Phonon Engineering: From Nanowires and Quantum Dots to Graphene and Topological Insulators

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Nano-Device Laboratory (NDL)
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Profile: experimental and theoretical research in phonon engineering and nanodevices

PI: Alexander A. Balandin
Thermal and Electrical Characterization

Research & Applications

Electronic Devices and Circuits

Direct Energy Conversion

Optoelectronics

Bio-Nanotech

Raman, Fluorescence and PL Spectroscopy

Theory and Modeling

Nanoscale Characterization

Device Design and Characterization

Research at NDL was funded by NSF, ONR, SRC, DARPA, NASA, ARO, AFOSR, CRDF, as well as industry, including IBM, Raytheon, TRW, etc.

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Outline

- Phonon Engineering Background
  - Definition and motivations
- Phonon Engineering for Mobility Enhancement
  - Acoustically mismatched nanostructures
- Phonon Engineering for Thermal Management
  - Tuning thermal conductivity at nanoscale
  - 2D phonon transport in graphene
- Nanometrology with Phonons
  - Viruses as nanotemplates
  - Graphene and topological insulators
Can We Engineer Phonons?

### Excitations in Solids

<table>
<thead>
<tr>
<th>Excitation</th>
<th>Field</th>
</tr>
</thead>
<tbody>
<tr>
<td>photon</td>
<td>EM wave</td>
</tr>
<tr>
<td>phonon</td>
<td>elastic wave</td>
</tr>
<tr>
<td>magnon</td>
<td>magnetization wave</td>
</tr>
<tr>
<td>exciton</td>
<td>polarization wave</td>
</tr>
<tr>
<td>polaron</td>
<td>“electron + phonon”</td>
</tr>
<tr>
<td>exciton polariton</td>
<td>“EM + exciton”</td>
</tr>
</tbody>
</table>

### Criteria

- we should obtain useful changes in properties of materials
- we should have means of controlling the elementary excitations

**Excitation Parameters**

<table>
<thead>
<tr>
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</tr>
</tbody>
</table>

**Bulk Limit**

\[
E = \frac{\hbar^2 k^2}{2m} \quad \omega = \frac{c}{\sqrt{\varepsilon}} k \quad \omega = c_{\text{L,T}} k
\]
Going Beyond “Classical” Size Effects:
From Phonon –Boundary Scattering to Phonon Spectrum Engineering

“Classical” size effects on heat conduction: phonon – boundary scattering
Casimir (1938), Berman (1955), Ziman (1960)

Phonon – boundary scattering rate:
\[
\frac{1}{\tau_B} = \zeta \frac{1 - p}{1 + p} \frac{<\nu>}{L}
\]

Significant decrease of the lattice thermal conductivity due to phonon confinement in a free-standing semiconductor quantum well

Alexander Balandin and Kang L. Wang
Device Research Laboratory, Electrical Engineering Department, University of California – Los Angeles, Los Angeles, California 90095-1594
(Received 17 February 1998; revised manuscript received 20 April 1998)

Tuning thermal conductivity... can be approached with appropriate modification of phonon modes, e.g., phonon engineering.
Phonon Engineering at Nanoscale

Analogy with the Electron Band-Structure Engineering

**Definition:** phonon engineering is an approach for modifying the thermal, electrical and optical properties of materials/devices via tuning the phonon characteristics at nanometer scale through the spatial confinement-induced changes in the phonon spectrum.

**Goals:**
- Change in electron – phonon scattering \( \rightarrow \) drift mobility
- Change in phonon group velocity \( \rightarrow \) thermal conductivity
- Control of the phonon energies \( \rightarrow \) optical response

**Tuning Parameters:**
- Crystalline structure
- Dimensions
- Sound velocity
- Mass density
- Acoustic Impedance
- Interface
- Optical phonon frequencies

**Band-gap engineering:** mismatch of \( E_G \)

**Phonon engineering:** mismatch of \( Z = \rho V_{sound} \)

Bulk vs. Confined Acoustic Phonons


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Part I: Phonon Engineering for Mobility Enhancement

Evolution of Phonon Transport in Nanowires with “Acoustically Hard” Barriers

Observations:

The size of the circles is proportional to the average divergence of displacement vector.

