Boltzmann Transport Equations for Nanoscience Applications

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Overview

- We want to understand electrical and thermal transport in nanoscale systems
- Simulate transport in nanotubes, nanoribbons, nanowires, etc.
  - Why BTE?
  - Derivation of the BTE
  - Classical vs. Quantum
  - Carbon Nanotubes
  - Simulation of 1D systems

Why Boltzmann Transport Eqn. (BTE)?

- Originally derived for a dilute gas of non-interacting particles
- Extended to the simulation of electron and phonon transport
- Particle motion treated classically as in the Liouville equation
- Particle interactions introduced through quantum-mechanical perturbation theory
- Very flexible, general, and powerful
- Can include many other important effects:
  - electron bandstructure
  - phonon dispersion
  - self-consistency (Poisson equation)
  - Electro-thermal transport

Distribution function

- Distribution function $f_T(r,k,t)$ represents the probability for a particle to occupy position **r** with momentum **k** at time t.
- Distribution function f_T contains all the information about the transport in the system.
- From f_T we can obtain average quantities like current, mobility, mean-free-path, etc.
- It is 7-D in general: 3-D spatial (**r**) + 3-D momentum (**k**) + time (t) dependence.
- In 1-D materials like CNTs and nanowires, space and momentum are 1-D, so f_T is 3-D altogether.

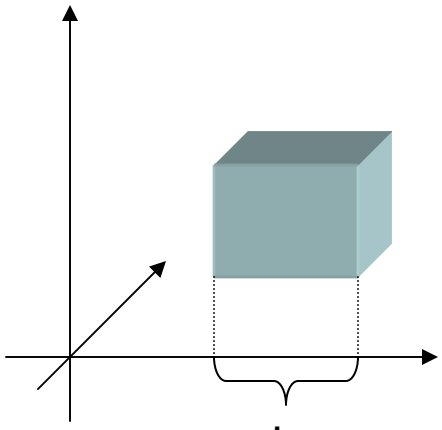
Semi-classical vs. Quantum

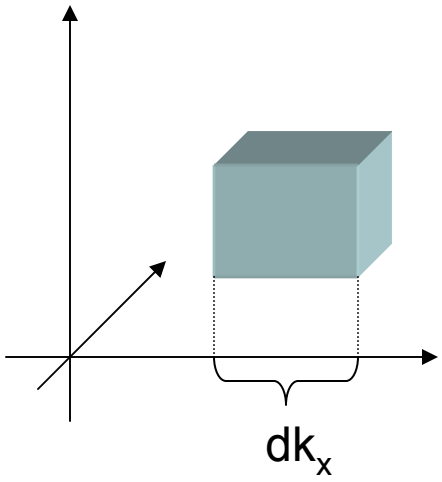
- Semi-classical BTE treats particles as classical point particles
  - Includes scattering through Fermi's Golden Rule
  - Assumes collisions are instantaneous
  - Position and momentum are independent and functions of time
- Quantum BTE is capable of including quantum transport effects
  - quasi-particle states
  - level shift and broadening
  - requires a straightforward modification to the scattering rates
- Wigner equation takes this another step further to include the effects of confining potentials
  - Add higher derivatives (3^{rd}, 5^{th}, etc.) of the potential and distribution

Particles change state by 3 different mechanisms:

1. Motion in real space due to electron velocity
2. Acceleration in momentum due to electric field
3. Scattering due to phonons

Consider a small cube in combined x and k space:





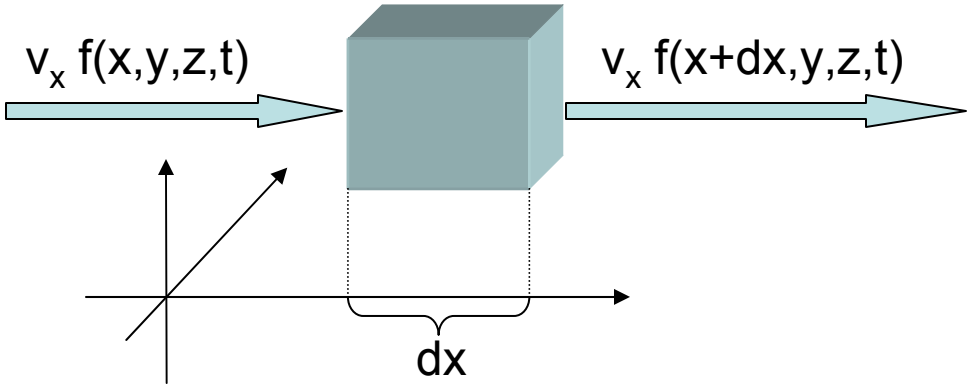
Particles change state by 3 different mechanisms:

1. Motion in real space due to electron velocity

The net particle gain is the difference at the two faces times the velocity in the x direction:

$$\frac{df(x,y,z,t)}{dt} = v_x \left[f(x,y,z,t) - f(x+dx,y,z,t) \right] dx$$

In the limit of small dx this becomes: $\frac{df(x,y,z,t)}{dt} = -v_x \frac{\partial f(x,y,z,t)}{\partial x}$



Particles change state by 3 different mechanisms:

1. Motion in 3D space:
   - In general 3-D space, when there is a spatial gradient to the electron distribution, electrons will travel from a region of higher density to region of lower density.
   - The gradient of the distribution points in the direction of greatest change, therefore direction of electron motion.
   - Therefore the rate of change of the distribution function (scalar!) is equal to the electron velocity (a vector!) dotted with the gradient (another vector!):

$$\frac{\partial f_T(\mathbf{r},\mathbf{k},t)}{\partial t} = v(\mathbf{r}) \cdot \nabla_{\mathbf{r}} f_T(\mathbf{r},\mathbf{k},t) = \frac{d\mathbf{r}}{dt} \cdot \nabla_{\mathbf{r}} f_T(\mathbf{r},\mathbf{k},t)$$

Particles change state by 3 different mechanisms:

