Effects of particle-hole asymmetry on the Mott-Hubbard Metal-Insulator Transition

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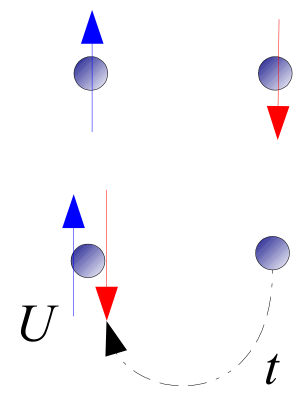
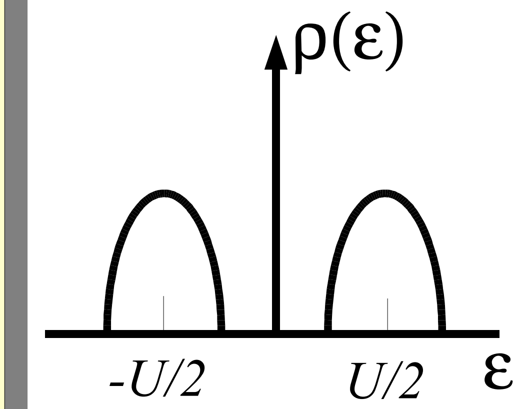
Metal-Insulator Transition (MIT)

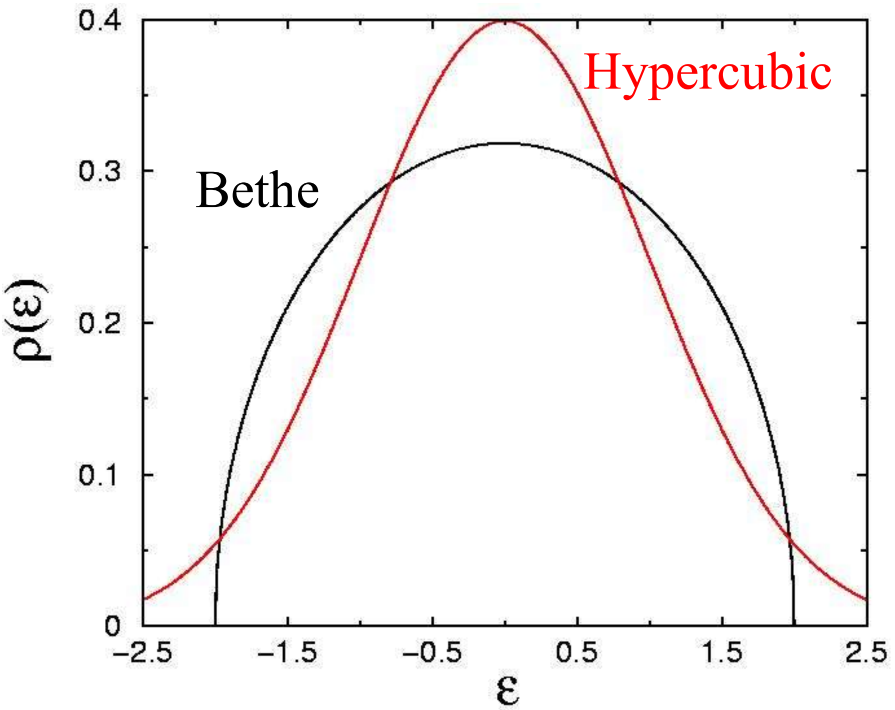
- Physics – local Coulomb repulsion *U* that forbids double occupancy of the electrons creating an insulator, when there is one electron per site on average
- Experiment - variety of materials (MnO, NiO, NiS, $YBa_2Cu_3O_6$), for which band structure calculations underestimate the gap or yield a metal
- Hubbard model – analyzed with many methods (NRG, QMC, etc.) well understood, difficult to develop an approximation which would describe both the weakly correlated Fermi liquid phase and strongly correlated insulating phase
- DMFT - Much progress has been made with Dynamical Mean Field Theory (DMFT), i.e. the limit of infinite dimensions, but numerics are complicated and delicate





MIT at half filling

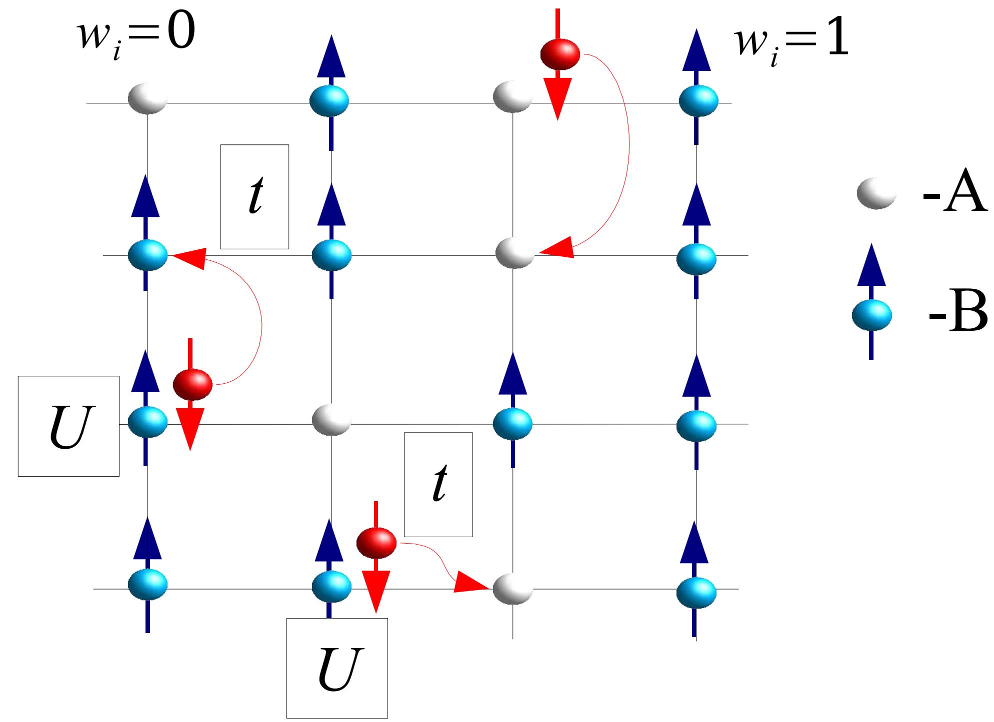
- Hubbard model - MIT occurs only at half filling
- Within DMFT, noninteracting density of states (DOS):
  - Hypercubic lattice – Gaussian, infinite bandwidth
  - Bethe lattice – semicircle, finite bandwidth



- Hypercubic: DOS can vanish only when the self-energy diverges, at a single point forming a pseudogap, i.e. MIT occurs when self-energy develops a pole
- Bethe: well defined gap, same scenario – MIT occurs when the self-energy develops a pole at the chemical potential
- What happens if MIT occurs in a system where particle-hole symmetry is broken? *(Jorge Hirsch)*
- Most real materials do not have particle-hole symmetry

How to choose the model?

