The 9th Asian Workshop on First-Principles
Electronic Structure Calculations

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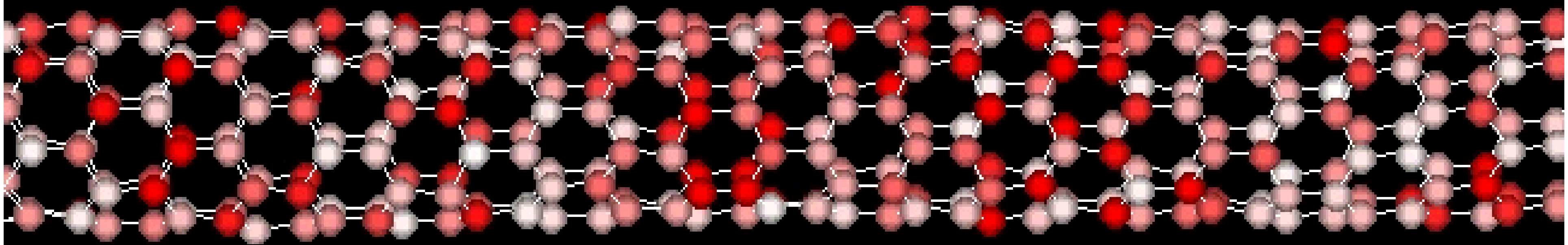
Quantum Theory of Thermal Transport in Carbon Nanotubes



Takahiro Yamamoto
Department of Physics, Tokyo University of Science

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- Basic Theory
  - NEGF method for phonon transport
- Results
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  - Electron-derived thermal transport in CNTs
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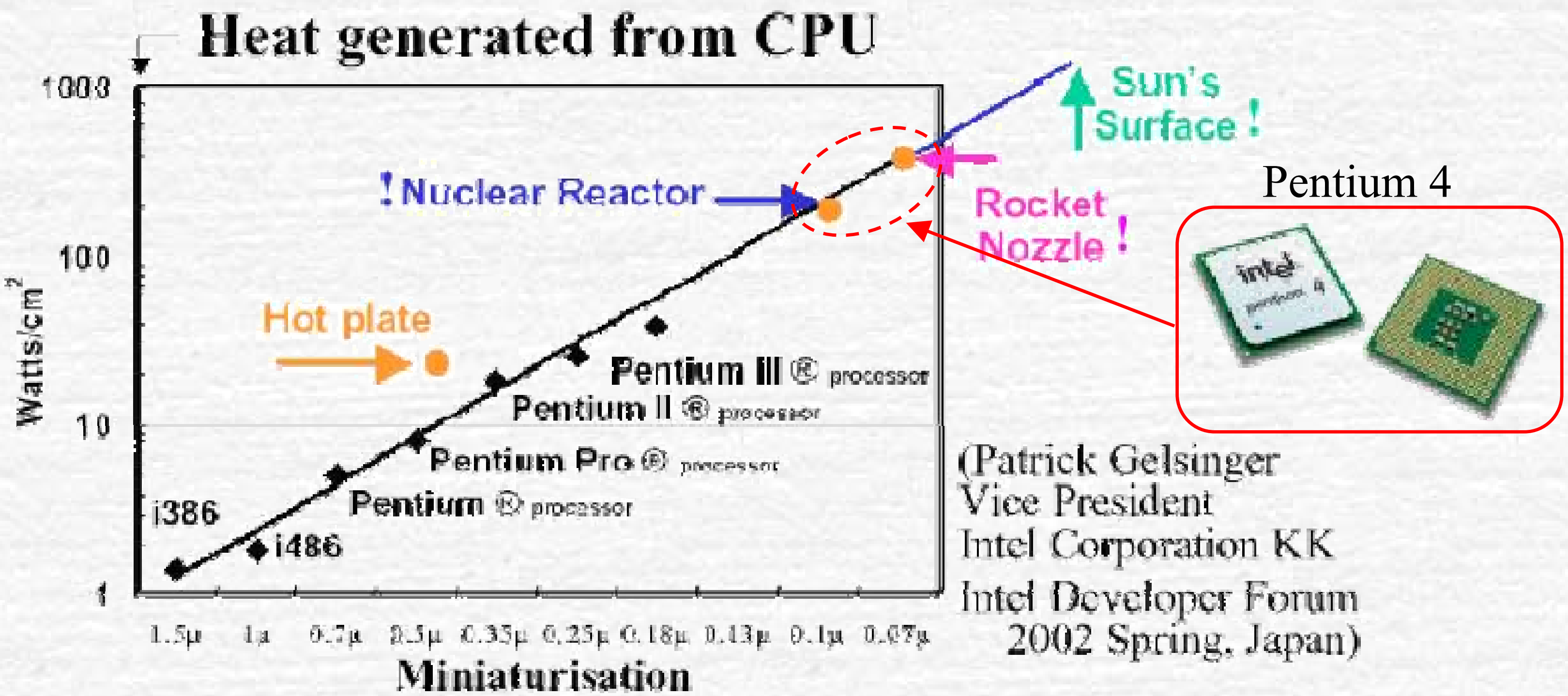
INTRODUCTION

- Miniaturization of Electrical Devices
  - Large Joule Heating
  - High Power Consumption

  } serious bottleneck

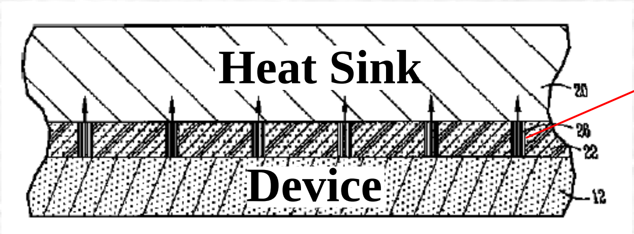


(Patrick Gelsinger
Vice President
Intel Corporation KK
Intel Developer Forum
2002 Spring, Japan)

CNT-based Heat Remover

- High Thermal Conductivity: ~3000W/m･K [Theoretical]
- Good Thermal Stability: ~2500°C (vacuum), 450°C(atmosphere)