1. Motion in real space:
   - Particle velocity is the time derivative of its position

$$v(\mathbf{k}) = \frac{d\mathbf{r}(t)}{dt}$$

   - Velocity can be obtained from the bandstructure or dispersion

$$v(\mathbf{k}) = \frac{1}{\hbar}\nabla_{\mathbf{k}} E(\mathbf{k},\mu) \quad , \quad v(\mathbf{q}) = \nabla_{\mathbf{q}}\omega(\mathbf{q},\mu)$$

   - Putting these together produces

$$\frac{\partial f_T(\mathbf{r},\mathbf{k},t)}{\partial t} = -\frac{1}{\hbar}\nabla_{\mathbf{k}} E(\mathbf{k},\mu)\cdot\nabla_{\mathbf{r}} f_T(\mathbf{r},\mathbf{k},t)$$

Particles change state by 3 different mechanisms:

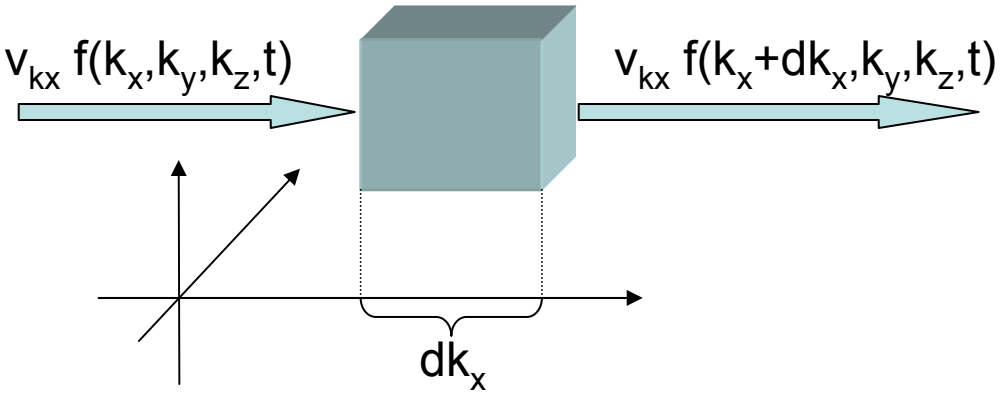
2. Acceleration in momentum due to electric field

Again consider a small cube in k-space, and look at k_x direction.

The net gain is the difference at the two faces times the velocity in the k_x direction:

$$\frac{df(k_x,k_y,k_z,t)}{dt} = v_{kx}\left[f(k_x,k_y,k_z,t) - f(k_x+dk_x,k_y,k_z,t)\right]dk_x$$

In the limit of small dk_x this becomes: $$\frac{df(k_x,k_y,k_z,t)}{dt} = \frac{dk_x}{dt}\frac{\partial f(k_x,k_y,k_z,t)}{\partial k_x}$$



Particles change state by 3 different mechanisms:

2. Acceleration under the force of the electric field:

- When an electric field **E** is applied to an electron, it produces an accelerating force **F**= –e**E** on the electron.
- Magnetic field can also be added **F**=-e(**E**+**v***x***B**)
- Analogous to F= ma = m*dv/dt = d**p**/dt, we have:

$$\frac{d\mathbf{k}}{dt} = \frac{1}{\hbar}\mathbf{F} = -\frac{e\mathbf{E}}{\hbar}$$

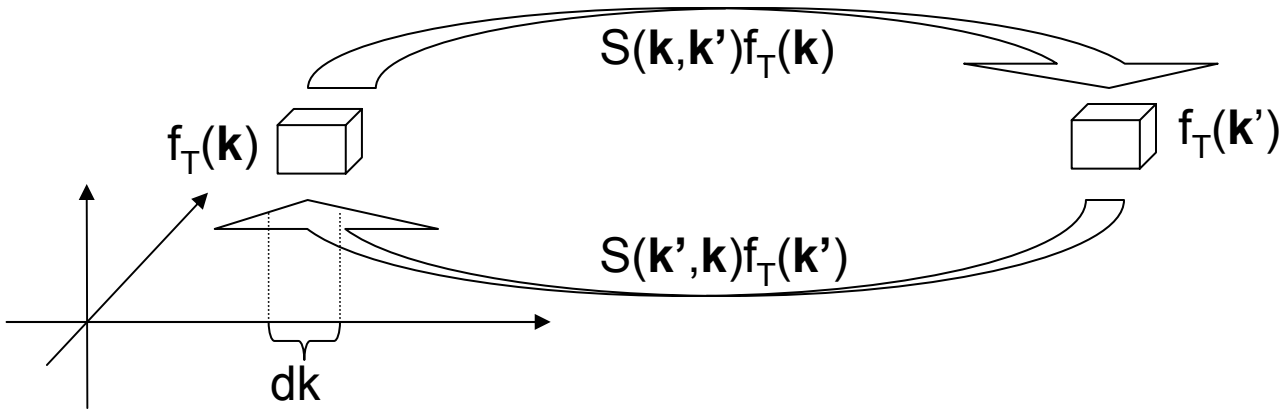
- Therefore the rate of change of the distribution function (scalar!) is equal to the applied force **F** (a vector!) dotted with the gradient in momentum (another vector!):

$$\frac{\partial f_T(\mathbf{r},\mathbf{k},t)}{\partial t} = -\frac{e\mathbf{E}}{\hbar}\cdot\nabla_{\mathbf{k}} f_T(\mathbf{r},\mathbf{k},t)$$

Electrons change state by 3 different mechanisms:

3. Scattering in and out of a momentum state:

- Can be derived by examining a small differential element in momentum space
- Particles occupying a state **k** with probability $f_T(\mathbf{k})$ can scatter out of **k** with transition probability $S(\mathbf{k},\mathbf{k}')$
- Particles occupying a state **k'** with probability $f_T(\mathbf{k}')$ can scatter into state **k** with transition probability $S(\mathbf{k}',\mathbf{k})$