Contribution of higher energy modes is negligible compared to the shown modes.

“True” acoustic mode changes velocity from that in Si to the diamond.

Contribution of the “true” acoustic mode to scattering decreases with increasing coating thickness.

Phonon Engineering of Electron Mobility in Silicon Nanowires

Observations:

→ at low T the mobility is limited by impurities and is proportional to $T^{1/2}$, while at high T it is limited by phonons and is proportional to $T^{-1/2}$

→ electron mobility for diamond coated Si nanowire is between the limits corresponding to free-standing and clamped nanowire

→ electron – phonon scattering in free-standing nanowires is so strong that it limits mobility even at low T

Design and Fabrication of Silicon Nanowire FETs with the Phonon-Engineered Mobility

Design of the Nanowire Field-Effect Transistor: NW-FET

Materials Issues
→ Si nanowire has to be crystalline
→ Nanometer radius
→ “Acoustically hard material”

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Experimental Demonstration of the Electron Mobility Enhancement in Silicon NW FETs

Results
- Mobility data is extracted from the IV and CV measurements and model calculations
- Preliminary data indicate electron mobility enhancement in Si NW FETs with the “acoustically hard” coating of Al₂O₃ compared to reference NW FETs
Part II: Phonon Engineering for Thermal Management

Artistic rendering of graphene lateral heat spreaders for advanced chips from a recent IEEE Spectrum feature article *Chill Out: New Materials and Designs Can Keep Chips Cool* by A.A. Balandin.
Thermal Conductivity and Heat Transport Basics

**Definitions and Basic Theory**

The ratio of the heat flow per unit area $S$ to the temperature gradient is defined as the thermal conductivity (Fourier’s law):

$$\frac{\dot{Q}}{S} = -K \nabla T$$

**RT thermal conductivity for important materials:**

- **Silicon (Si):** 145 W/mK
- **SiO$_2$:** 1-13 W/mK
- **Copper:** 400 W/mK

**RT thermal conductivity for carbon materials:**

- **Diamond:** 1000 – 2200 W/mK
- **Graphite:** 200 – 2000 W/mK (orientation)
- **DLC:** 0.1 – 10 W/mK
- **CNTs:** 3000 – 3500 W/mK

Heat current carried by phonons is a sum of contributions from all normal modes:

$$Q = \sum_{q,j} N_{q,i}(\mathbf{q}) \hbar \omega_i(\mathbf{q}) \frac{\partial \omega}{\partial \mathbf{q}}$$

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Phonon Thermal Transport at Nanoscale in Crystalline Semiconductors

Thermal conductivity usually decreases as one goes from bulk material to nanostructure or thin film

 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: $K = 13 \text{ W/mK}$

$\rightarrow$ Phonon - boundary scattering:

$$\frac{1}{\tau_B} = \zeta \frac{1 - p <v>}{1 + p L}$$

$\rightarrow$ Phonon spectrum modification

$\rightarrow$ Phonon group velocity changes

$\rightarrow$ Phonon density of states change

$\rightarrow$ Changes for electrons
Tuning Thermal Conduction in Quantum Dot Superlattices

Measured and Calculated Thermal Conductivity

Transition to the Bulk Limit

Bulk limit: $t \to$ very large or $d \to$ very large


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Phonon Engineering of Thermal Conductivity of QDS for ZT Enhancement

Partially Ordered Si/Ge Quantum Dot Superlattices

Figure of Merit:

\[ ZT = \frac{\alpha^2 \sigma T}{\kappa} \]

\( \alpha \) - Seebeck coefficient
\( \sigma \) – electrical conductivity
\( \kappa \) – thermal conductivity
\( T \) – absolute temperature
\( Z \) – figure of merit

Thermal Conductivity of Carbon Materials and Nanostructures

Dependence of thermal conductivity of carbon materials on film thickness and atomic structure (sp² vs sp³ phase; H content)

Collaboration with A.C. Ferrari Group (Cambridge, U.K.)