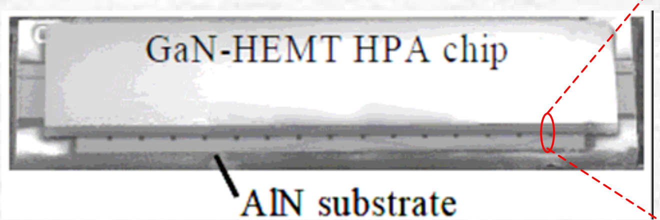
Intel Co. (2003)

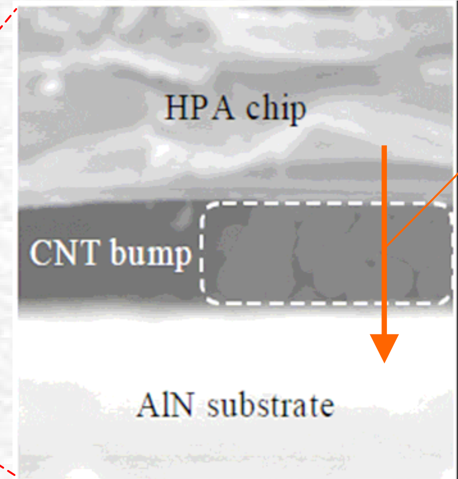


CNT bundles

Laid-open disclosure public patent bulletin : 2003-249613

Fujitsu Lab. Ltd (2005)



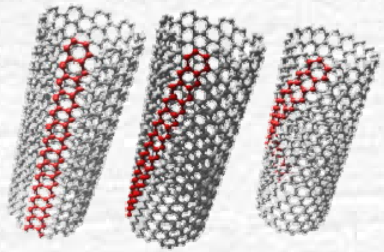


Heat flow

Thermal conductivity: 1400 W/m･K

Iwai *et al*., IEEE IITC Tech. Digest, pp. 257-260 (2005)

Experimental Values of Thermal Conductivity



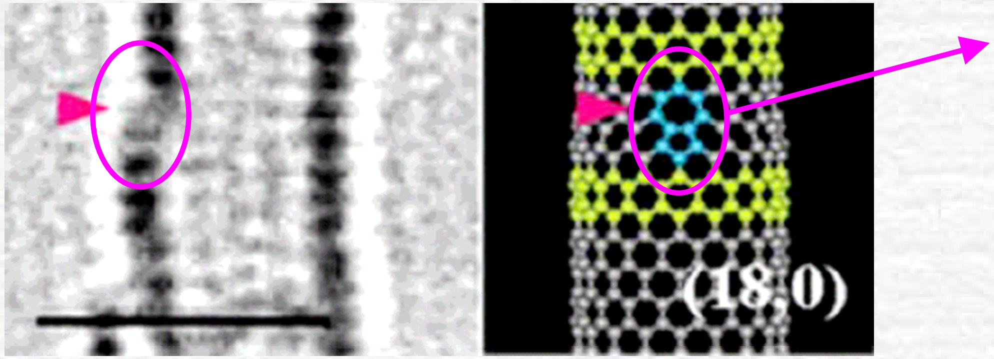
- J. Hone *et al*., Phys. Rev. B **59**, 2514 (1999). => 35
- W. Yi et al., Phys. Rev. B **59**, 9015 (1999). => 25
- J. Hone *et al*., Appl. Phys. Lett. **77**, 666 (2000). => 200
- P. Kim *et al*., Phys. Rev. Lett. **87**, 215502 (2001). => 3000
- D. J. Yang *et al*., Phys. Rev. B **66**, 165440 (2002). => 200
- M. Fujii *et al*., Phys. Rev. Lett. **95** 065502 (2005). => 2000
- Iwai *et al*., IEEE IITC Tech. Digest, pp. 257 (2005). => 1400 [W/m･K]

Possible Reasons

1) Difference among experimental method
2) Purity of samples: defect influence
3) Contact thermal resistance at interface between CNT and substrate/device
4) Heat dissipation to surrounding
5) Phonon-phonon scattering

Various Types of Defect in CNTs

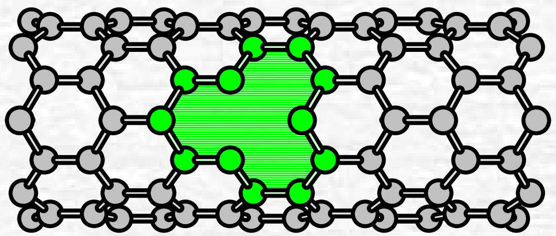
- Defects in CNTs



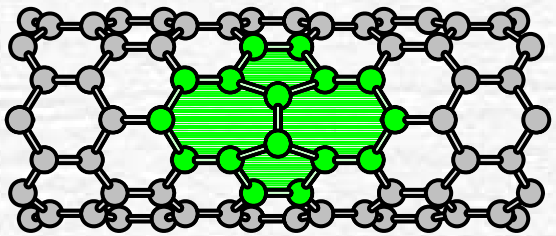
Many defects exist in synthesized CNTs.