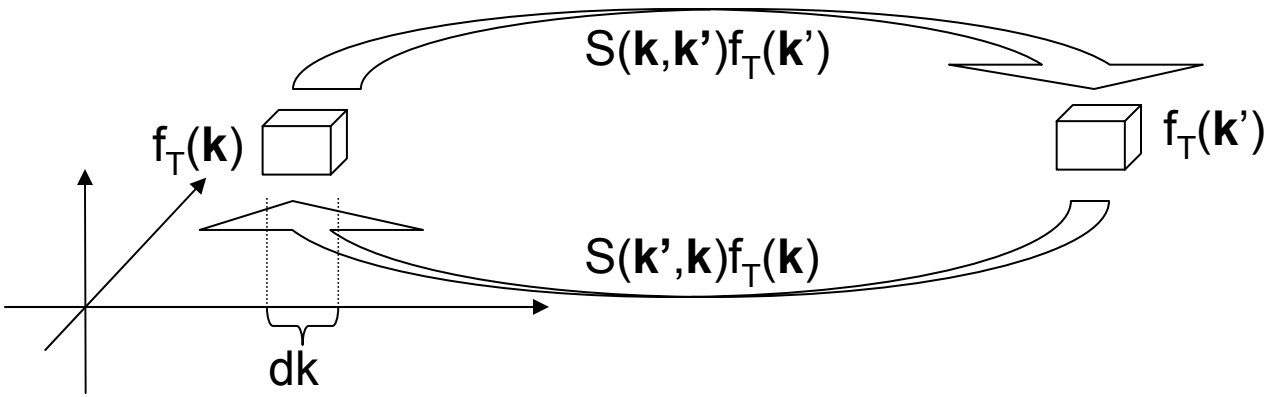


Electrons change state by 3 different mechanisms:

3. Scattering in and out of a momentum state:

- Every scattering into **k** increases the occupancy $f_T(\mathbf{k})$
- Every scattering out of **k** decreases $f_T(\mathbf{k})$
- The net change in occupancy $f_T(\mathbf{k})$ is the in-scattering minus the out-scattering
- For each state **k**, add up contributions from all other states **k'**

$$\frac{\partial f_T(\mathbf{k})}{\partial t} = \sum_{\mathbf{k}\prime} [S(\mathbf{k}\prime, \mathbf{k}) f_T(\mathbf{k}\prime) - S(\mathbf{k}, \mathbf{k}\prime) f_T(\mathbf{k})]$$



Degeneracy and exclusion

- Pauli's Exclusion Principle tells us that only one electron can occupy a given state at a given time (ignoring spin).
- Because of exclusion, an electron can scatter into a state only if it is empty.
- To account for exclusion, we multiply the transition rate by the probability that the state is not occupied, given by (1-$f_T(\mathbf{k})$).
- Finally we add all the contributions by summing over all the possible final states **k**'

$$\frac{\partial f_T(\mathbf{k})}{\partial t} = \sum_{\mathbf{k}\prime} [S(\mathbf{k}\prime, \mathbf{k}) f_T(\mathbf{k}\prime)(1 - f_T(\mathbf{k})) - S(\mathbf{k}, \mathbf{k}\prime) f_T(\mathbf{k})(1 - f_T(\mathbf{k}\prime))]$$

- This form referred to as "degenerate statistics"

Boltzman Transport Eqn. (BTE)

- Particles are conserved so rate of change in time has to equal the change due to scattering
- Therefore we simply equate the two rates to obtain the BTE :

$$\frac{df_T(\mathbf{r},\mathbf{k},t)}{dt} = \left(\frac{df_T(\mathbf{r},\mathbf{k},t)}{dt}\right)_{scat.}$$

- The sum can be converted to an integral in the limit of small dk.
- This makes the BTE a difficult integro-differential equation.

$$\frac{\partial f_T(\mathbf{r},\mathbf{k},t)}{\partial t} - \frac{e\mathbf{E}(\mathbf{r})}{\hbar}\cdot\nabla_{\mathbf{k}} f_T(\mathbf{r},\mathbf{k},t) + \frac{1}{\hbar}\nabla_{\mathbf{k}} E(\mathbf{k},\mu)\cdot\nabla_{\mathbf{r}} f_T(\mathbf{r},\mathbf{k},t) =$$
$$\frac{\Omega}{(2\pi)^3}\int d^3\mathbf{k}\prime\,[S(\mathbf{k}\prime,\mathbf{k})f_T(\mathbf{k}\prime)(1-f_T(\mathbf{k})) - S(\mathbf{k},\mathbf{k}\prime)f_T(\mathbf{k})(1-f_T(\mathbf{k}\prime))]$$

"Shorthand" BTE

- The BTE can be derived quickly by starting with the semiclassical assumption and applying the chain rule
- Start by noting the distribution function f_T is a function of position **r**, momentum **k**, and time t
- Assume **r**(t) and **k**(t) are independent and only functions of time
- REMINDER: Chain rule in 1-D and n-dimensions

$$\frac{df(g(t))}{dt} = \frac{\partial f(g)}{\partial g}\frac{dg(t)}{dt} \qquad \frac{df(\mathbf{g}(t))}{dt} = \nabla_{\mathbf{g}} f(\mathbf{g}) \cdot \frac{d\mathbf{g}(t)}{dt}$$

- Apply the chain rule to obtain the complete time derivative:

$$\frac{df_T(\mathbf{r},\mathbf{k},t)}{dt} = \frac{\partial f_T(\mathbf{r},\mathbf{k},t)}{\partial t} + \nabla_{\mathbf{r}} f_T(\mathbf{r},\mathbf{k},t) \cdot \frac{d\mathbf{r}}{dt} + \nabla_{\mathbf{k}} f_T(\mathbf{r},\mathbf{k},t) \cdot \frac{d\mathbf{k}}{dt}$$

Interpreting the BTE:

The BTE is saying that probability is conserved along the path of the particle

- Use Taylor expansion in phase space:

$$f_T(\mathbf{r}+d\mathbf{r},\mathbf{k}+d\mathbf{k},t+dt) =$$
$$f_T(\mathbf{r},\mathbf{k},t)+\frac{\partial f_T(\mathbf{r},\mathbf{k},t)}{\partial t}dt+\nabla_{\mathbf{r}}f_T(\mathbf{r},\mathbf{k},t)\cdot d\mathbf{r}+\nabla_{\mathbf{k}}f_T(\mathbf{r},\mathbf{k},t)\cdot d\mathbf{k}$$

- Factor out the “dt” term and group together:

$$f_T(\mathbf{r}+d\mathbf{r},\mathbf{k}+d\mathbf{k},t+dt) =$$
$$f_T(\mathbf{r},\mathbf{k},t)+dt\left(\frac{\partial f_T(\mathbf{r},\mathbf{k},t)}{\partial t}+\nabla_{\mathbf{r}}f_T(\mathbf{r},\mathbf{k},t)\cdot\frac{d\mathbf{r}}{dt}+\nabla_{\mathbf{k}}f_T(\mathbf{r},\mathbf{k},t)\cdot\frac{d\mathbf{k}}{dt}\right)$$

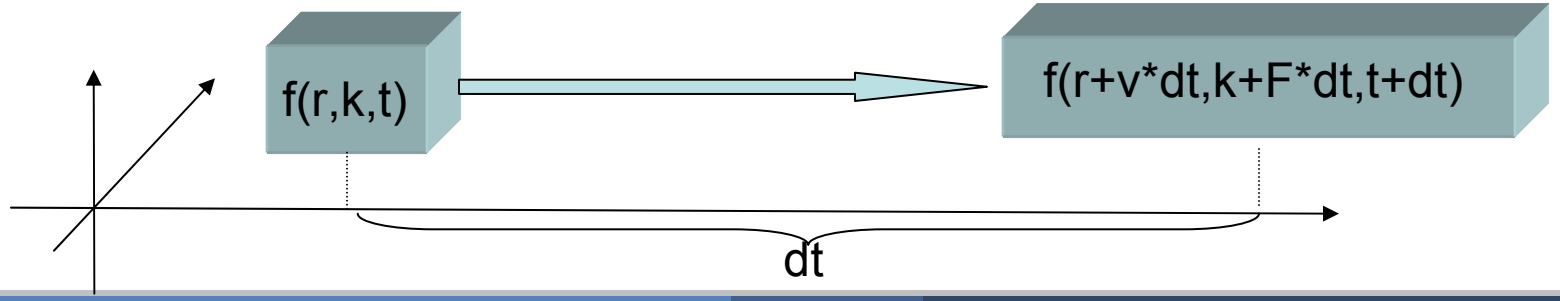
Interpreting the BTE:

The BTE is saying that probability is conserved along the path of the particle

- Recognize the expression for total time derivative
- Substitute in the conservation equation:

$$f_T(\mathbf{r}+d\mathbf{r},\mathbf{k}+d\mathbf{k},t+dt) =$$

$$f_T(\mathbf{r},\mathbf{k},t) + dt\left(\frac{f_T(\mathbf{r},\mathbf{k},t)}{dt}\right) = f_T(\mathbf{r},\mathbf{k},t) + dt\left(\frac{f_T(\mathbf{r},\mathbf{k},t)}{dt}\right)_{scat.}$$



Interpreting the BTE:

The BTE is saying that probability is conserved along the path of the particle:

- Particles will move in space according to their velocity:

$$d\mathbf{r} = \frac{d\mathbf{r}(t)}{dt}dt = v(\mathbf{k})dt$$

- Particles change momentum according to the forces acting on them

$$d\mathbf{k} = \frac{d\mathbf{k}}{dt}dt = -\frac{e\mathbf{E}(\mathbf{r})}{\hbar}dt$$

- Particles can scatter from a momentum state **k** into another momentum state **k'** due to interactions with phonons, photons, plasmons, impurities, boundaries, etc.

Solving the BTE

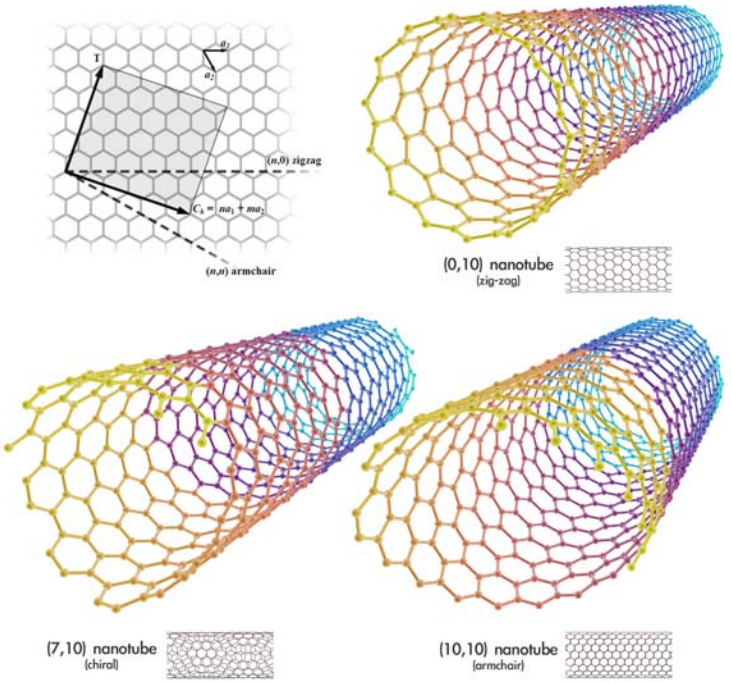
- The BTE poses tremendous computational burdens due to high dimensionality (7-D=3-D space+3-D momentum + time)
- In order to solve it, we must simplify:
  - Consider momentum space only (homogeneous/bulk materials)
    - Iterative methods, spherical harmonics expansions, Rode's Method
  - Consider real space only
    - Moments of the BTE, Hydrodynamic equations, Drift-Diffusion
  - Assume distribution is near equilibrium
    - Relaxation time approximation, Analytical methods
  - Only consider samples of the f(r,k,t)
    - Monte Carlo method for device simulation
  - Consider 1-D systems (produces a 3-D problem)
    - Good for carbon nanotubes, silicon nanowires, etc.