- Needed a modified model which has MIT away from half-filling
- Falicov-Kimball model
- Binary alloy picture
- Exhibit MIT for a wide range of fillings $0<w_1<1$
- Numerics are under excellent control
- Scale effective bandwidth for different lattices $W=\sqrt{\int \epsilon^2 \rho(\epsilon) d\epsilon}$
- Ta_xN – example



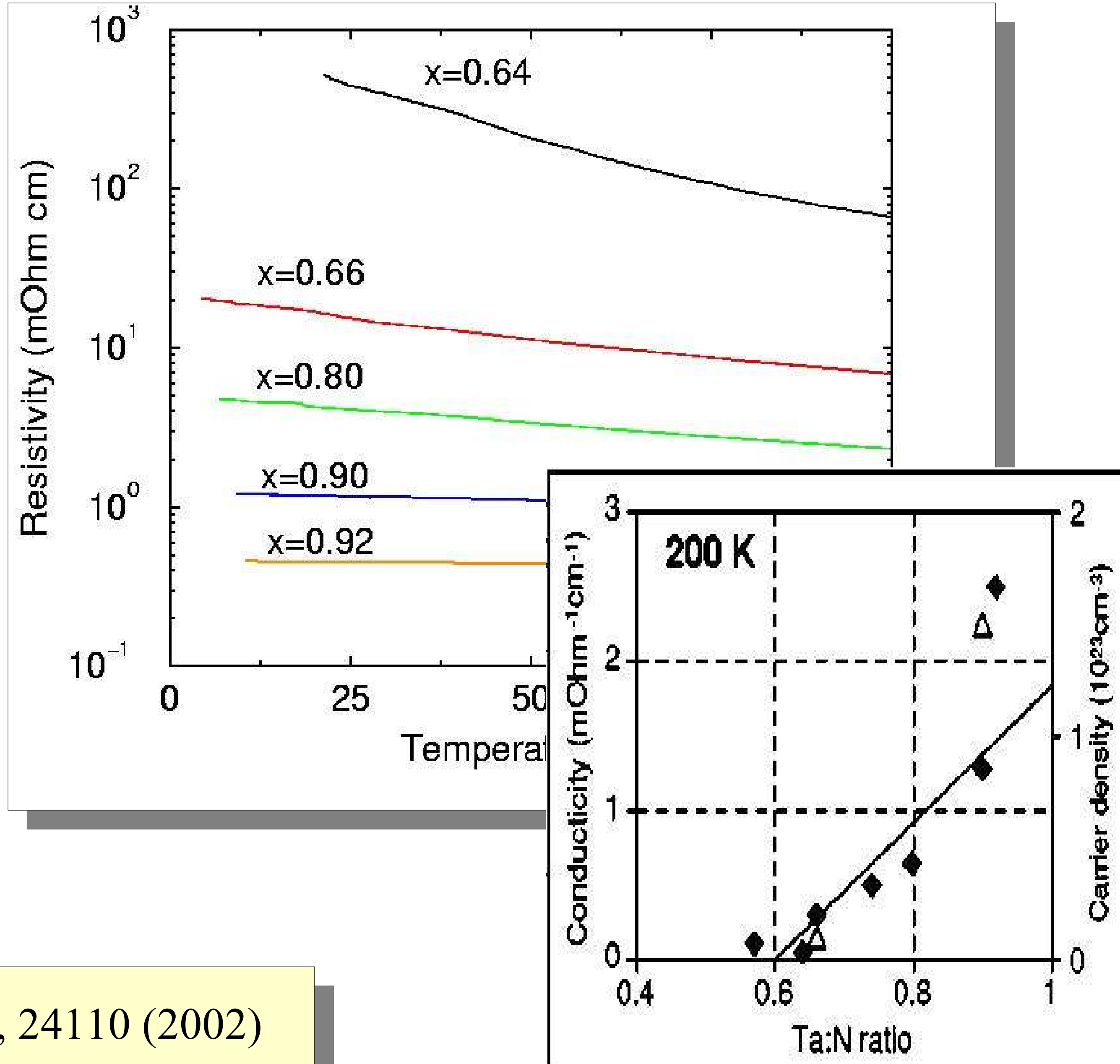
$$H=-t\sum_{\langle i,j\rangle} c_i^+ c_j + U\sum_i w_i c_i^+ c_i$$

$w_1=\langle w_i\rangle$ - average filling

$\rho_e=1-w_1$ - fix total number of particles

Binary alloy picture

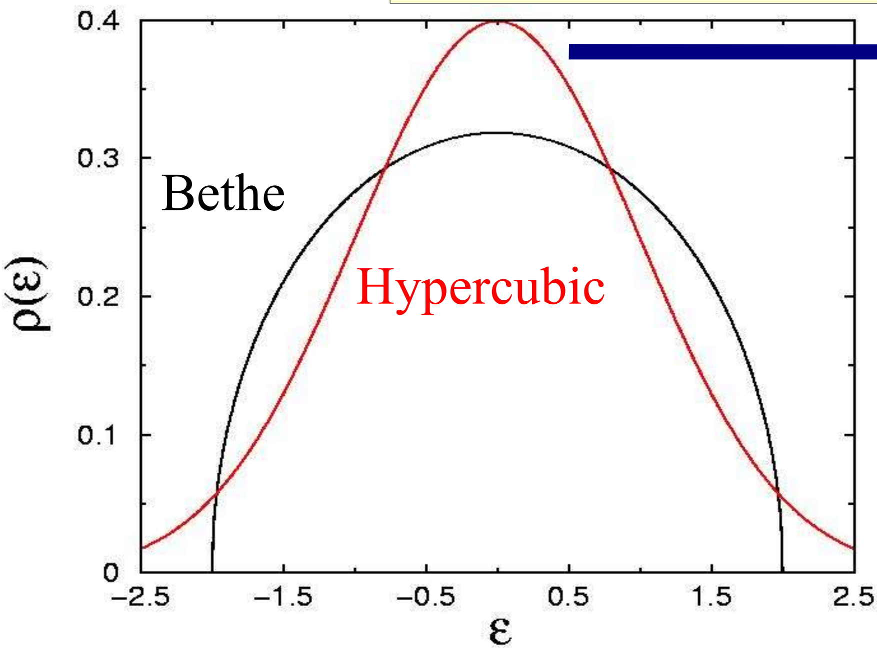
- Ta_xN – binary alloy: A(Ta atoms)+ B(vacancies)
- Vacancies interact with conduction e^- and may trap them
- MIT is observed at x=0.6 (Ta:N ratio)
- Potential problem: localization due to disorder becomes increasingly important as x decreases



L. Yu *et al. Phys. Rev. B* **65**, 24110 (2002)

DMFT

Feed in noninteracting density of states



Converge $\Sigma(\omega)$

Hilbert transformation:

$$G(\omega)=\int d\epsilon\,\rho(\epsilon)\frac{1}{\omega+\mu-\Sigma(\omega)-\epsilon+i\delta}$$

Dyson's equation:

$$G_0(\omega)=\left[G(\omega)^{-1}+\Sigma(\omega)^{-1}\right]^{-1}$$

Exact impurity solution:

