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Effects of electron correlations on bulk thermal transport

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Motivation

- Cooling and power generation applications
- Figure-of-merit ZT evaluates the efficiency of a bulk material as a cooling element.
- Commercially available semiconductor devices have ZT ~1
- To be competitive with the efficiency of conventional mechanical refrigerators we need ZT of 3 or 4

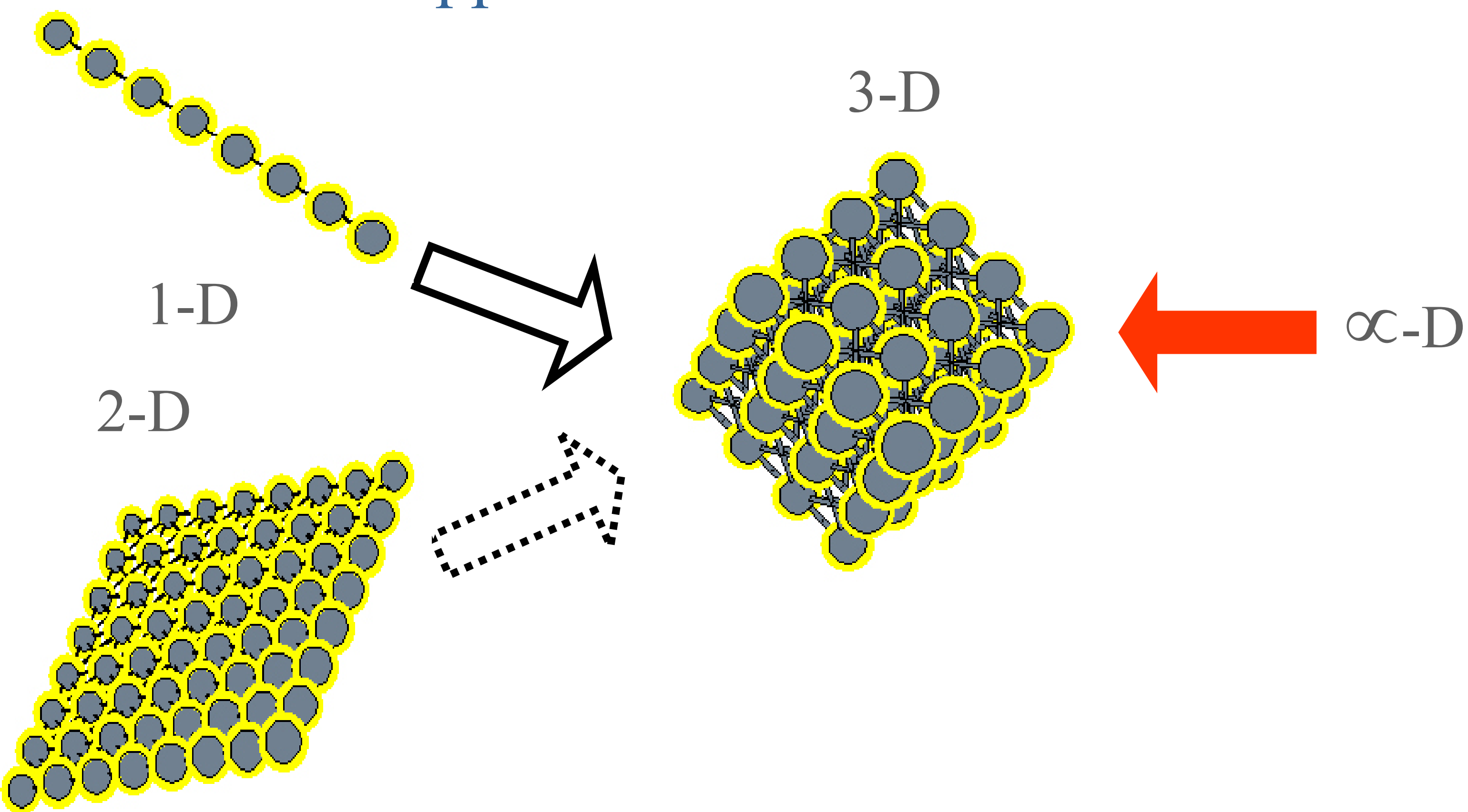
Correlated electron systems/models

- Coulomb repulsion between electrons is taken into account.