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Divergence of the Lattice Thermal Conductivity in 2-D Crystal Lattices

K \sim \log(N) \text{ in 2D}

K \sim N^\alpha \text{ in 1D, } \alpha \neq 1

N - system size

→ Consensus: The intrinsic thermal conductivity of 2-D or 1-D anharmonic crystals is anomalous.

Klemens Theory of the Phonon
Thermal Conductivity of Bulk Graphite

\[ Q = \sum_{q,j} N_{q,j}(q) \hbar \omega_j(q) \frac{\partial \omega_j}{\partial q}, \]

Intrinsic Umklapp-Limited Thermal Conductivity:

\[ K = \frac{1}{4\pi k_B T^2} \hbar \sum_{q = TA, LA} q_{\text{max}} \int \left\{ (\hbar \omega_j(q) \frac{d\omega_j(q)}{dq})^2 \tau_{U,s} \frac{\exp[\hbar \omega_j(q)/kT]}{[\exp[\hbar \omega_j(q)/kT] - 1]^2} \right\} dq \]

Umklapp life-time, which defines MFP:

\[ \tau_{U,s} = \frac{1}{\gamma_s^2} \frac{M \nu_s^2}{k_B T} \frac{\omega_{s,\text{max}}}{\omega^2} \]

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

3-D: \( C(\omega) \sim \omega^2 \)

In bulk graphite there is natural low-bound cut-off at \(~4\) THz. The heat transport is two-dimensional only above this frequency. Klemens’s value for graphite: \( K = 1900 \) W/mK


Phonon Dispersion and Gruneisen Parameter in Graphene

Valence Force Field (VFF) calculation of the phonon dispersion in graphene

The mode-dependent Gruneisen parameters are measure of sensitivity of the phonon frequencies to changes in the system volume.

\[ \gamma_{\lambda,k} = -\frac{\partial \ln (\omega_{\lambda,k})}{\partial \ln V} \]


Klemens Theory of the Phonon Heat Conduction in Graphene

Thermal conductivity in graphene:

\[
K = \frac{1}{4\pi k_B T^2} \sum_{s=TA,LA}^{q_{\text{max}}} \int_{q_{\text{min}}}^{q_{\text{max}}} \left\{ (\hbar \omega_s(q) \frac{d\omega_s(q)}{dq})^2 \tau_{U,s} \frac{\exp[\hbar \omega_s(q)/kT]}{[\exp[\hbar \omega_s(q)/kT] - 1]^2} \right\} dq
\]


Graphene:

MFP = \( L \) – physical size of the system

\( \tau_{U,s} = \frac{1}{\gamma_s^2} \frac{M \nu_s^2}{k_B T} \frac{\omega_{s,\text{max}}}{\omega^2} \)

Limiting low-bound frequency:

\( \omega_{s,\text{min}} = \frac{\nu_s}{\gamma_s} \sqrt{\frac{M \nu_s}{k_B T L} \frac{\omega_{s,\text{max}}}{L}} \)

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Klemens Approximation for Graphene

"Intrinsic" Thermal Conductivity Value is Defined by the Flake Size

The phonon transport in graphene is 2D all the way down to zero frequency

Low-bound cut-off frequency is defined by the condition that the phonon MFP can not exceed the physical size of the graphene flake:

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

UCR Experiment: Heating Up Graphene

IEEE Spectrum illustration of the first measurements of thermal conductivity of graphene carried out at UCR.

See details in A.A. Balandin et al., Nano Letters, 8, 902 (2008).