$$G(\omega)=(1-w_1)G_0(\omega)+w_1\frac{1}{G_0(\omega)^{-1}-U}$$

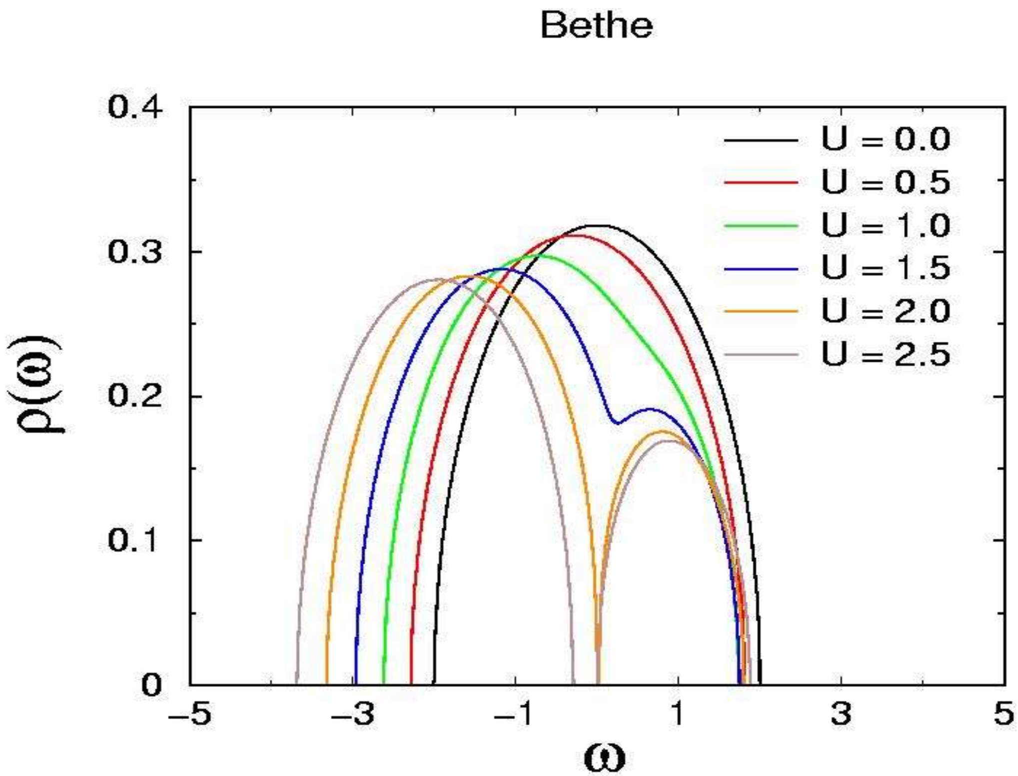
Dyson's equation for $\Sigma(\omega)$

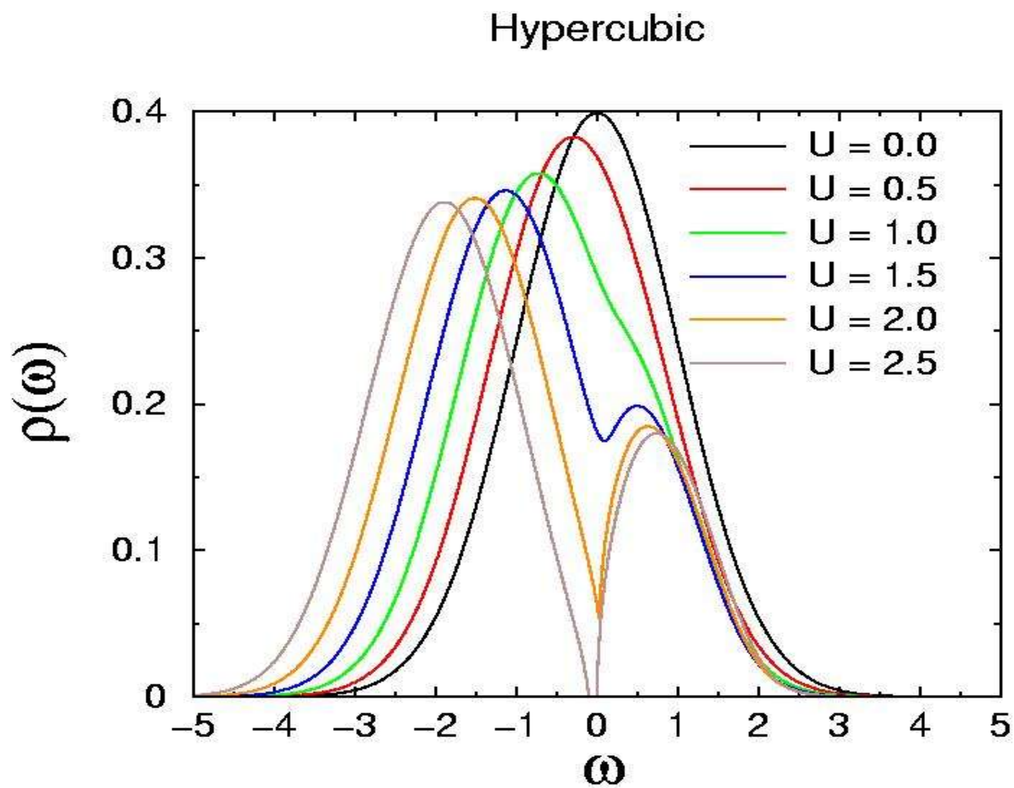
M. Jarrell, Phys. Rev. Lett. **69**, 168 (1992)

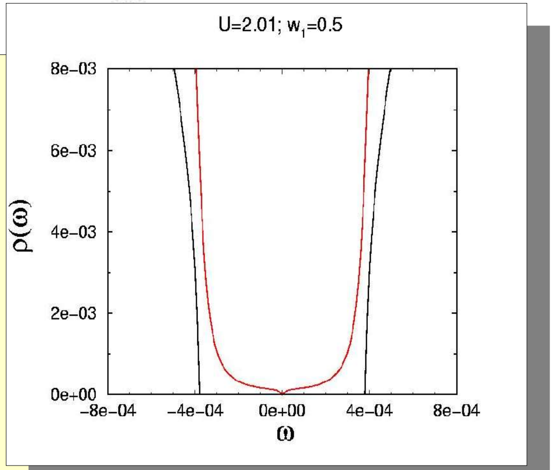
- In the limit $d\to\infty$ FK model solved exactly[1] with DMFT [2]
- Self-energy $\Sigma(\omega)$ has no momentum dependence
- Non-interacting DOS in infinite dimensions:
  - Bethe $\rho_{Bethe}(\epsilon)=\sqrt{4-\epsilon^2}/2\pi$ - finite bandwidth
  - Hypercubic $\rho_{HC}(\epsilon)=\exp(-\epsilon^2/2)/\sqrt{2\pi}$ - infinite bandwidth

[1] J.K.Freericks, V. Zlatic Rev. Mod. Phys. **75**, 1333 (2003)
U.Brandt, C.Mielsch, Z. Phys.**75**, 365 (1989); **79**, 295 (1990)
[2] W.Metzner, D.Vollhardt Phys. Rev. Lett. **62**, 324 (1989)

DOS away from half-filling (w_1=0.25)







- Algorithm converges to about 13 digits

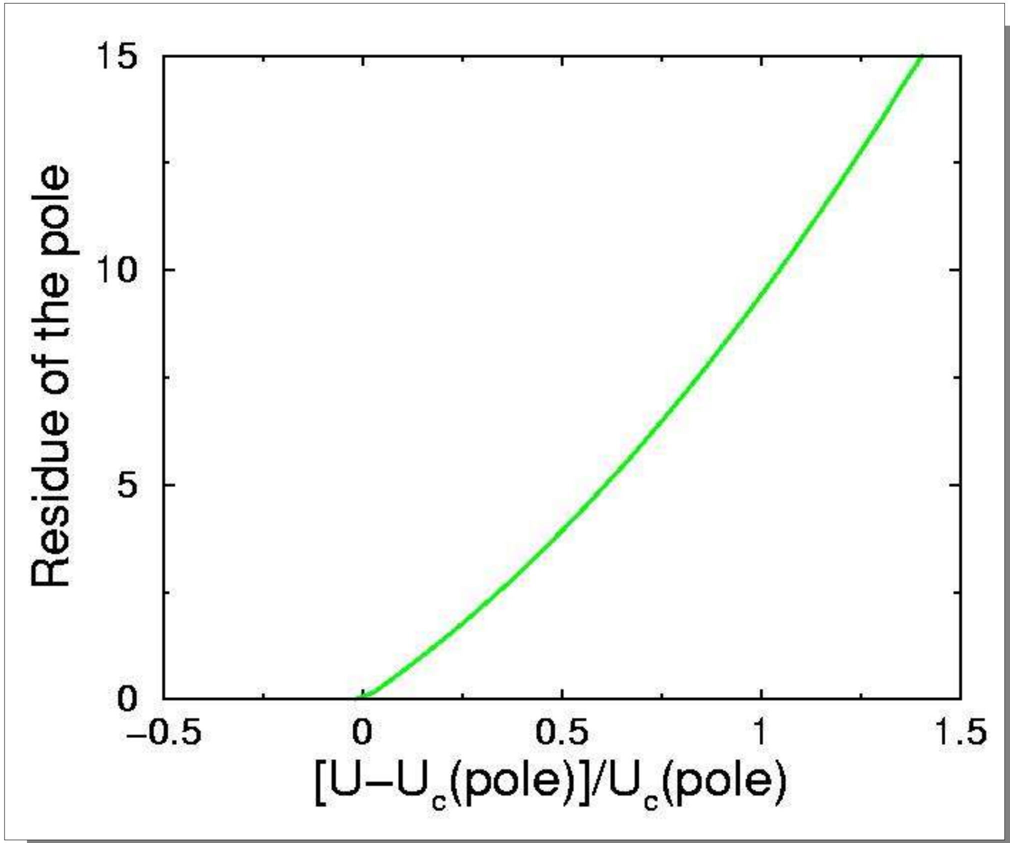
Interacting DOS are defined

$$\rho_{int}(\omega) = -Im[G(\omega)]/\pi$$

- Insulating phase – real gap or pseudogap (blow up)
- No quasiparticle peak, no *T*-dependence