- As a consequence materials exhibit properties which are absent in single electron approximation.

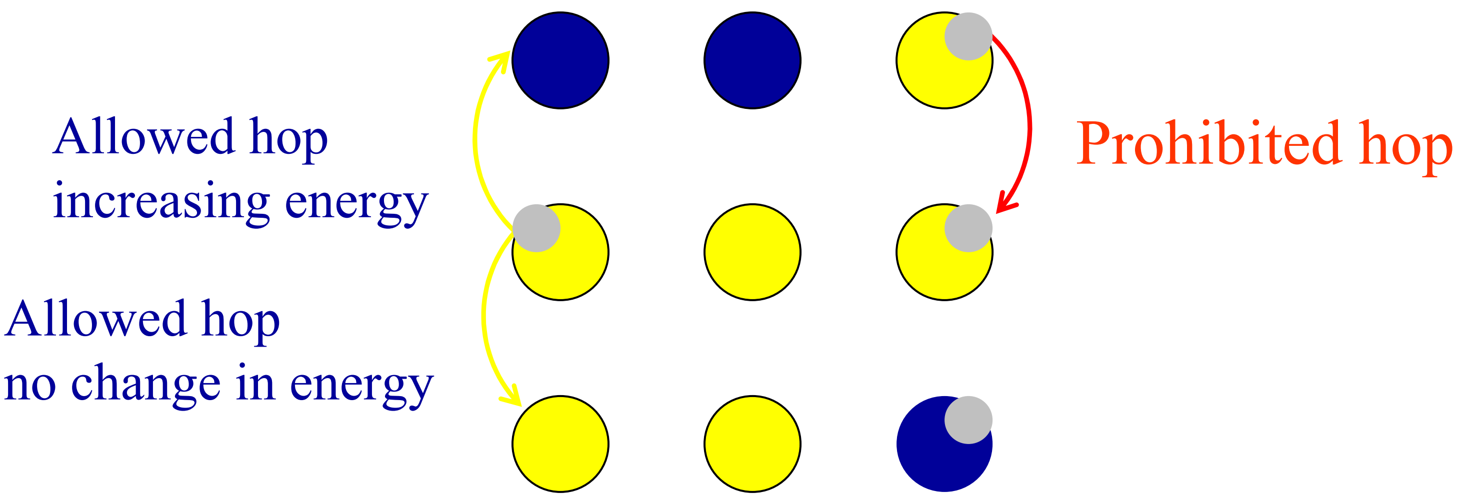
- Those of interest are difficult to solve even numerically.

Predicting properties of real materials – which approach is most realistic?



*W. Metzner and D. Vollhardt, Phys. Rev. Lett. 62, 324 (1989)

Spinless Falicov-Kimball model – underlying physics



- Two species of atoms, A and B, form an alloy $A_X B_{1-X}$
- Electrons from atoms A form a conduction band
- Electrons from atoms B are localized on ions B
- Hops of conduction electrons to sites B are unfavorable because of Coulomb repulsion U
- $Ta_{0.6}N$ is believed to be described by the model

Spinless Falicov-Kimball model – binary alloy picture

$$H = -\frac{t^*}{\sqrt{Z}} \sum_{\langle i,j \rangle} c_i^\dagger c_j + U \sum_i w_i c_i^\dagger c_i$$

- c^+,c^+ – conduction electrons
- w_i – random variable equal 0 or 1 (presence of ion with localized electron)
- Z – number of nearest neighbors
- U – same site Coulomb interaction strength
- hopping occurs only to nearest neighbors
- t^* –energy scale

DMFT solution

- Self energy Σ(ω) in Fourier space doesn't depend on $\vec{k}$

- Bethe lattice of infinite connectivity

- Number of conduction electrons + Number of localized electrons (w_1) = Number of sites => System undergoes MIT at high U's

- Green's function G(ω) satisfies an algebraic cubic equation, which can be solved "exactly"