SEM image of the suspended graphene flake connected to heat sinks.
Raman Spectroscopy of Graphene

Phonons for Raman-based Nanometrology of Graphene

Optical visualization on Si/SiO₂ substrates

AFM inspection

Low-T transport measurements

D band: A₁g (~1350 cm⁻¹); G peak: E₂g; 2D band

Graphene Temperature Coefficients: Raman Spectrometer as “Thermometer”

Note: the sign is negative

Temperature is controlled externally; very low excitation power on the sample surface is used (< 0.5 – 1 mW).

Phonon frequency downshift with T is unusual when the bond-bond distances shorten with T since normally lattice contraction leads to the upward shift of the frequencies.

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Measurement of the Thermal Conductivity

Laser acts as a heater (confirmed by Stokes–Anti-Stokes intensity ratio): $\Delta P_G$

Raman “thermometer”: $\Delta T_G = \Delta \omega / \chi_G$

Thermal conductivity: $K = (L / 2a_G W)(\Delta P_G / \Delta T_G)$

Thermal conductivity of rectangular flake ($L$ is the half-length):

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

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


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Few-Layer Suspended Graphene Measurements

→ Massive metal heat sinks
→ Data extraction for actual shape of the
→ Monitoring SiO$_2$ temperature: $W_2$ and $W_3$ Si-O-Si stretching bond position (800 – 1100 cm$^{-1}$)
→ Method verification at macro-scale

→ Collaboration with C.N. Lau group (UCR)
→ Free-standing samples; A. Geim group (UM)
Modeling Based Data Extraction for Arbitrary Shaped Graphene Flakes

Investigation of heat conduction in multi-layer graphene led to the use of the irregular shaped graphene flakes due to the difficulty of the sample preparation.

The excitation source follows Gaussian distribution:

\[ Q = P(x, y) = P_0 \exp\left(-\frac{x^2 + y^2}{2\sigma^2}\right) \]

FWHM occurs at 0.5 \( \mu \text{m} \)

Finite-element solution of the heat diffusion through the graphene flake was obtained in order to take into account the actual shape of the flake.

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Thermal Conductivity of Single Layer Graphene: Comparison with Carbon Nanotubes

Table I: Experimental RT Thermal Conductivity of Graphene and CNTs

<table>
<thead>
<tr>
<th>Sample</th>
<th>( K ) (W/mK)</th>
<th>Method</th>
<th>Comments</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>~2800</td>
<td></td>
<td>suspended large-area</td>
<td></td>
</tr>
<tr>
<td></td>
<td>~1500</td>
<td></td>
<td></td>
<td>Ghosh et al., Nature Mat. (2010)</td>
</tr>
<tr>
<td>BLG</td>
<td>2800</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>FLG (n=4)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SW-CNT</td>
<td>~3500</td>
<td>electrical</td>
<td>individual</td>
<td>Pop et al., Nano Lett. (2006)</td>
</tr>
<tr>
<td>SW-CNT</td>
<td>3000 - 7000</td>
<td>thermocouples</td>
<td>individual</td>
<td>Yu et al., Nano Lett. (2005)</td>
</tr>
</tbody>
</table>

Table II: Theoretical RT Thermal Conductivity of Graphene and CNTs

<table>
<thead>
<tr>
<th>Sample</th>
<th>( K ) (W/mK)</th>
<th>Method</th>
<th>Comments</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>CNTs</td>
<td>~6600</td>
<td>MD</td>
<td>predicted higher ( K ) for graphene</td>
<td>Berber et al. PRL (2000)</td>
</tr>
<tr>
<td>CNTs</td>
<td>~2980</td>
<td>MD</td>
<td>strong defect dependence</td>
<td>Che et al., Nanotech. (2000)</td>
</tr>
<tr>
<td>graphite</td>
<td>~2000</td>
<td>C-K</td>
<td>basal plane (a-plane)</td>
<td>Klemens et al., Carbon (1994)</td>
</tr>
</tbody>
</table>
Phonon Transport in Graphene: Accurate Theory vs Experiment