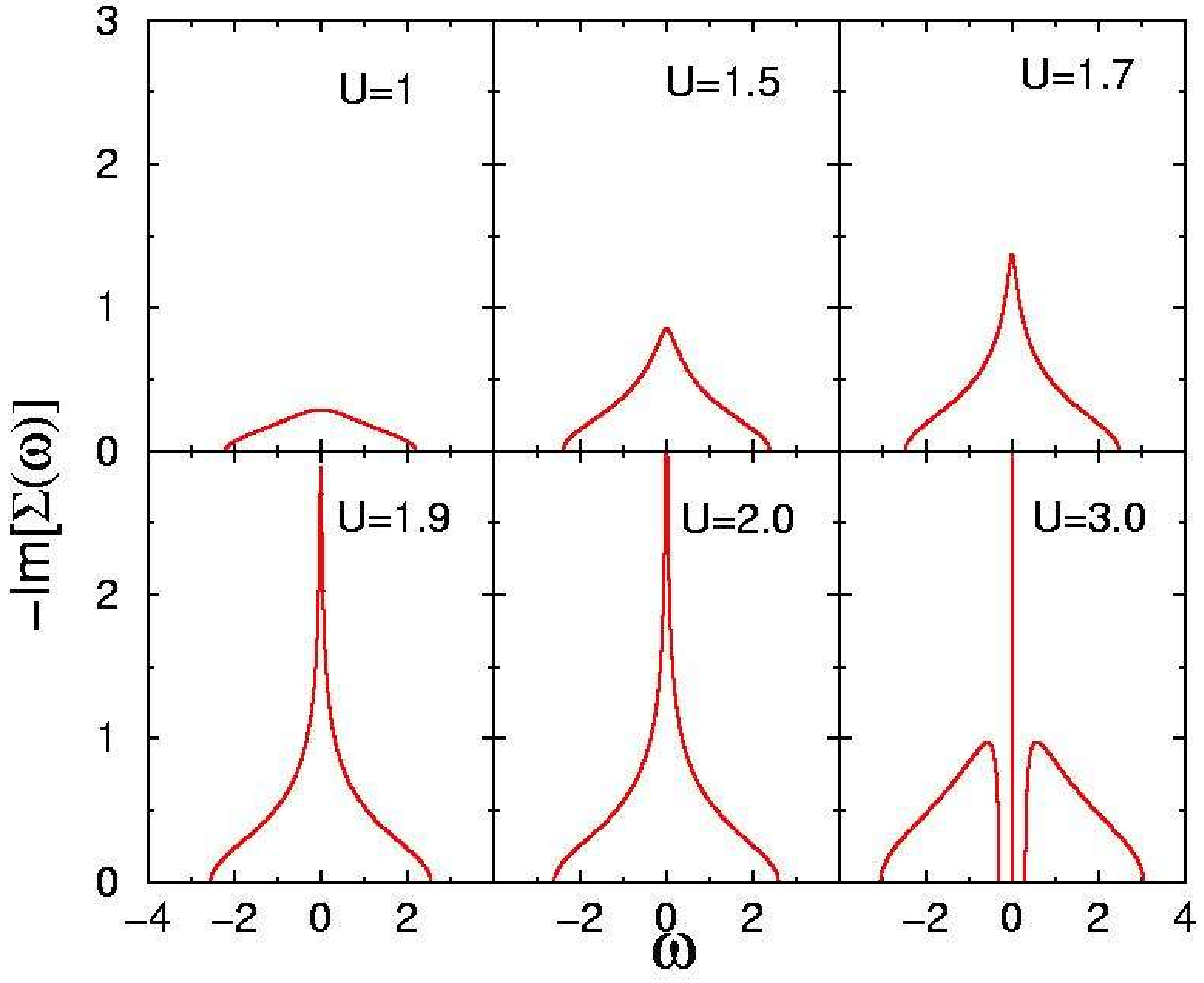
MIT and pole in self-energy

- U_c(gap) – critical U for the gap opening
- U_c(pole) – critical U for the pole formation
- Half-filling – *U_c(gap)=U_c(pole)* always – self-energy develops a pole at MIT
- Pole may indicate MIT
- Residue is a universal plot for all fillings on both lattices (scaling holds)
- Can residue of the pole be an order parameter?



MIT and pole in self-energy (Im part)

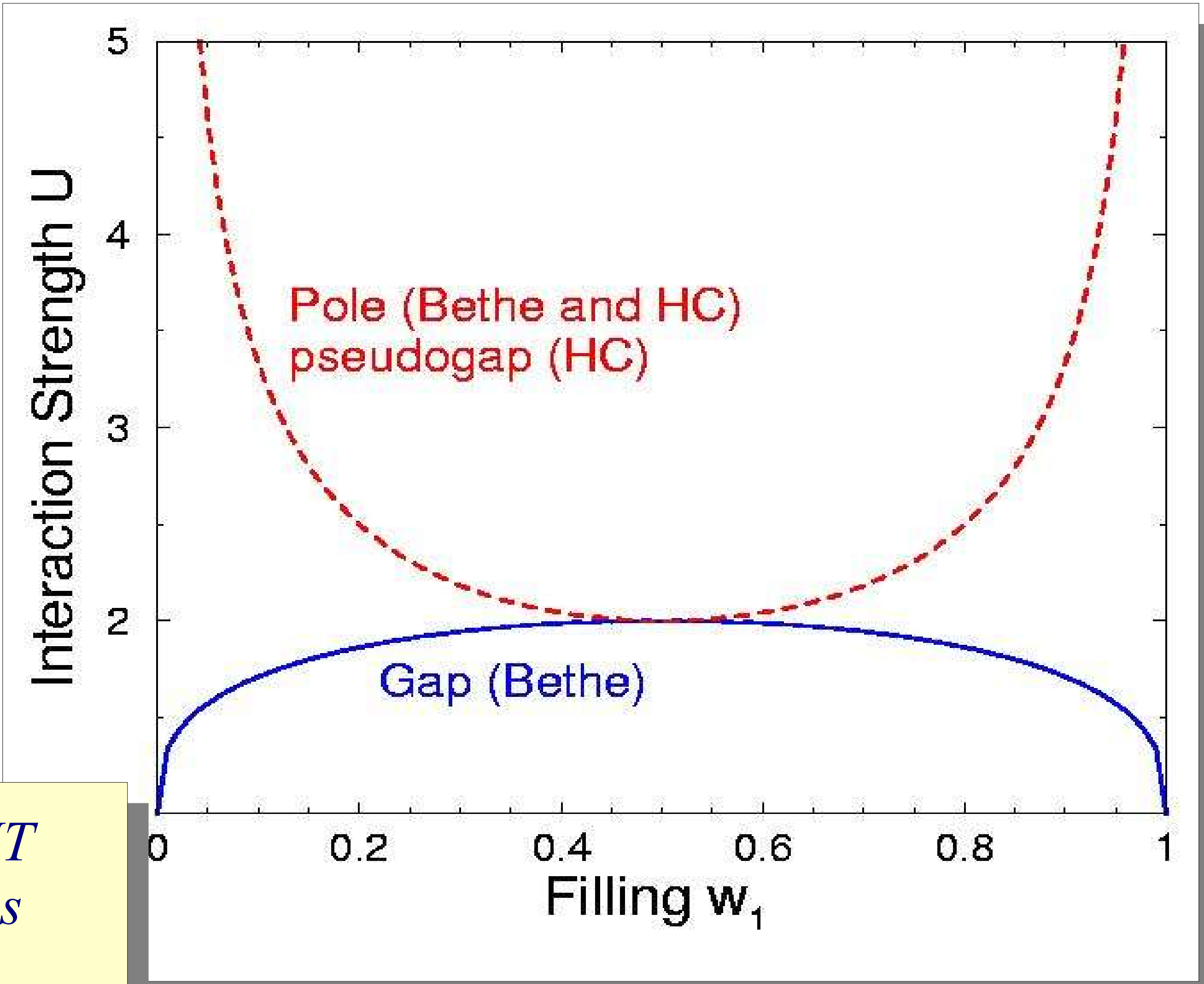
- Bethe lattice
- Non-Fermi liqud
- Pole is formed together with opened gap
- Pole shows as delta function in Im[Σ]



