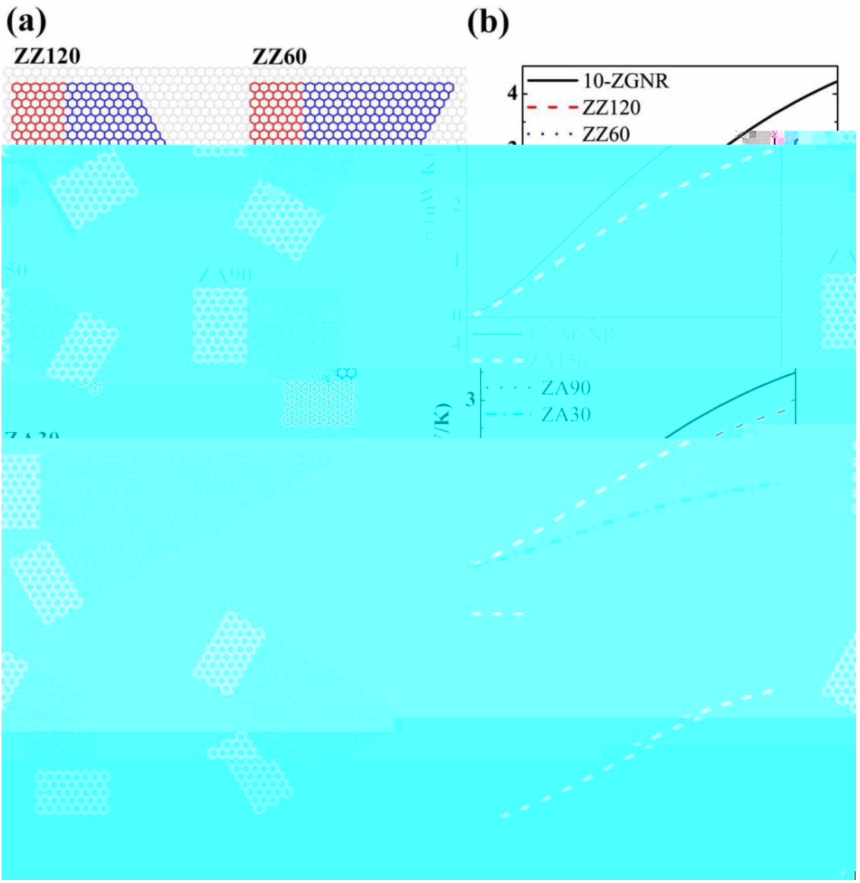
L. C. Campos, *et al*.
Nano Lett. 9, 2600 (2009).

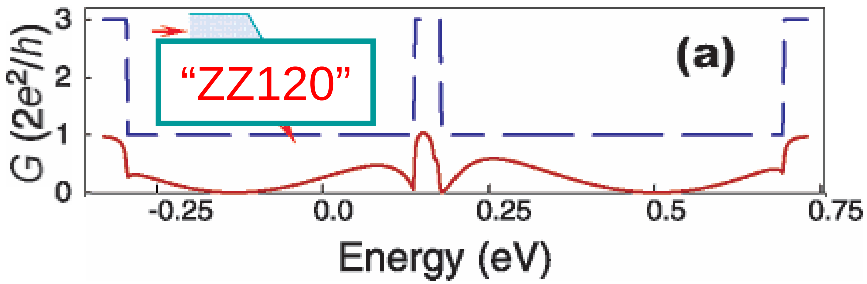


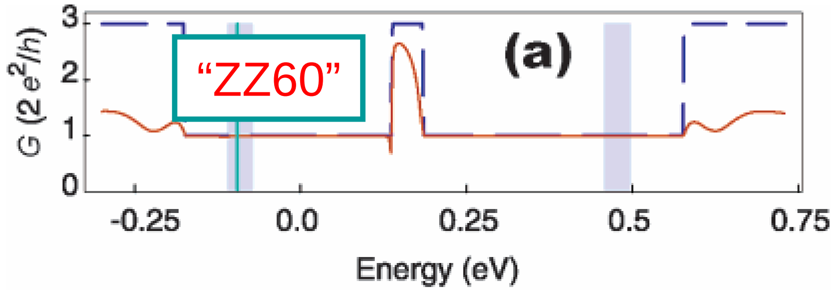
X. Li, *et al*.
Science 319, 1229 (2008).