Transport simulation in Carbon Nanotubes

- CNT bandstructure and Density-of-States (DOS)
- CNT phonon dispersion
- 1-D BTE for CNTs
- Upwind Discretization
- Stability and Boundary Conditions (BCs)
- Poisson Equation (self-consistent potentials)
- Scattering Rates
- Linear Analytic method
- Results and Future Work

Single-walled Carbon Nanotubes

- CNTs are rolled-up sheets of monolayer graphene
- Have many interesting properties:
  - Extremely strong
  - Great thermal conductors
  - High optical phonon frequency
  - Can be both semiconducting or metallic depending on how the graphene sheet is rolled up (zig-zag, armchair, chiral)
- Potential applications as
  - FET devices
  - Interconnects
  - Sensors
  - Cooling solutions
  - Filters, etc.



CNT Bandstructure

- In general, bandstructure is obtained by solving the stationary Schroedinger equation for the periodic atomic potential
- CNT Bandstructure obtained by zone folding tight-binding graphene data according to:

$$\mathbf{k}_{zf} = k\frac{\mathbf{K}_2}{\|\mathbf{K}_2\|} + \mu\mathbf{K}_1 \qquad \mu = 0, 1, \ldots, N-1$$

- N is the number of atoms in the unit cell, and k is the CNT wave-vector
- K_1 and K_2 are reciprocal basis vectors of the honeycomb lattice
- This allows a simple and sufficiently accurate treatment of electronic structure
- Other methods, including *ab initio*/DFT possible

CNT Bandstructure

- Graphene bandstructure can be computed using tight-binding by solving the secular equation:

$$det\left[H - ES\right] = 0$$

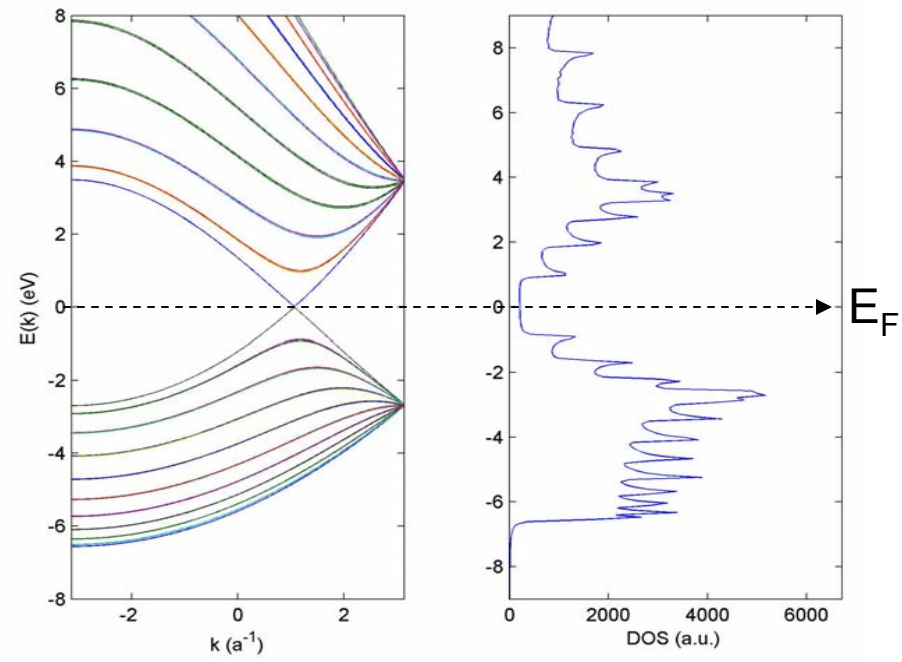
- E is the energy we are solving for, and H and S are given by:

$$H = \begin{pmatrix} \epsilon_{2p} & tf(k) \\ tf(k)^* & \epsilon_{2p} \end{pmatrix} \quad , \quad S = \begin{pmatrix} 1 & sf(k) \\ sf(k)^* & 1 \end{pmatrix}$$

- Momentum dependence enters through the form factor f(k)
- Parameters s and t are the overlap and transfer integrals, and are computed from first-principles calculations.
- Typical values are: epsilon=0, s=0.129, and t=-3.033 eV.

CNT Bands and DOS results

- Results for a (10,10) metallic tube
- Note the bands crossing at zero energy. These will contribute most to electronic transport.
- Often only this portion is taken into account.
- Also note the non-zero density of states around Fermi level
- This makes the nanotube metallic (states available for transport even in equilibrium).