A. Hashimoto, *et al.*, Nature **430**, 870 (2004)

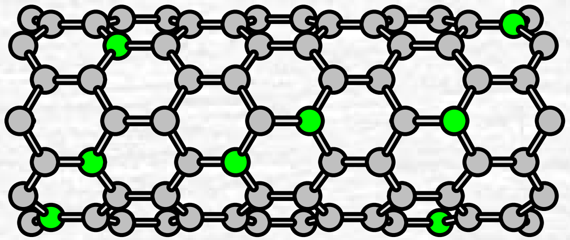
- Various Types of Defects in CNTs



Vacancy defect

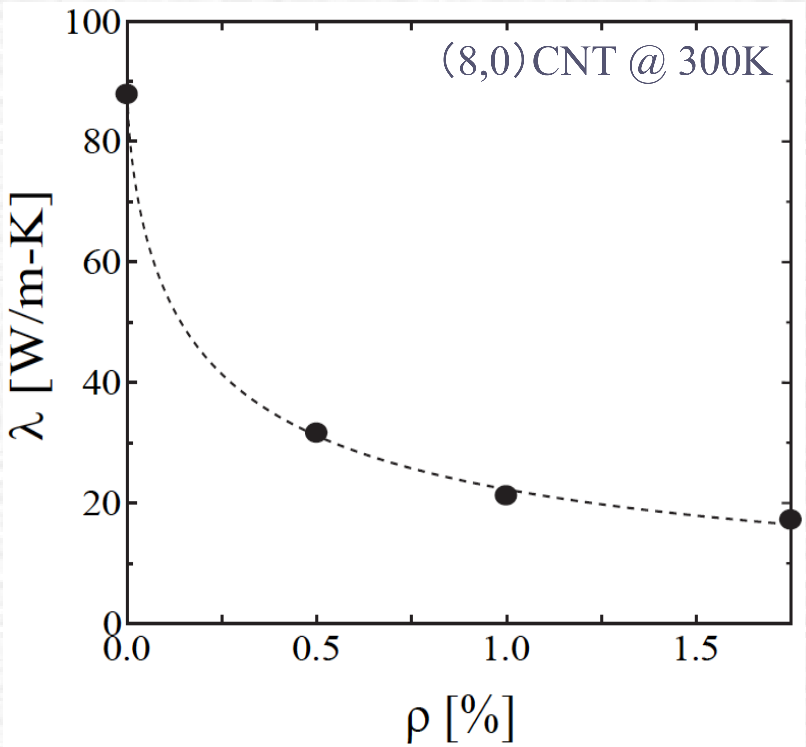


SW defect



Isotope Impurities

Reduction of Thermal Conductivity due to Mono-Vacancies



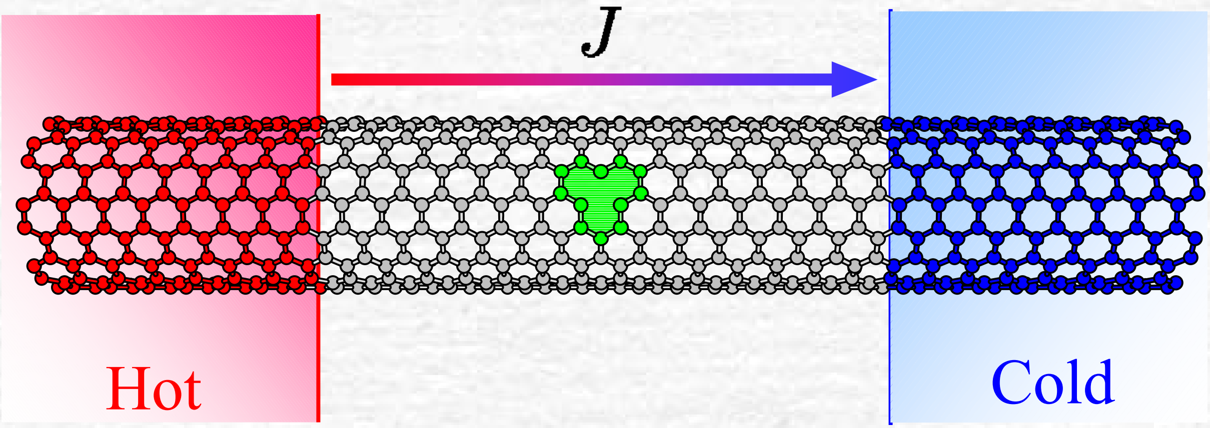
Kondo, Yamamoto, Watanabe:
e-J. Surf. Sci. Nanotech. **4**, 239 (2006).

Thermal conductivity decreases rapidly due to vacancies

Objectives

- Clarification of
  - Effect of defects on thermal transport in CNTs
  - Mechanism of phonon scattering with the defects

- Provision of
  - Way to improve reduced thermal conductivity



The NEGF Method for Phonon Transport at Nanoscale

Phonon-derived thermal current

$$J_{\rm th} = \int_0^\infty \frac{d\omega}{2\pi} \hbar\omega {\rm Tr}\left[\boldsymbol{\Sigma}_L^{>}(\omega)\boldsymbol{D}_S^{<}(\omega) - \boldsymbol{\Sigma}_L^{<}(\omega)\boldsymbol{D}_S^{>}(\omega)\right]$$

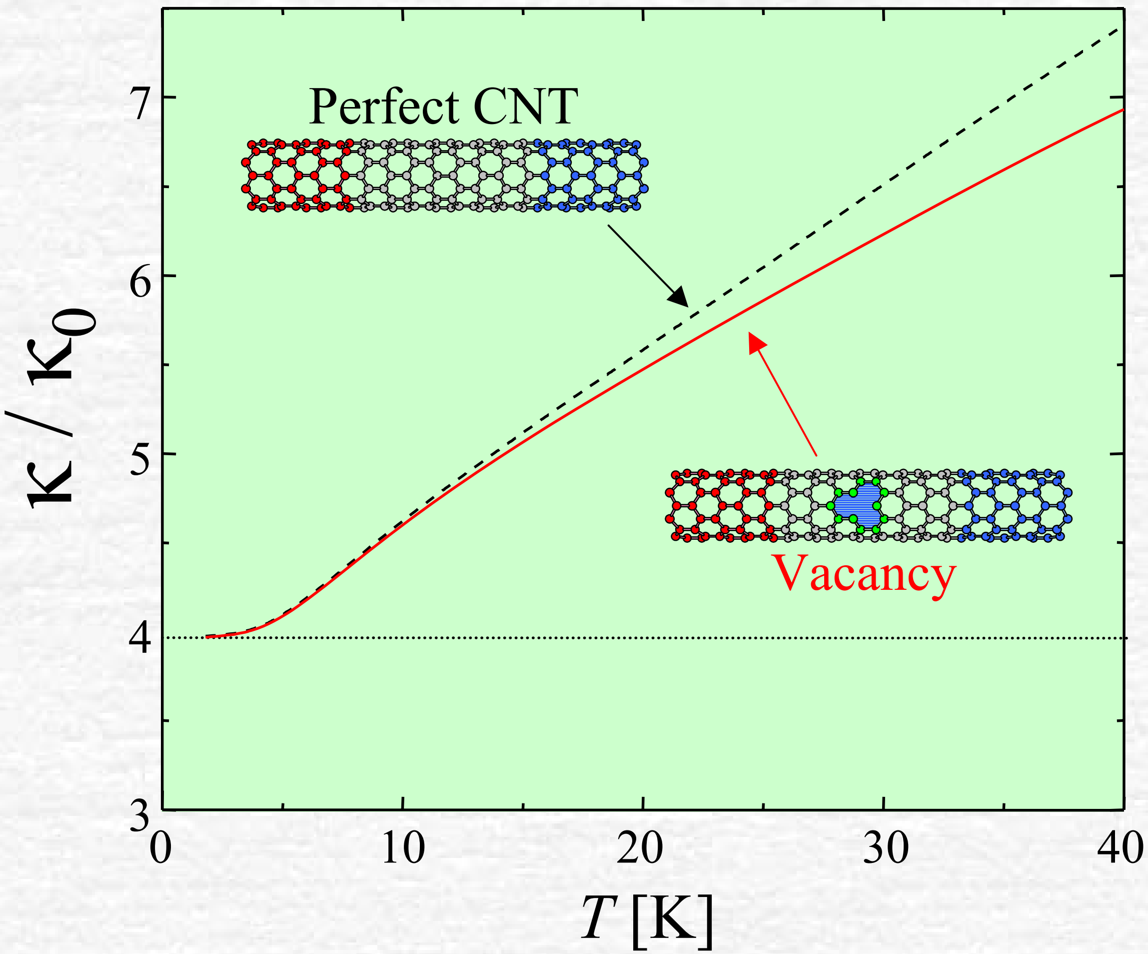
- $\boldsymbol{\Sigma}_L^{\gtrless}(\omega)$: Greater/Lesser self-energy for the left lead
- $\boldsymbol{D}_S^{\gtrless}(\omega)$: Greater/Lesser Green's function for the scattering region