Connection angles: 30˚, 60˚, 90˚,120˚, 150˚

Connection angles



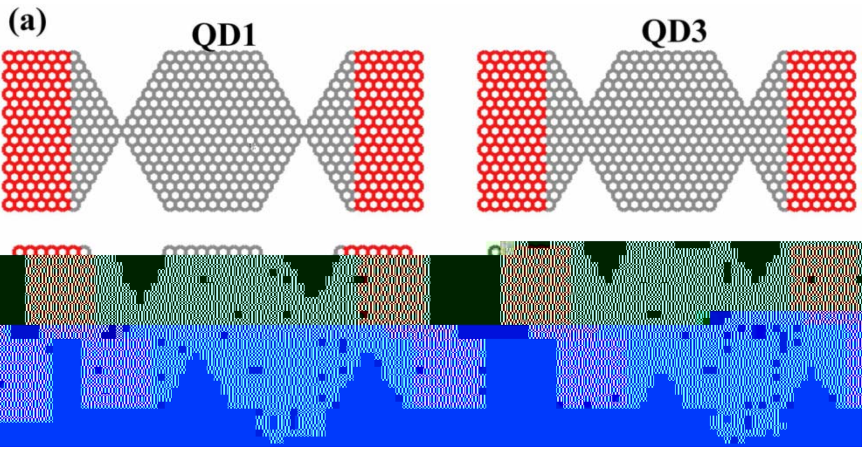


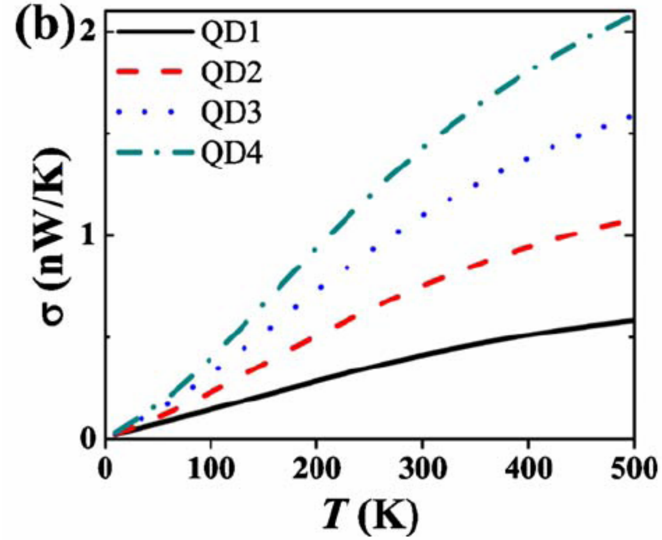


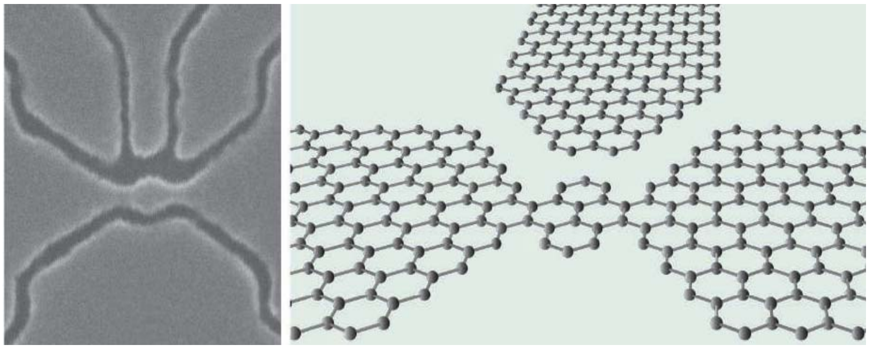
D. A. Areshkin and C. T. White
Nano Lett. 7, 3253 (2007).

Graphene junctions with ***smaller connection angles*** show ***lower thermal conductance*** and ***higher electronic conductance***.

Graphene QDs







R. M. Westervelt , Science 320, 324 (2008).

QDs have extremely low σ :
1.Phonons are easily scattered into the protruding parts of QDs;
2.The constrictions could be very narrow.

1. Heat dissipation is difficult in QDs.
2. The thermoelectric efficiency of QD structures may be largely enhanced.

Conclusion

- Thermal conductance is insensitive to the detailed structure of the contact region but substantially limited by the narrowest part (bottleneck!) of the systems.

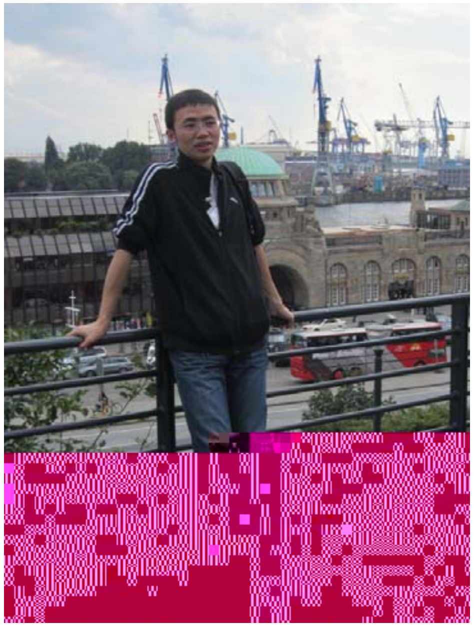
- Thermal contact resistance R_C in graphene nanodevices is quite low (10^{-10} m^2K/W at 300 K). Interestingly, R_C of double-interface junctions is just slightly higher than that of single-interface junctions.

- Different even opposite dependences of thermal and electronic transport properties on the structural characteristics may find wide applications in nanoelectronics and thermoelectricity.

Summary

- Calculation method for quantum thermal transport is developed.
- CNTs are robust phonon waveguides against radial deformations.
- GNRs exhibits intrinsic anisotropy (up to 30%) which originates from different boundary condition at ribbon edges.
- Thermal conductance is substantially limited by the narrowest part of the systems.
- Low thermal contact resistance in graphene.

Collaborators / students




Thank You !