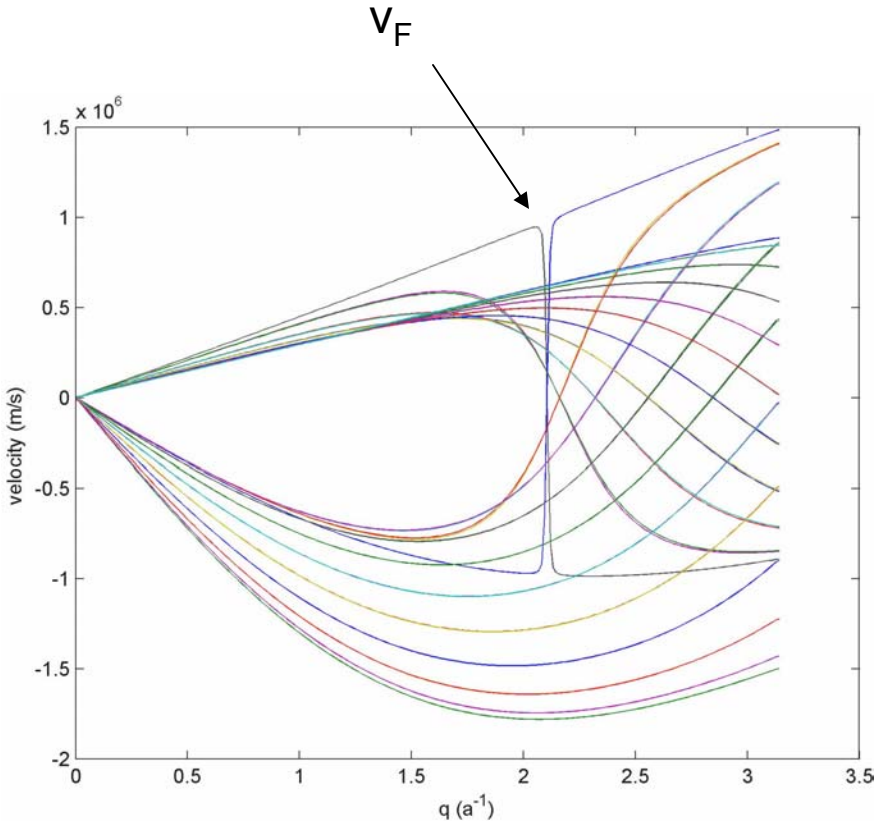


Electron velocities

- Metallic (10,10) tube
- Velocity given by the gradient of the dispersion:

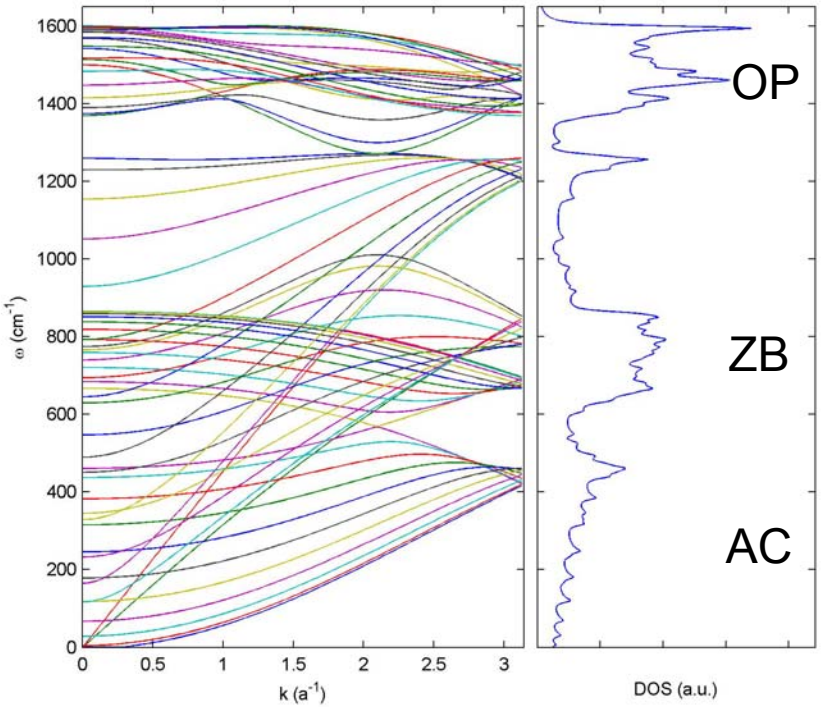
$$v(k,\mu) = \frac{1}{\hbar}\frac{dE(k,\mu)}{dk} = \frac{1}{\hbar}\frac{b_{1,2}^T\,(H' - ES')\,b_{1,2}}{b_{1,2}^T S b_{1,2}}$$

- Velocity highest near Fermi level E_F. This is the typical value of around $8.1*10^5$ m/s.



CNT Phonon dispersion

- Obtained by zone-folding the graphene dispersion
- Force Constant approach by fitting to measured data
- Factors due to bending of the graphene sheet into a tube
- High density of optical (OP) and zone-boundary (ZB) modes
- Strong interaction between electrons and OP and ZB modes

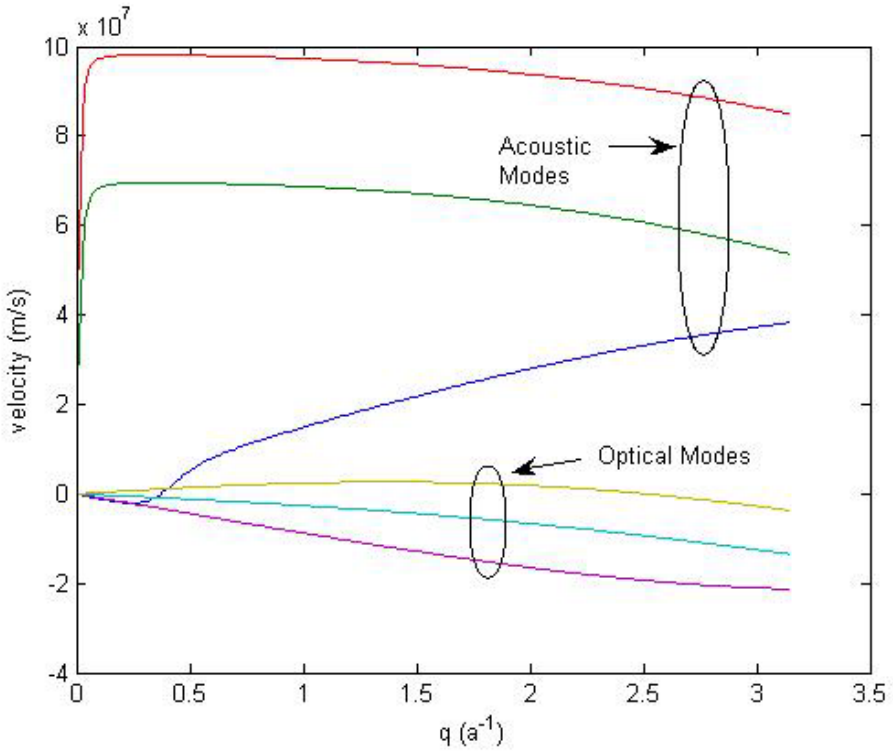


CNT Phonon velocities

- Phonon group velocities also obtained from the gradient of the dispersion:

$$v_g(q,\mu) = \frac{d\omega(q,\mu)}{dq}$$

- Optical modes have flat dispersion giving rise to low group velocities
- Optical modes contribute little to thermal transport