Yamamoto and Watanabe, Phys. Rev. Lett. **96**, 255503 (2006)

Advantages:

1) Applicable to nanoscale objects with complex atomic structure
2) Local physical quantities
3) Applicable to the interacting phonon transport

Reduction of Thermal Conductance due to Vacancy



Universal quantum
of thermal conductance

$$\kappa_0 = \frac{\pi k_B T}{3h}$$

Fig: Thermal conductance κ scaled by the universal quantum κ_0 of (8,8) CNT at low *T*.

Phonon Transmission Functions

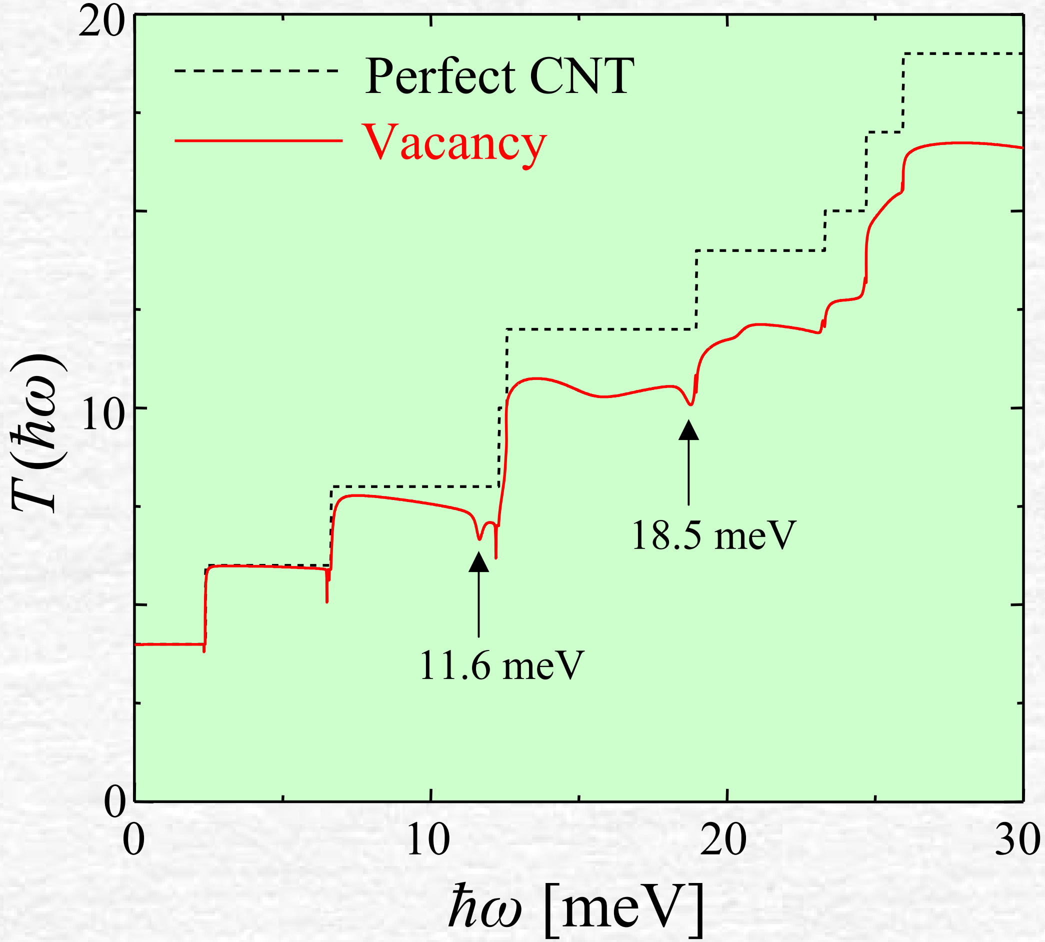
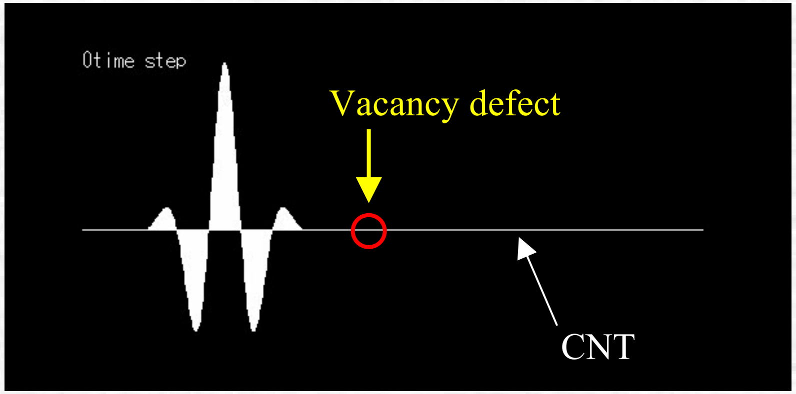
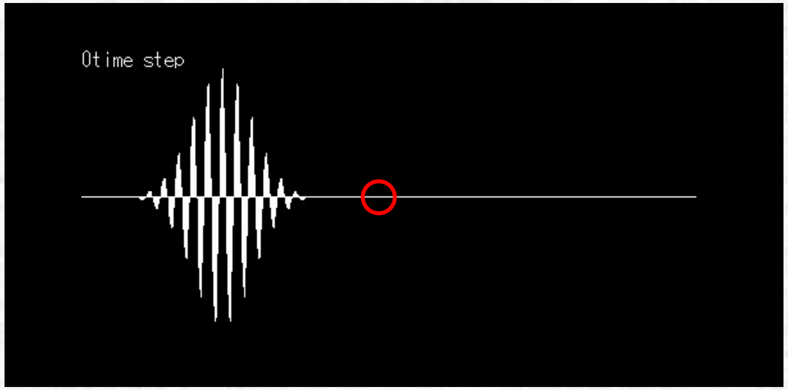