Gap and pole in self-energy

- Green's function satisfies the cubic equation (Bethe)
- Condition for the development of the pole in the self-energy
- Condition for band edges yields critical U for the gap opening

Different U's away from half-filling

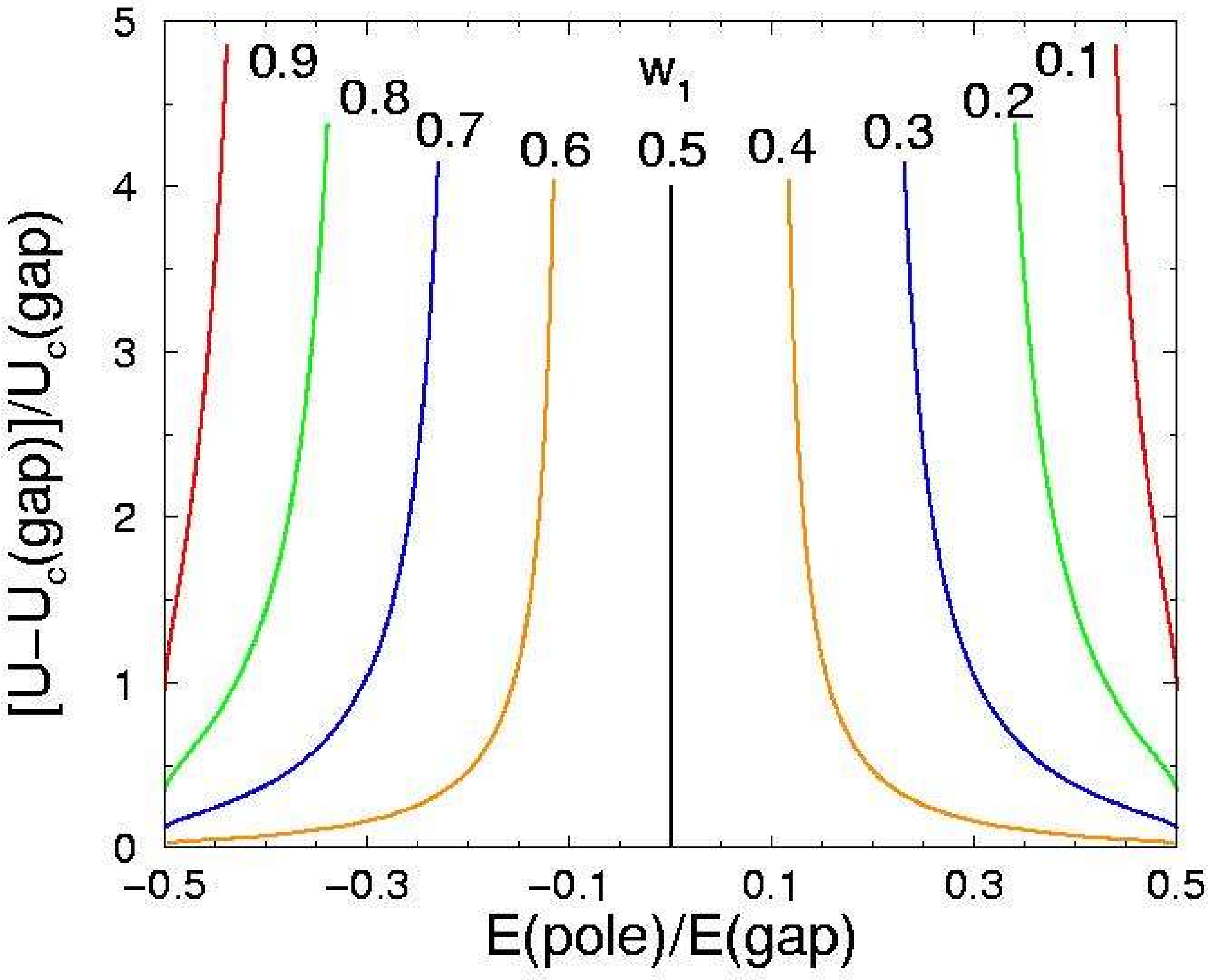


The scenarios for the MIT on HC and Bethe lattices are NOT the same

$$G^3 - 2xG^2 + \left(1 + x^2 - \frac{U^2}{4}\right)G - (x + \alpha) = 0 \qquad x = \omega + \mu - U/2;\ \ \alpha = U\left(w_1 - \frac{1}{2}\right)$$

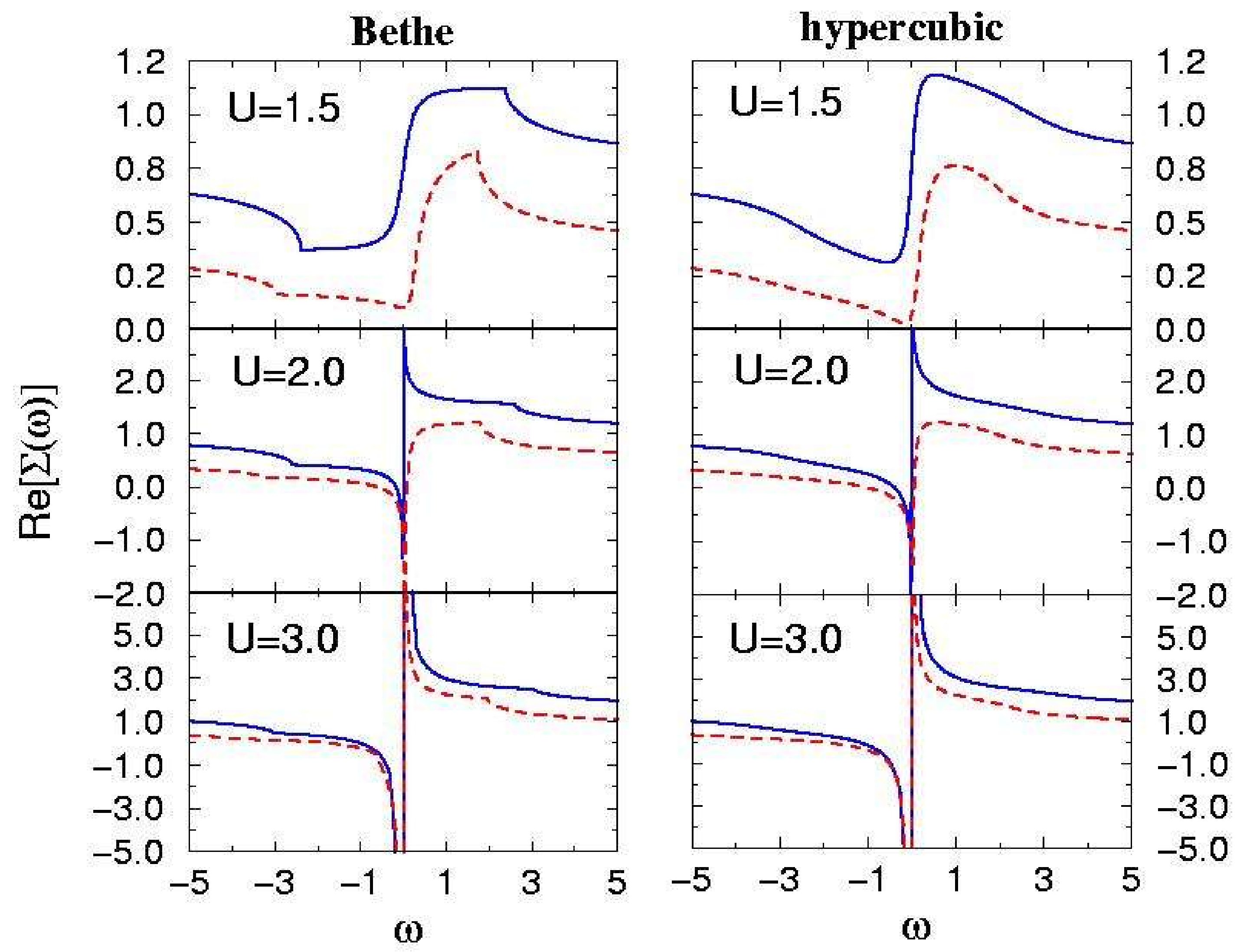
Pole formation (Bethe lattice)