1D Boltzmann Transport Eqn. (BTE)

- Electron BTE (1D):

$$\frac{\partial f_T(x,k,t)}{\partial t} + \frac{eF}{\hbar}\frac{d}{dk} f_T(x,k,t) + v(k,\mu)\frac{d}{dx} f_T(x,k,t) =$$
$$\frac{\Omega}{2\pi}\int dk\prime \left[S(k\prime,k) f_T(k\prime)(1-f_T(k)) - S(k,k\prime) f_T(k)(1-f_T(k\prime))\right]$$

- Sum converted to an integral in the limit of small dk.
- RHS looks like a standard 2D advection equation.
- Can apply standard discretization techniques.

Upwind Discretization

- Determine direction of differencing based on the sign of velocity and field at each (j,k) point

$$f_{j,k}^{n+1} = f_{j,k}^{n} - \frac{f_{j,k}^{n} - f_{eq,j,k}^{n}}{\tau_k} - \frac{1+sgn(v_k)}{2}\nu_k(f_{j,k}^{n} - f_{j,k-1}^{n}) - \frac{1-sgn(v_k)}{2}\nu_k(f_{j,k+1}^{n} - f_{j,k}^{n}) - \frac{1+sgn(F_j)}{2}\nu_j(f_{j,k}^{n} - f_{j-1,k}^{n}) - \frac{1-sgn(F_j)}{2}\nu_j(f_{j+1,k}^{n} - f_{j,k}^{n})$$

relaxation time

- Constant τ_k is the “ralaxation time” computed from the scattering rate integral over all k

Stability and BCs

- Explicit time-stepping places a restriction on step-size Δt dependent on the discretization

$$\left|\frac{eE}{\hbar}\right| \frac{\Delta t}{\Delta k} < 1$$

$$|v_{max}| \frac{\Delta t}{\Delta x} < 1$$

- For Δx~1nm, Δt~1fs
- This is comparable to the relaxation time (10~50fs)
- Relaxation time poses another limitation on the timestep ($\Delta t << \min_k(\tau_k)$)
- Periodic BCs in momentum (lattice is periodic)
- Homogenous Neumann BCs in space (quasi-equilibrium)
- Fermi-Dirac initial condition (start off with equilibrium)

Poisson Equation

- Charge and current can be obtained from

$$\rho(t,x) = e\int f(x,k,t)dk$$

$$I(x,t) = e\int v(k)f(x,k,t)dk$$

- Solve the Poisson equation for the potential along the tube

$$V_{j+1}^n - 2V_j^n + V_{j-1}^n = \frac{\Delta x^2 \rho_j^n}{\epsilon}$$

- Boundary conditions given by applied potentials
- Extend to full 3-D Poisson for semiconducting CNTs

CNT Scattering Rates

- Scattering rates derived from quantum-mechanical “Fermi’s Golden Rule”
- Coupling potentials between electrons and phonons given by Bardeen’s Deformation Potential theory
- Acoustic rates have a factor of q squared:

$$\frac{1}{\tau(k_i,\mu_i)} = \sum_{k_f,\mu_f} \frac{\hbar D_{ac}^2 \left[q^2 + \left(\frac{2\mu_p}{D}\right)^2\right]}{2\rho\omega_{q,\mu_p}} \left(N_{q,\mu_p} + \frac{1}{2} \mp \frac{1}{2}\right) \delta\left(E(k_i,\mu_i) - E(k_f,\mu_f) \pm \omega_{q,\mu_p}\right)$$

- The signs depend on absorption or emission of a phonon.
- The δ function controls energy conservation
- Can be replaced by a Lorentzian to allow collisional broadening

CNT Scattering Rates

- Zone Boundary:

$$\frac{1}{\tau(k_i,\mu_i)} = \sum_{k_f,\mu_f} \frac{\hbar D_{ZB}^2}{2\rho\omega_{q,\mu_p}} \left(N_{q,\mu_p} + \frac{1}{2} \mp \frac{1}{2} \right) \delta\left(E(k_i,\mu_i) - E(k_f,\mu_f) \pm \omega_{q,\mu_p} \right)$$

- Optical rate:

$$\frac{1}{\tau(k_i,\mu_i)} = \sum_{k_f,\mu_f} \frac{\hbar D_{OP}^2}{2\rho\omega_{q,\mu_p}} \left(N_{q,\mu_p} + \frac{1}{2} \mp \frac{1}{2} \right) \delta\left(E(k_i,\mu_i) - E(k_f,\mu_f) \pm \omega_{q,\mu_p} \right)$$

Broadening

- When the scattering rate is high ($\delta \sim k_B T$) transitions can occur between perturbed "quasi-particle" states
- This is described by the particle "self-energy"
- For simplicity assume self-energy is pure imaginary (no level shift, only broadening).
- Replace δ-function with a Lorentzian distribution
- Can add self-consistency by using optical theorem:

$$\delta_{k,\mu} = -\frac{\hbar}{2} Im\ \Sigma(k,\mu) = \frac{\hbar}{2\tau(k,\mu)}$$

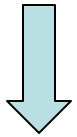
- Take into account initial and final state broadening:

$$\delta = \delta(k_i,\mu_i) + \delta(k_f,\mu_f)$$

Broadening

- Replace δ-function with a Lorentzian distribution

$$\delta\left(E(k_i,\mu_i) - E(k_f,\mu_f) \pm \hbar\omega(q,\mu_p)\right)$$



$$\frac{1}{\pi}\frac{\delta}{\delta^2 + \left(E(k_i,\mu_i) - E(k_f,\mu_f) \pm \hbar\omega(q,\mu_p)\right)^2}$$