Fig: The phonon transmission functions of (8,8) CNT with/without defects

Perfect Transmission



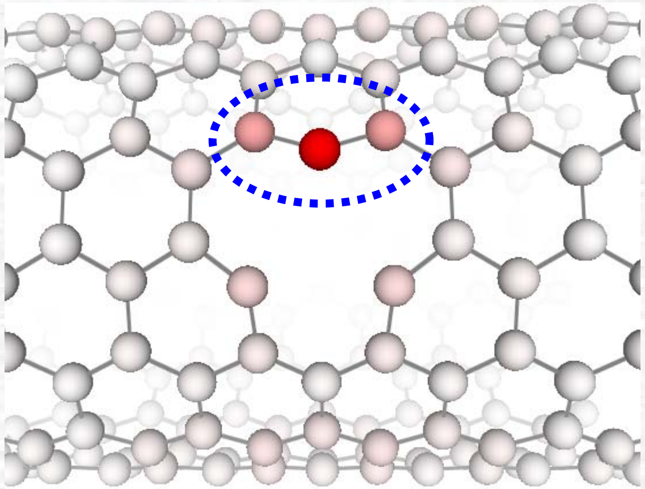
Strong Reflection @ Dip

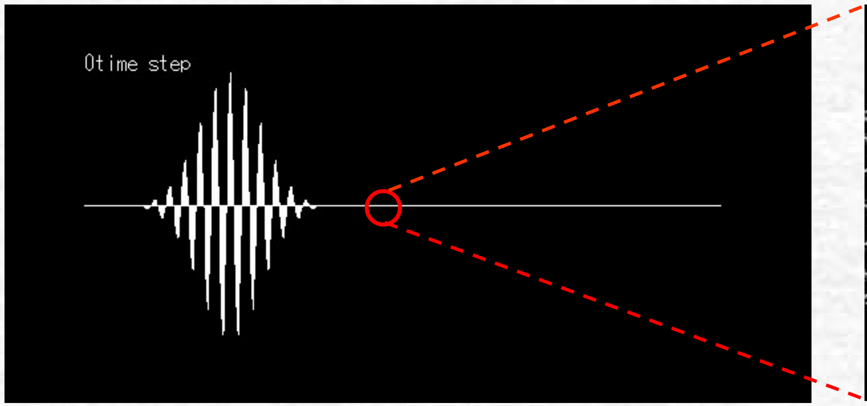


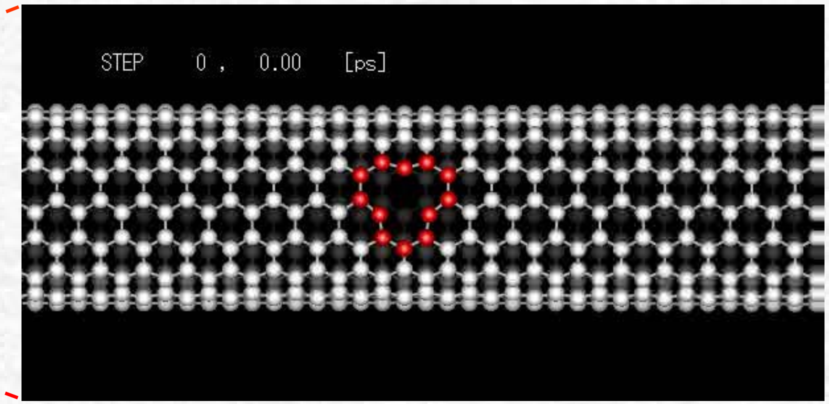
N. Kondo, T.Y., K. Watanabe, Jpn. J. Appl. Phys., **45**, L963 (2006)