- Relative *U* vs. relative location of the pole in the gap
- Half-filling – pole is in the middle of the gap
- For $w_1 \neq 0.5$ the pole first appears at one of the band edges at $U=U_c(pole)$ then drifts closer to the center
- There is no smooth transition between half-filled and particle-hole asymmetric cases

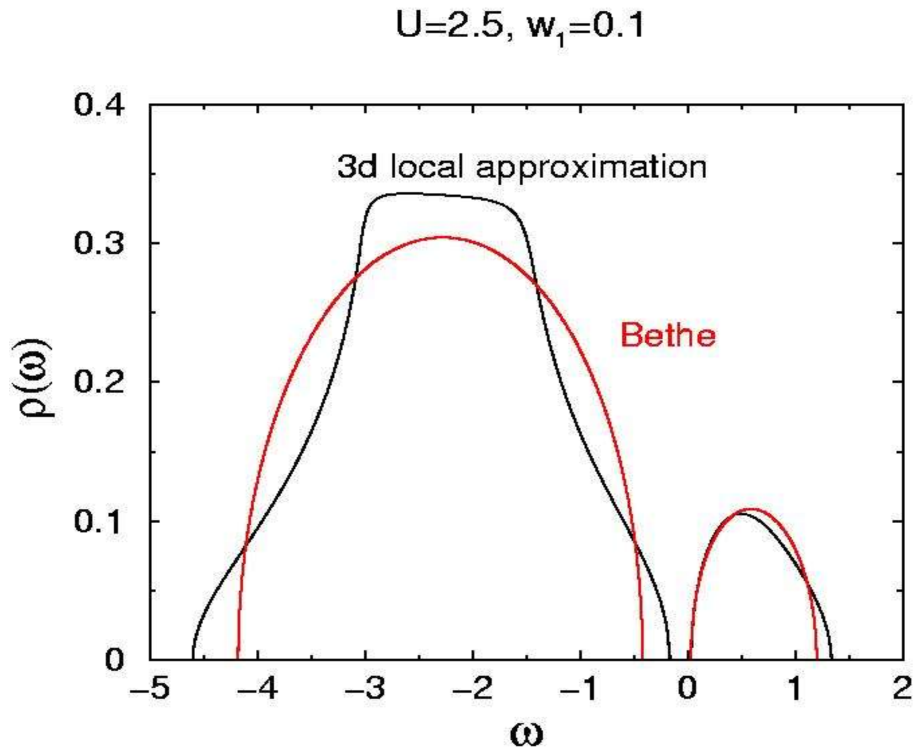


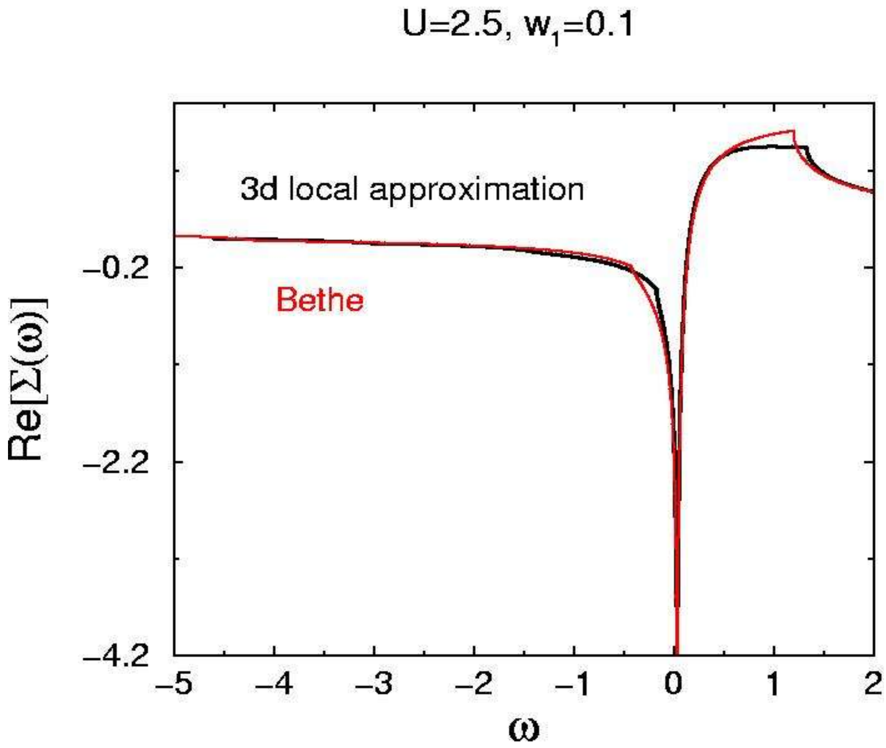
Evolution of self-energy (Re part)

- Blue – w_1=0.5
- Red – w_1=0.25
- U=2 insulator for both w_1=0.5 and w_1=0.25
- w_1=0.25 - no pole
- Re[Σ(ω)] (Re[G(ω)]) - kinks at the new band edges
- Residue of the pole does not describe transition on the Bethe lattice



What causes the differences?