- This makes numerical calculation of scattering rate (relaxation time) easier
- Energy no longer conserved exactly, only on the average

Linear Analytic Method

- Break integral apart into many small segments in k
- Expand energies to 1st order and integrate analytically over each small segment in k-space
- Add up contributions form all segments in k-space

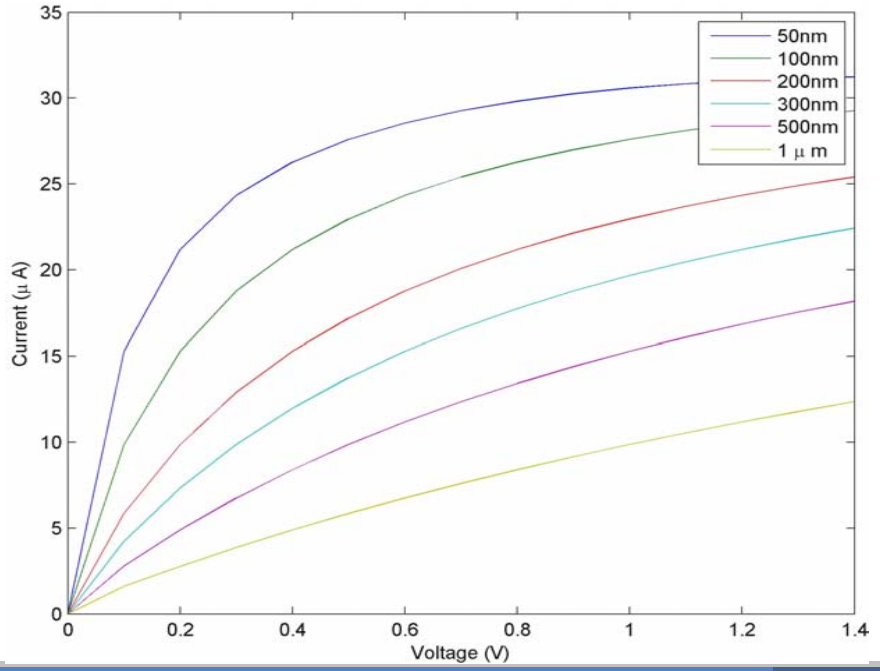
$$\int dk \frac{1}{\pi} \frac{\delta}{\delta^2 + \left(E(k_i,\mu_i) - E(k_f,\mu_f) \pm \hbar\omega(q,\mu_p)\right)^2}$$

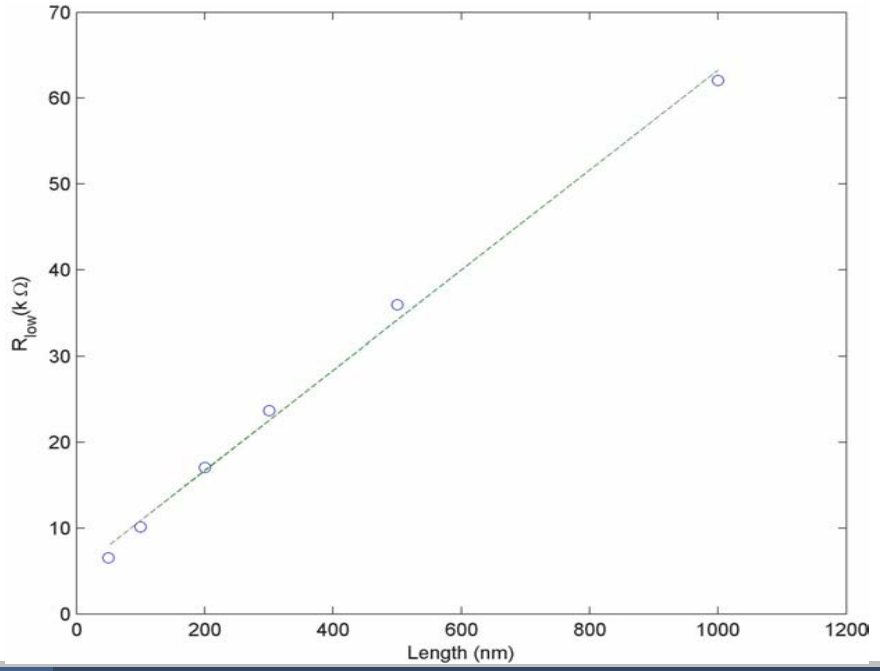
$$= \sum_{k_f,\mu_f} \int_{dk} \frac{1}{\pi} \frac{\delta}{\delta^2 + \left(\Delta E + \hbar v(k_f)(k-k_f)\right)^2}$$

$$= \sum_{k_f,\mu_f} \frac{1}{\pi\hbar v(k)} \left(tan^{-1} \frac{\hbar v(k_f)dk - \Delta E}{\delta} + tan^{-1} \frac{\hbar v(k_f)dk + \Delta E}{\delta} \right)$$

Results: IV curves for (10,10) SWNT

- Current saturates around 25μA due to onset of strong optical scattering
- Resistance scales linearly with length in the low-field regime (interconnect applications)





Comments and Extensions

- Extends naturally to many other 1-D systems:
  - Carbon Nanoribbons (CNRs) are candidates for future FET devices
  - Semiconducting CNTs show interesting current up-kick
  - Rough Si nanowires show great potential for energy harvesting
- Phonon (thermal) transport is treated with a similar discretization scheme (no interaction with the electric field)
- Non-equilibrium transport can be explored in detail
- Thermo-electric properties can be simulated
- This requires coupling through scattering integrals (for each k sum over all k', expensive ~1hr/tstep)
- Possible efficient parallel implementation

References

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- “Basic Semiconductor Physics” by Chihiro Hamaguchi, Springer (2001), chapter 6
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