Phonon Density around Vacancy

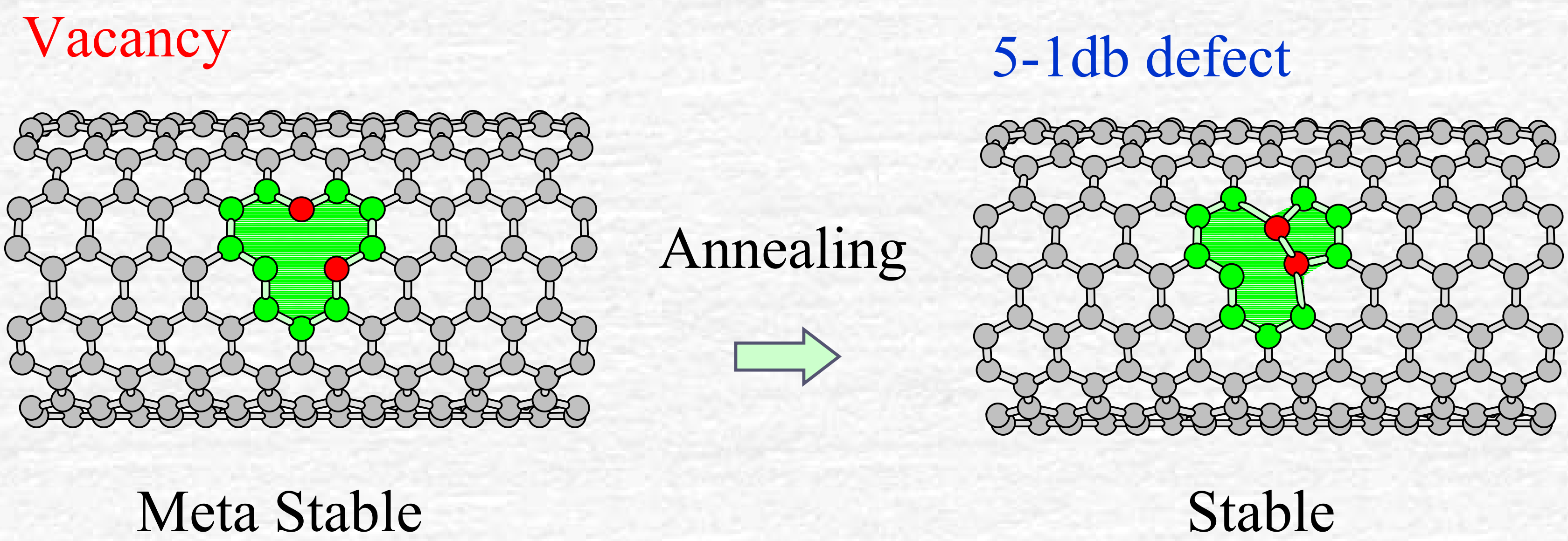
$\hbar\omega$=11.6 meV







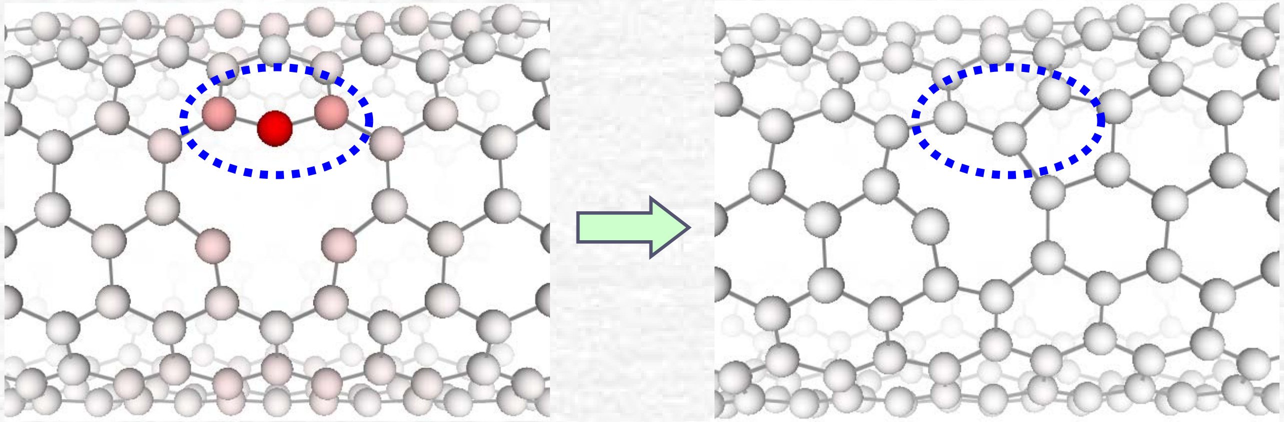
Structural Change due to Thermal Annealing



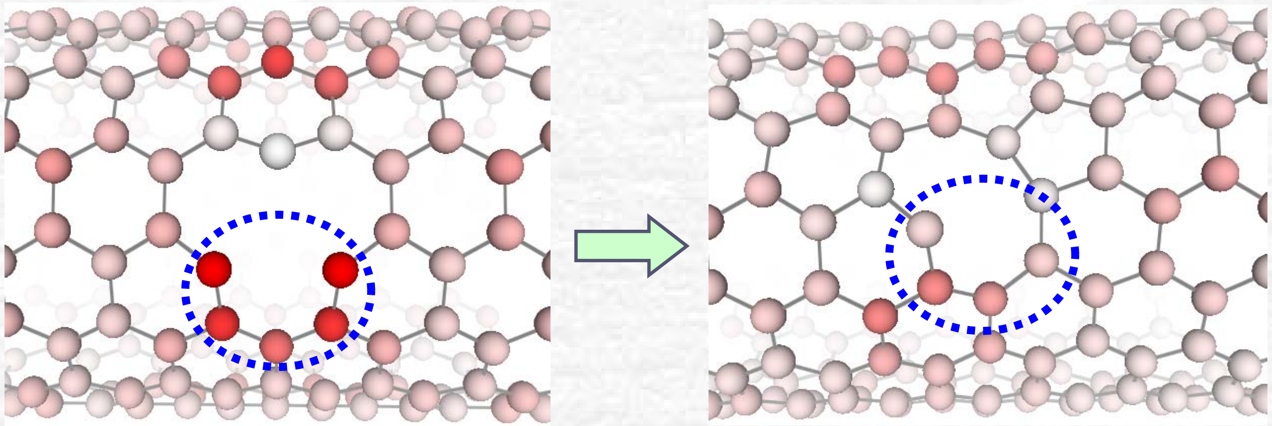
1) A.V. Krasheninnikov and K. Nordlund, J. Vac. Sci. Technol. B **20**, 728 (2002)
2) Y. Miyamoto *et al*., Physica B **323**, 78 (2002)