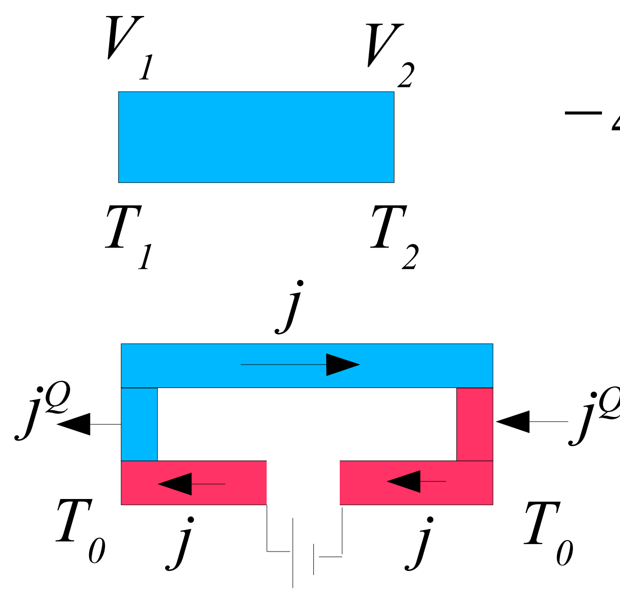
- Is it due to the absence of percolation loops on Bethe lattice?
- Local approximation calculations on the 3*d* lattice, it has loops, but also has finite bandwidth
- Example: U=2.5, w_1=0.1, well developed gap DOS but no pole
- Bandwidth matters
- Bethe lattice is more physical than HC

Bulk transport and thermal properties

- Is pole formation significant?
- Use Kubo-Greenwood approach
- Invoking Johnson-Mahan theorem[3] for transport coefficients

$$L_{ij}=\frac{\sigma_0}{e^2}\int d\omega\left(-\frac{df(\omega)}{d\omega}\right)\tau(\omega)\omega^{i+j-2}$$

- dc conductivity $\sigma=e^2 L_{11}$
- Thermopower $S=-\frac{k_B}{|e|T}\frac{L_{12}}{L_{11}}$
- Thermal conductivity $\kappa_e=\frac{k_B^2}{T}\left[L_{22}-\frac{L_{12}L_{21}}{L_{11}}\right]$
- Thermoelectric figure-of-merit $ZT=\frac{L_{12}^2}{L_{11}L_{22}-L_{12}^2}$



$$-\Delta V=S\Delta T$$

$$j^Q=\Pi j$$

$$\Pi=TS$$

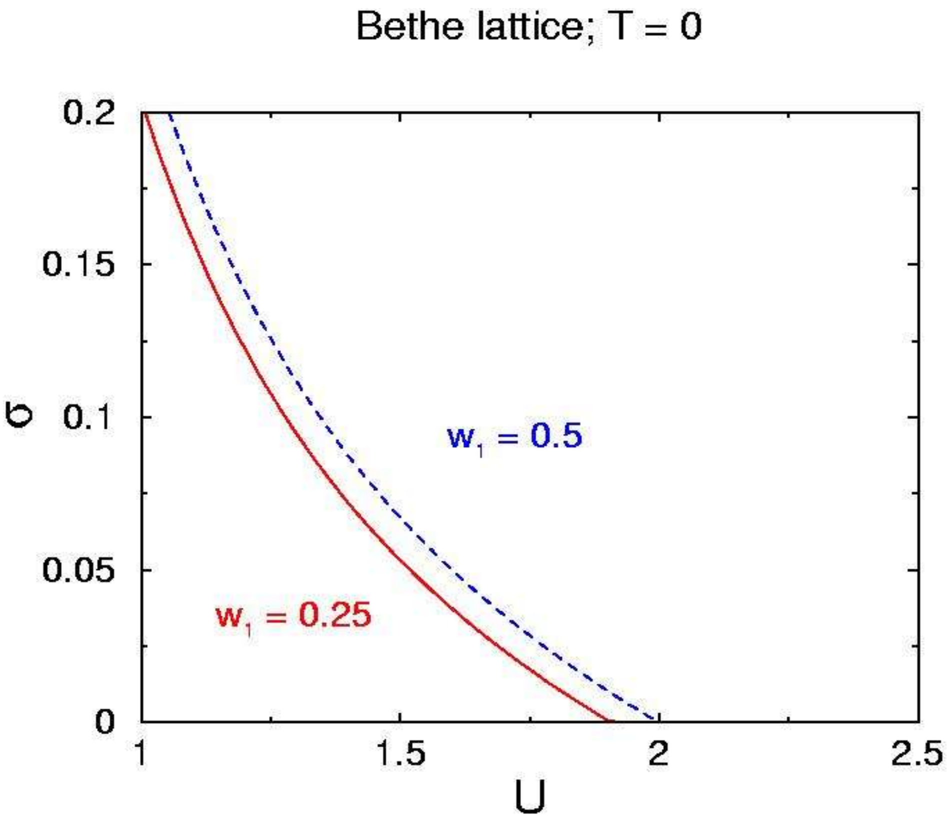
– Peltier set up

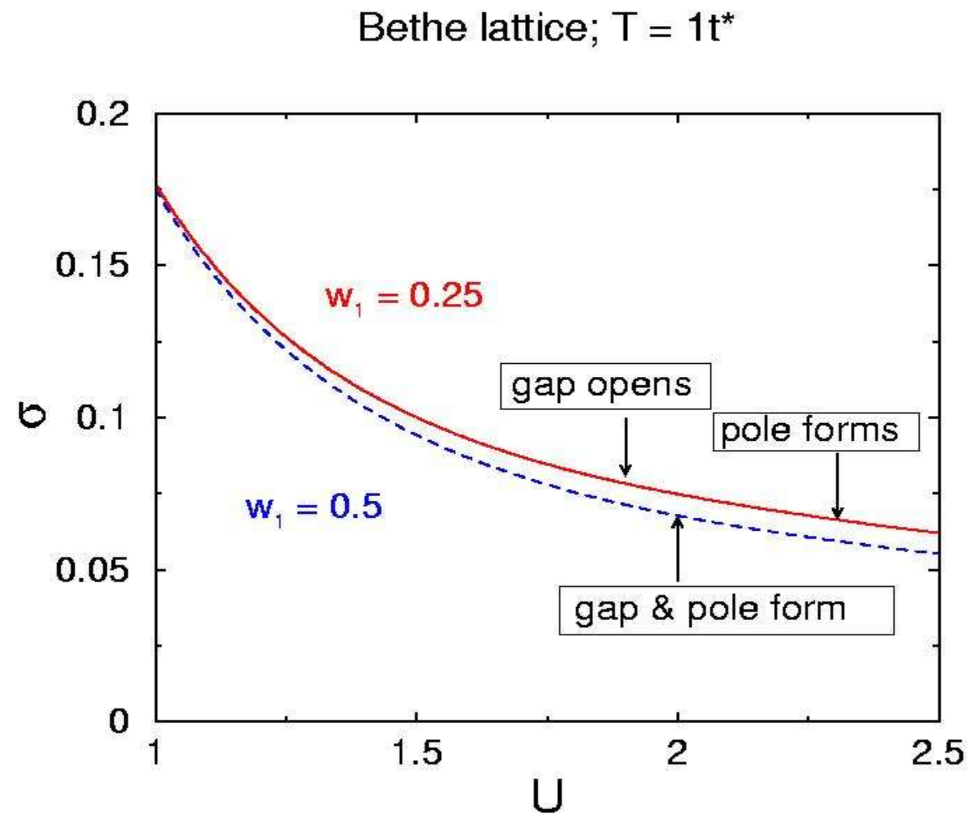
$$\vec{j}^Q=-\kappa\vec{\nabla}T$$

$ZT > 1$ commercially viable thermoelectric applications
$ZT \sim 4$ (freon refrigerators)
$ZT \to \infty$ - Carnot efficiency

[3] M. Johnson and G.D. Mahan, Phys. Rev. **B** 21, 4223 (1980)