- **4-fold enhancement** of Umklapp scatterings in bilayer in comparison with monolayer due to the two-fold degeneracy of phonon energy spectra in bilayer

- **Dimensional crossover** between 2D and 3D systems


http://www.nature.com/nmat/journal/vaop/ncurrent/full/nmat2753.html
Enhancement of Umklapp Scattering in Bilayer Graphene

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Thermal Management Applications: Engineering Phonon Heat Fluxes

High thermal conductivity materials can be used as lateral hot-spot spreaders or thermal interface materials (TIM)

Issues:
- Compatibility with Si CMOS technology
- Electrical insulator vs conductor
- Bulk vs nanostructure
- Anisotropy
- Coefficient of thermal expansion
- Temperature stability

Theoretical Suggestions:
- Graphene should have very high thermal conductivity; flat geometry is a major benefit

Extra Benefits:
- Graphene and CNTs can become foundations of the carbon or hybrid Si-carbon electronics

Table: Room-temperature thermal conductivity of best heat conductors

<table>
<thead>
<tr>
<th>Sample</th>
<th>K (W/mK)</th>
<th>Method</th>
<th>Comments</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>diamond</td>
<td>~ 1000 – 2200</td>
<td>3-omega; other</td>
<td>bulk</td>
<td>Berman et al.</td>
</tr>
<tr>
<td>MW-CNT</td>
<td>&gt; 3000</td>
<td>electrical</td>
<td>individual</td>
<td>Kim et al.</td>
</tr>
<tr>
<td>SW-CNT</td>
<td>~ 3500</td>
<td>electrical</td>
<td>individual</td>
<td>Pop et al.</td>
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<td>SW-CNT</td>
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<td>thermocouples</td>
<td>bundles</td>
<td>Hone et al.</td>
</tr>
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</table>
Graphene Heat Spreaders Designs: Interconnect Applications


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Part III: Nanometrology with Phonons

← Growth (literally) of tobacco mosaic viruses for applications as nanotemplates
Viruses as Nanotemplates for Large-Scale Self-Assembly of Nanodevices


Nanofabrication Benefits:

- suitable dimensions
- small size dispersion
- selective attachment
- robust

SEM of a pure TMV and TMV end-to-end assembly (left); nanowire "interconnect" made of metal coated TMV assembly (right).

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Nanostructure “Growth”, Self-Assembly and Characterization

Nanostructure Growth:

TEM micrograph of the pure TMV and metal coated TMV. Scale bar is 50 nm.

X-Ray Characterization

Phonons for Nanometrology of Bio-Inorganic Nanostructures and Self-Assembly Process

Raman spectra of TMV, Pt coated TMV and Au coated TMV: the Amide I line at 1655 cm\(^{-1}\), C-H deformation lines at 1454.5 cm\(^{-1}\) and 1332 cm\(^{-1}\), and the phenylalanine residue line at 1005 cm\(^{-1}\). The Amide I lines of TMV-Pt and TMV-Au are at 1664 cm\(^{-1}\) and 1672 cm\(^{-1}\) respectively.

Amide I line is related to TMV coat protein capsid, the line shift indicates the change of vibrational modes due to the binding of metal with certain functional group in the shell protein.

“Graphene-Like” Exfoliation of Quasi 2D Crystals of Different Materials

Quintuple thickness: 
~1 nm

Identification: AFM, SEM, TEM

Quasi-2D Crystals of Bi$_2$Te$_3$ Family as Thermoelectrics and Topological Insulators

→ Resistivity is $\sim 10^{-4}$ $\Omega$m, which is near ZT optimum of $\sim 10^{-5}$ $\Omega$m


Raman Nanometrology of Graphene: Counting the Number of Atomic Planes

Deconvolution of 2D (G') Band

Graphene @ 300K
\( \lambda_{\text{exc}} = 488 \text{ nm} \)