Phonon Number Density

$\hbar\omega$=11.6 meV



$\hbar\omega$=18.5 meV



Improvement of Thermal Conductance

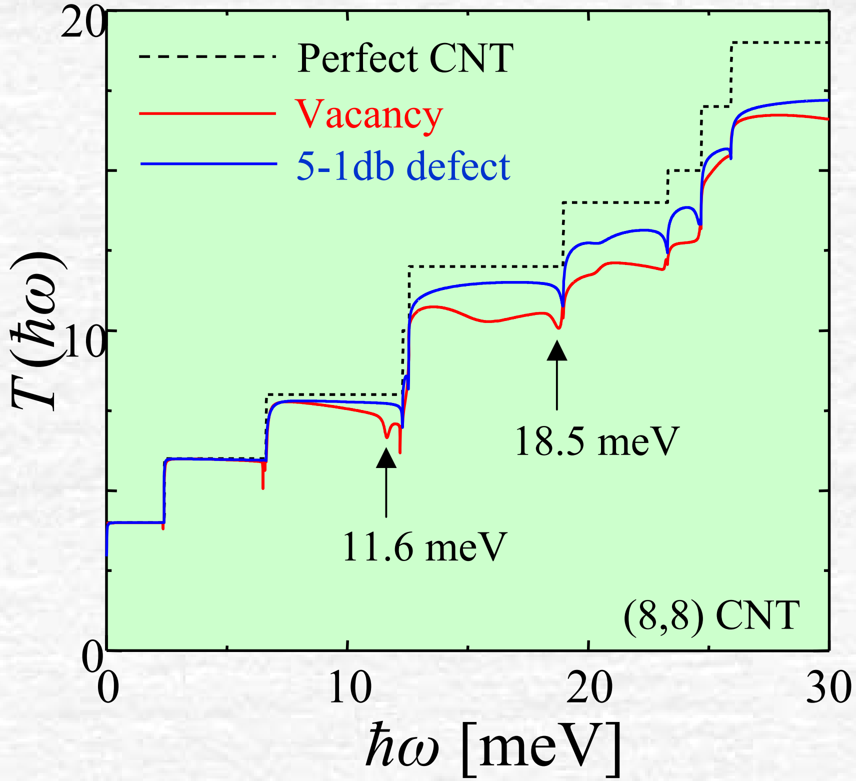


Fig: Phonon transmisssions

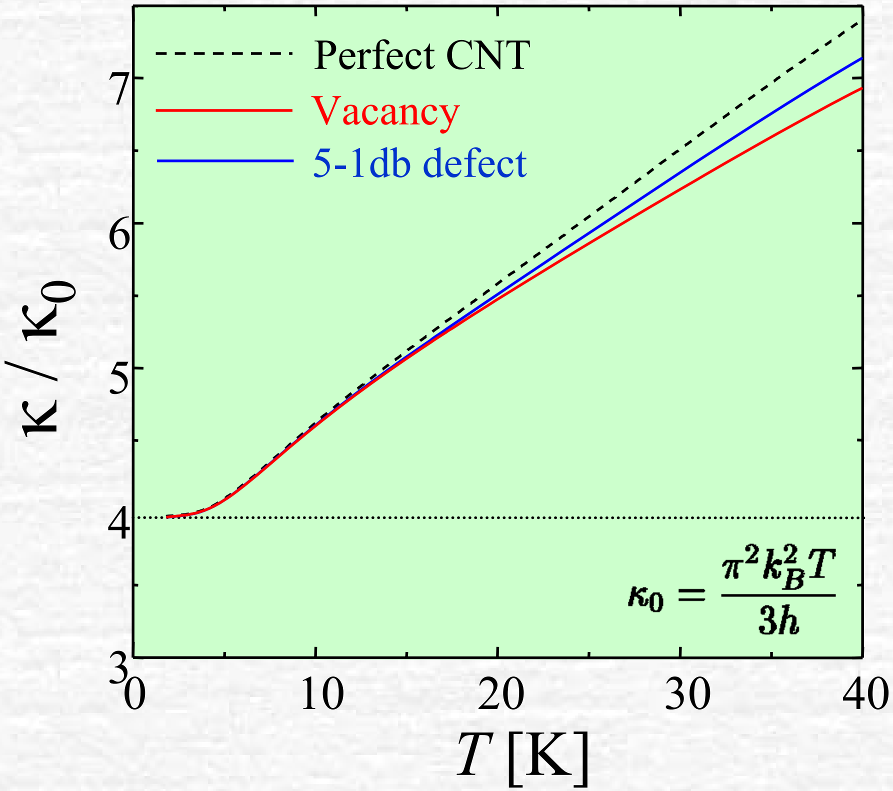
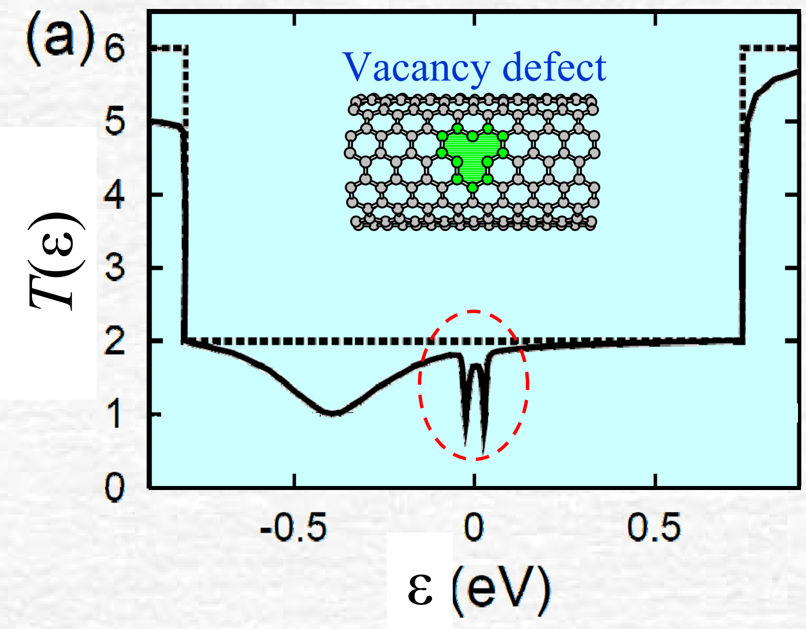