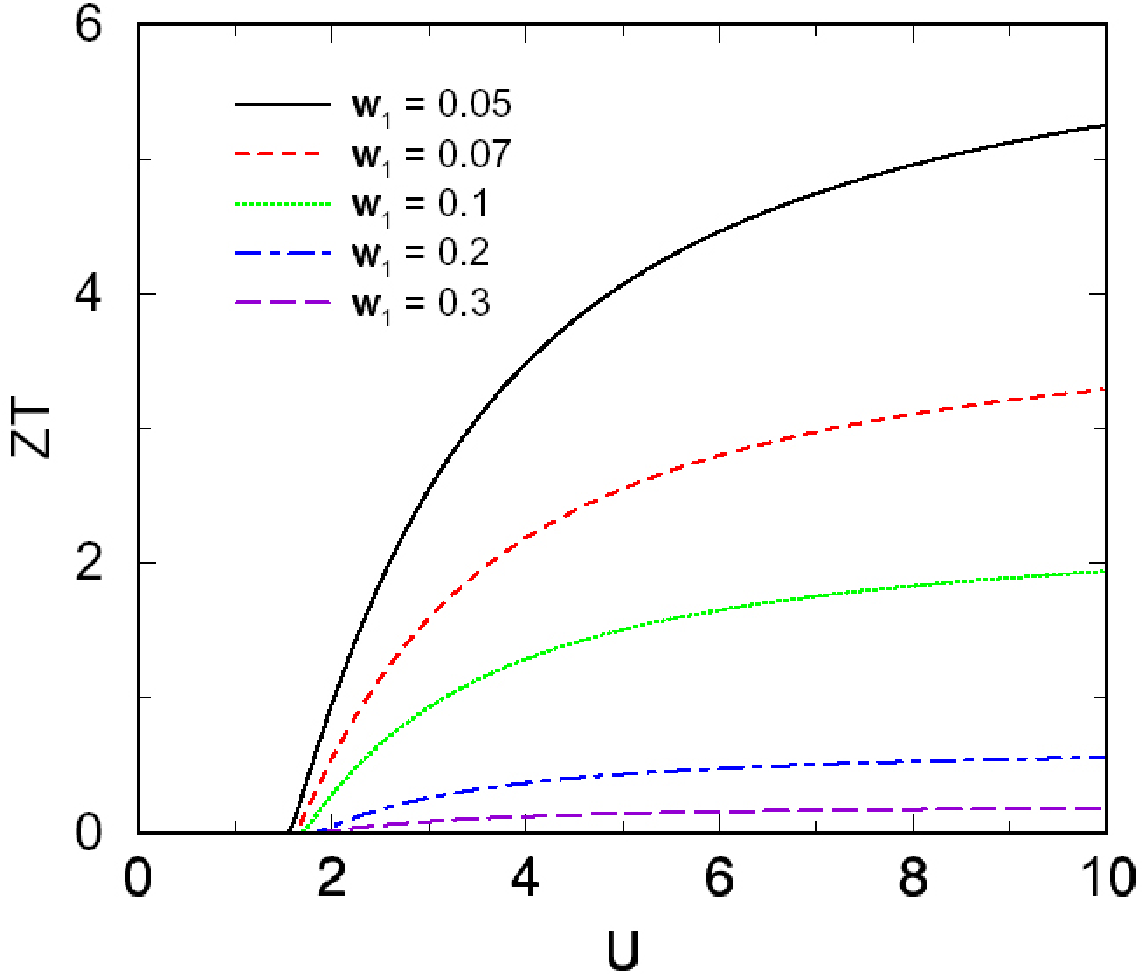
$$G^3 - 2(\omega + \mu - U/2)G^2 + [1 + (\omega + \mu - U/2)^2 - \frac{U^2}{4}]G - [\omega + \mu - U/2 + U(w_1 - \frac{1}{2})] = 0$$

Transport properties within linear response approximation

- Electrical conductivity σ_{dc}
- Thermopower S
- Thermal conductivity κ_e (electronic contribution)
- Lorenz number $L \sim \kappa_e / (\sigma_{dc} T)$
- Figure of merit $ZT_e = T\, \sigma_{dc} S^2/\kappa_e$ $(ZT = T\, \sigma_{dc} S^2/(\kappa_e + \kappa_l))$

All characteristics can be expressed as integrals of functions expressed in terms of Green's function G(ω) (using Jonson-Mahan theorem)