1 layer
2 layers
3 layers
4 layers
5 layers

I. Calizo, F. Miao, W. Bao, C.N. Lau and A.A. Balandin

Raman Spectroscopy of Quasi 2-D Bismuth Telluride Crystals

Major Issue: overheating and melting under low power excitation

The use of the power levels above 0.5 mW (corresponds to ~0.22 mW on the sample) results in the holes due to local melting of the atomically-thin flakes

Low thermal conductivity of Bi-Te quintuples is good news for thermoelectric applications but bad news for characterization

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Raman Spectra of Suspended Bi-Te Atomic Films

The Raman phonon peaks were identified as $A_{1g}$ at $\sim 62$ cm$^{-1}$, $E_{g2}$ at $\sim 104$ cm$^{-1}$, $A_{1u}$ at $\sim 120$ cm$^{-1}$ and $A_{2g}$ at $\sim 137$ cm$^{-1}$.

Possible enhancement of out-of-plane modes in suspended atomically-thin samples

Crystalline or poly with large grains

The out-of-plane vibrations (at $\sim 137$ cm$^{-1}$ and $\sim 119$ cm$^{-1}$) in the suspended Bi-Te atomic films have higher intensity.
Phonon Nanometrology of the Atomically Thin Films of Bi-Te Topological Insulators


TABLE I. Raman peaks in Bi2Te3 crystals and few-quintuple films.

<table>
<thead>
<tr>
<th></th>
<th>E^1_g</th>
<th>A^1_g</th>
<th>E^2_g</th>
<th>A^1_a</th>
<th>A^2_1g</th>
<th>I(A^2_1g)/I(E^2_g)</th>
<th>I(A^1_u)/I(E^2_g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk</td>
<td>34.4</td>
<td>62.1</td>
<td>101.7</td>
<td>⋯</td>
<td>134.0</td>
<td>0.75</td>
<td>⋯</td>
</tr>
<tr>
<td>82 nm</td>
<td>35.8</td>
<td>61.5</td>
<td>101.9</td>
<td>116.9</td>
<td>132.7</td>
<td>0.83</td>
<td>0.62</td>
</tr>
<tr>
<td>40 nm</td>
<td>38.9</td>
<td>61.3</td>
<td>101.3</td>
<td>116.2</td>
<td>133.0</td>
<td>0.92</td>
<td>0.76</td>
</tr>
<tr>
<td>4 nm</td>
<td>38.9</td>
<td>60.9</td>
<td>101.4</td>
<td>116.7</td>
<td>132.9</td>
<td>1.30</td>
<td>0.80</td>
</tr>
<tr>
<td>Bulk</td>
<td>36.5</td>
<td>62.0</td>
<td>102.3</td>
<td>⋯</td>
<td>134.0</td>
<td>⋯</td>
<td>⋯</td>
</tr>
<tr>
<td>Bulk</td>
<td>⋯</td>
<td>62.3</td>
<td>103.7</td>
<td>⋯</td>
<td>134.2</td>
<td>⋯</td>
<td>⋯</td>
</tr>
</tbody>
</table>
Conclusions and Outlook

- Phonon engineering can be used for improvement of the charge carrier mobility
  - acoustically hard barriers work at room temperature
- Phonon engineering can be used for strong suppression of thermal conductivity in nanostructured materials
  - thermoelectric applications
- “Phonon optimization” of conventional technology:
  - thermal constrains early at the device-design stage
  - material selection for the mobility enhancement: few-layer graphene
- Technological challenges:
  - interface quality
  - few-nanometer scale requirement for RT effects
  - heterostructures of dissimilar materials
- Phonons for nanoscale characterization
  - hybrid bio-inorganic nanostructures and self-assembly processes
  - graphene materials and devices
  - topological insulators
Acknowledgements

Nano-Device Laboratory (NDL) Group, UC - Riverside

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