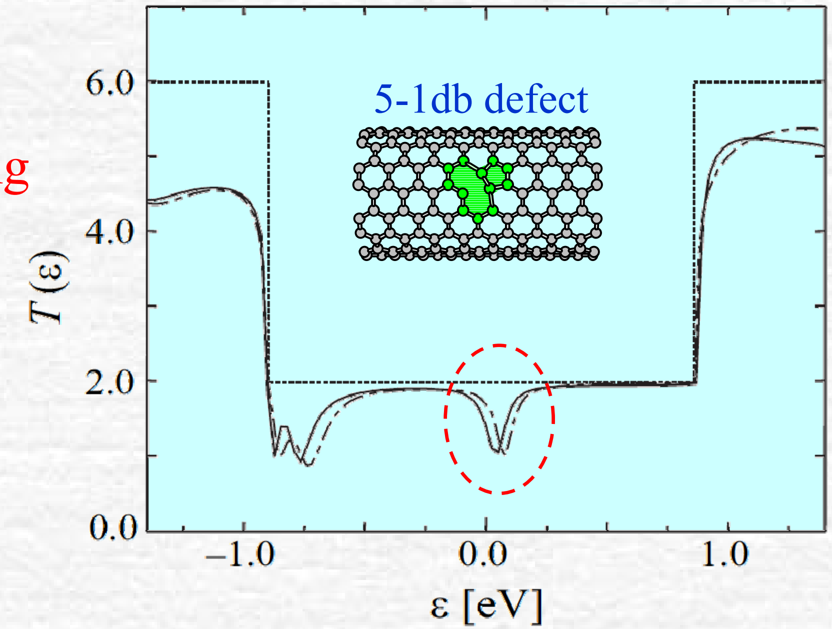
Fig: Thermal Conductances

"*Electron Contribution*" to Thermal Conductance in Metallic CNTs

Electron-derived thermal conductance at low bias: $\kappa_{\rm el} = \frac{\pi k_B T}{3h} T(\epsilon)$

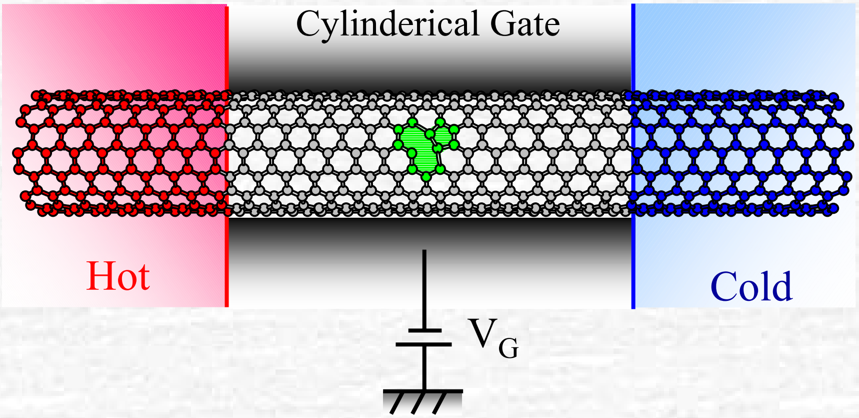


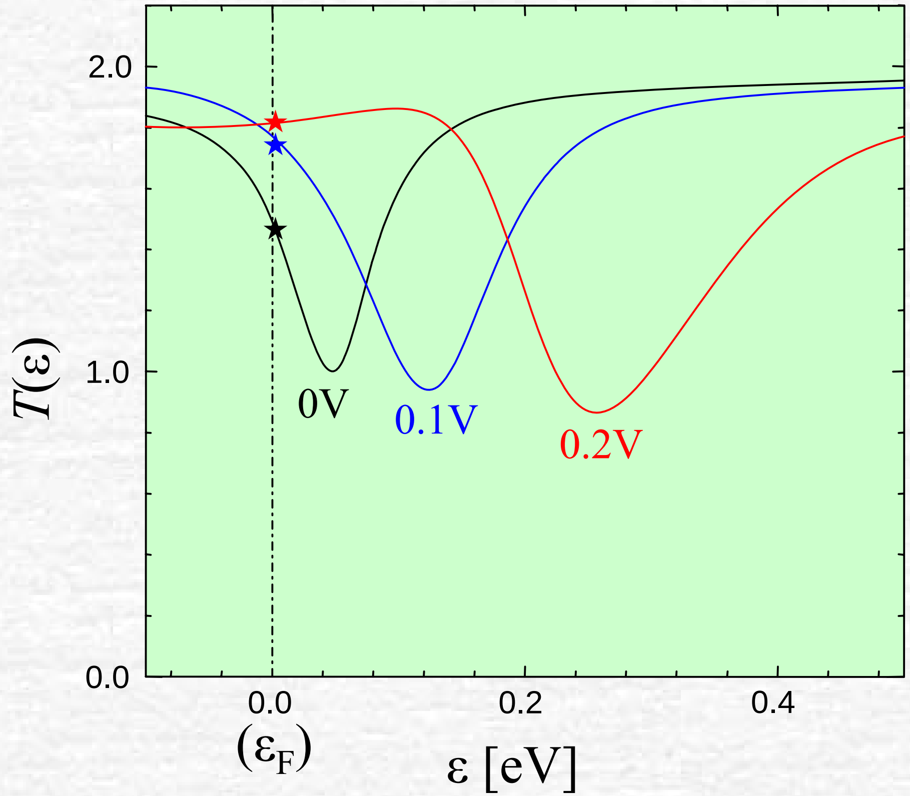
Annealing



H.J. Choi, *et al.*, PRL, **84**, 2917 (2000)

Improvement of Reduced Electron-Derived Thermal Conductance using *Gate Voltage*





The transmission dip can be removed by applying gate voltages.

Summary

We study the thermal transport in carbon nanotubes, especially focusing on influences of vacancy defect on thermal transport in CNTs.

- "*Phonon*"-derived thermal conductance is
  - reduced by phonon scattering with the localized phonon state around vacancy.
  - repaired by rearrangement of the vacancy to the 5-1db defect.
- "*Electron*"-derived thermal conductance is
  - reduced by electron scattering with σ dangling-bond state.
  - repaired by tuning a gate voltage in addition to the annealing.