Figure of merit at zero temperature



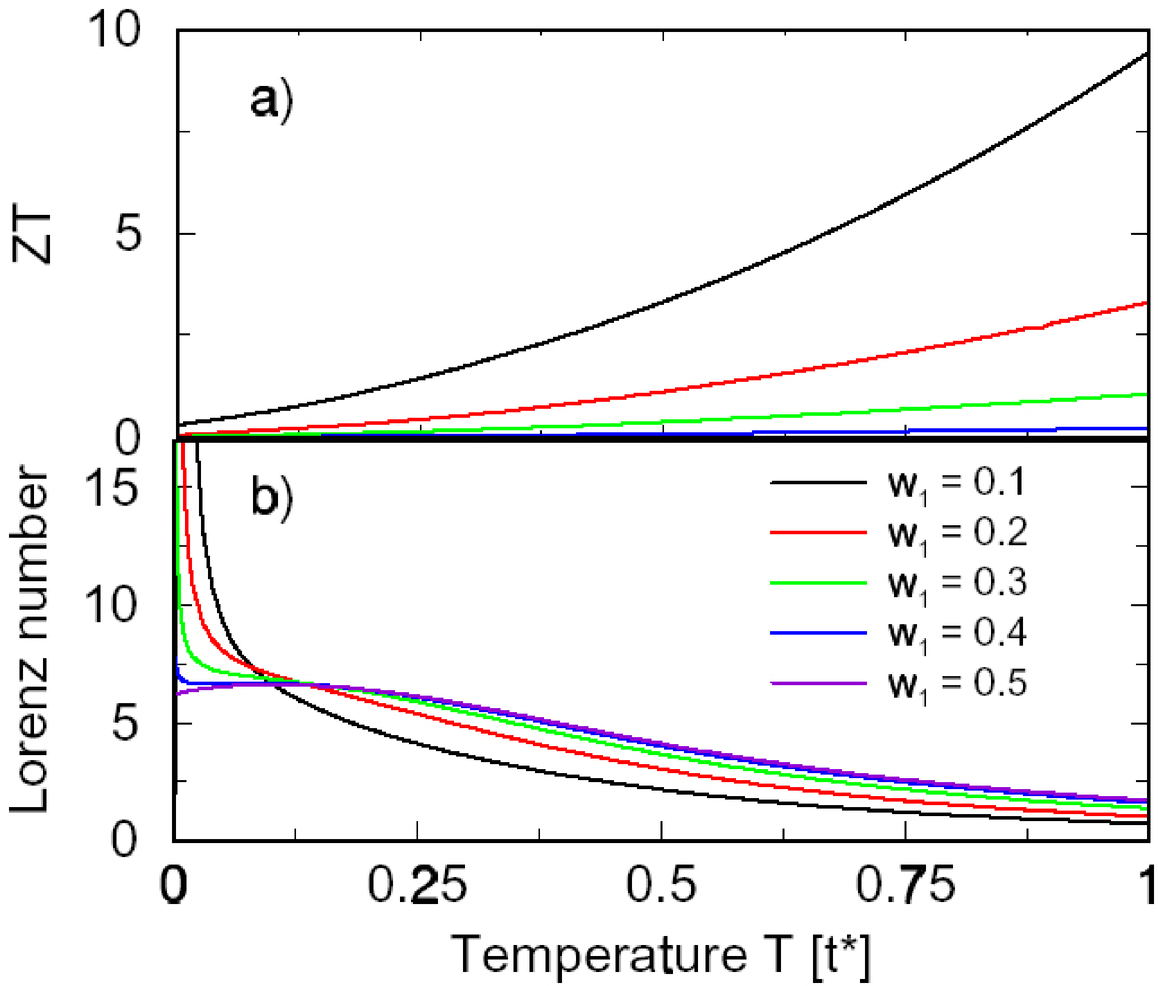
T=0

$\rho_e = 1 - w_1$

When there is no gap in DOS ZT(T=0) = 0

DOS= $-\,\mathrm{Im}[G(\omega)]/\pi$

Transport at finite temperatures



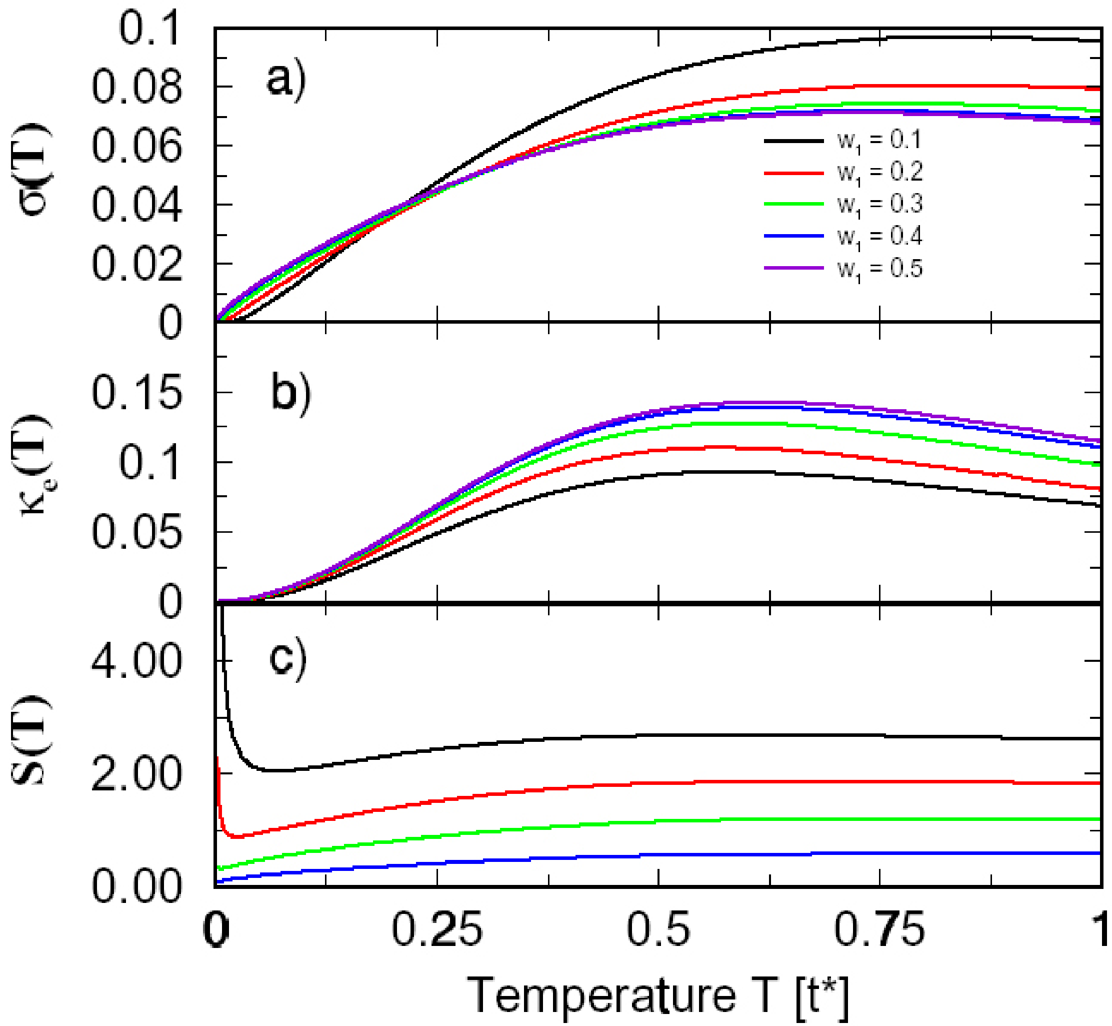
U=2 (MIT has occurred)

ZT grows with temperature

ZT (w_1=0.5) ≡ 0

w_1=0.5 – particle-hole symmetric case

Transport at finite temperatures



U=2 (MIT has occurred)

Electric and thermal conductivities at low temperatures are exponentially small

Divergence of thermopower signals the breakdown of linear response approximation at low temperatures

Conclusions

- Linear response approximation fails in zero T limit

- Very high ZT's are achievable at small "dopings"and high temperatures

- It's unlikely to have an effective cooling / power generating device at low T's because of :
  1. dominant lattice contribution to thermal conductivity
  2. exponentially small electrical conductivity