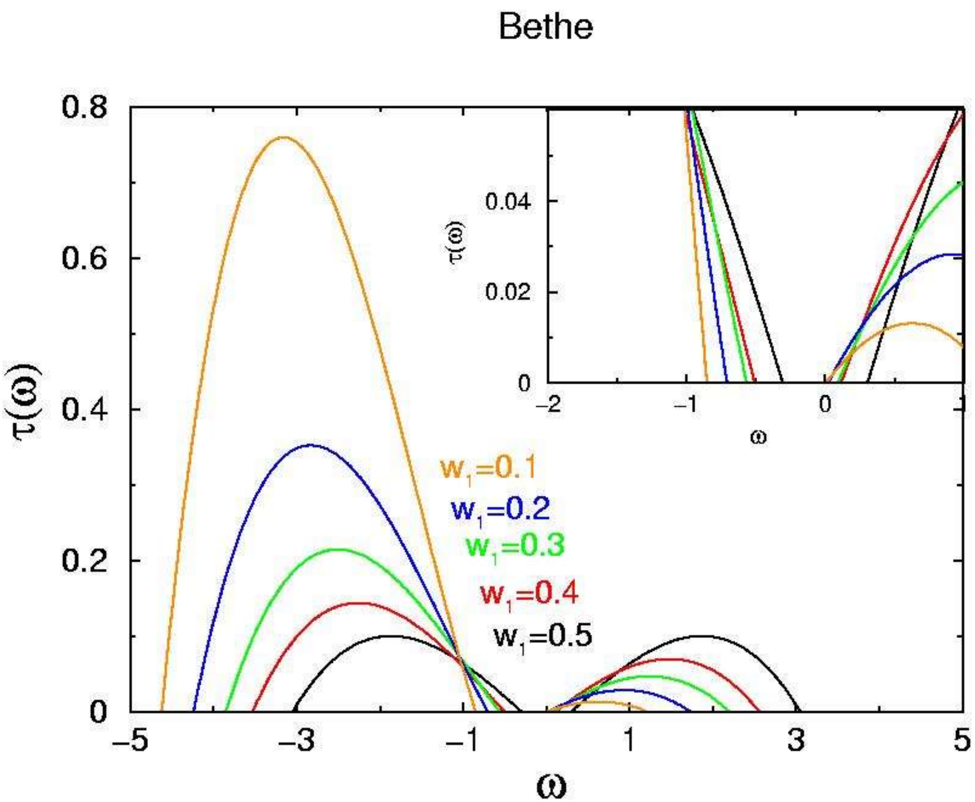
Is pole formation significant?

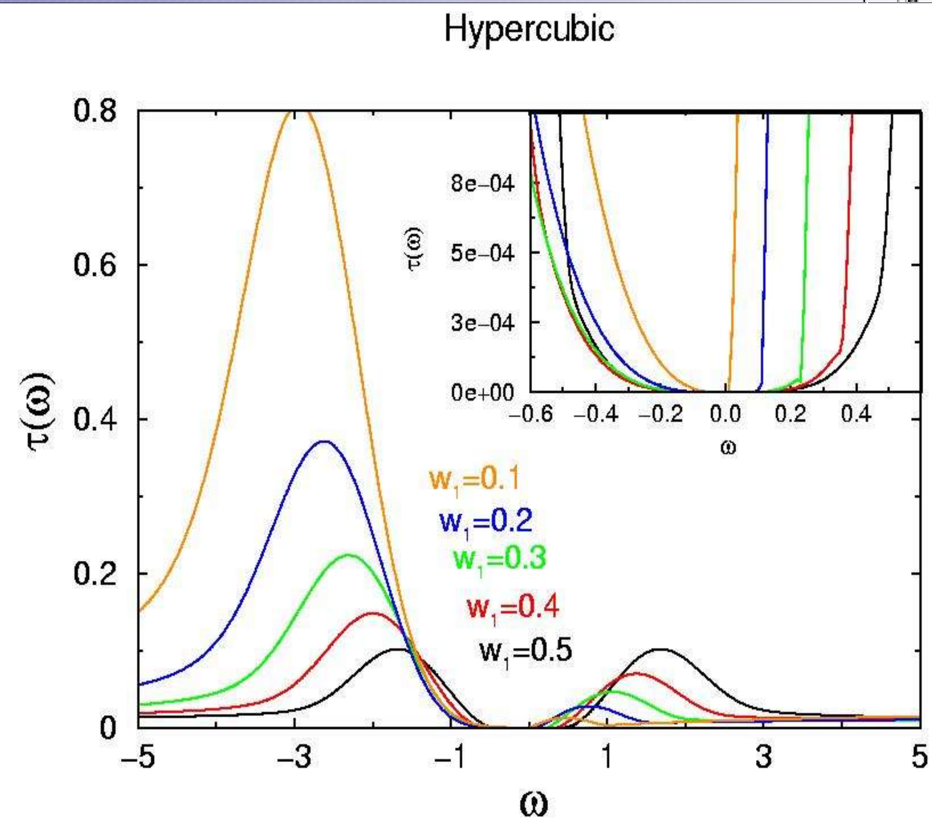




- Example: calculated dc conductivity on the Bethe lattice
- Conductivity shows continuous transition at T=0
- No influence of the pole on the transport at finite T
- Other differences between HC and Bethe lattices?

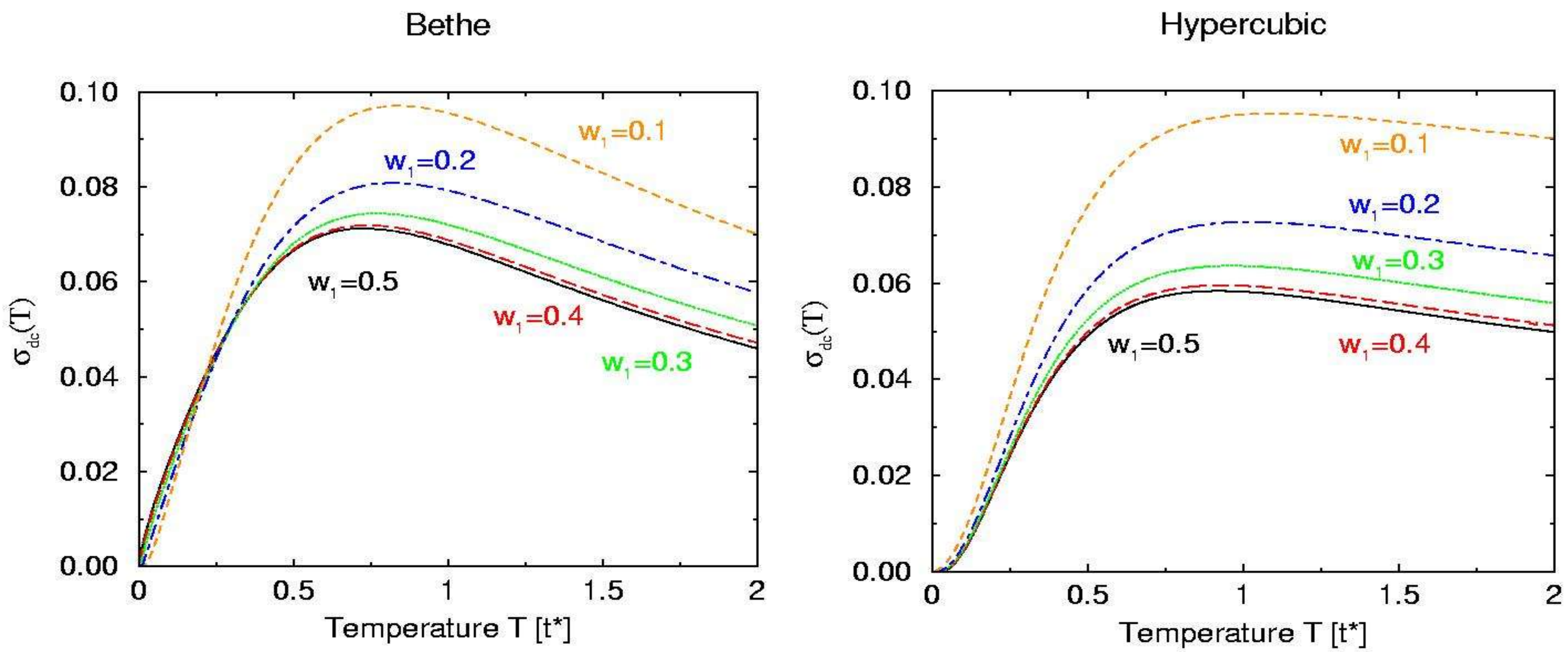
Unphysical relaxation time (HC)





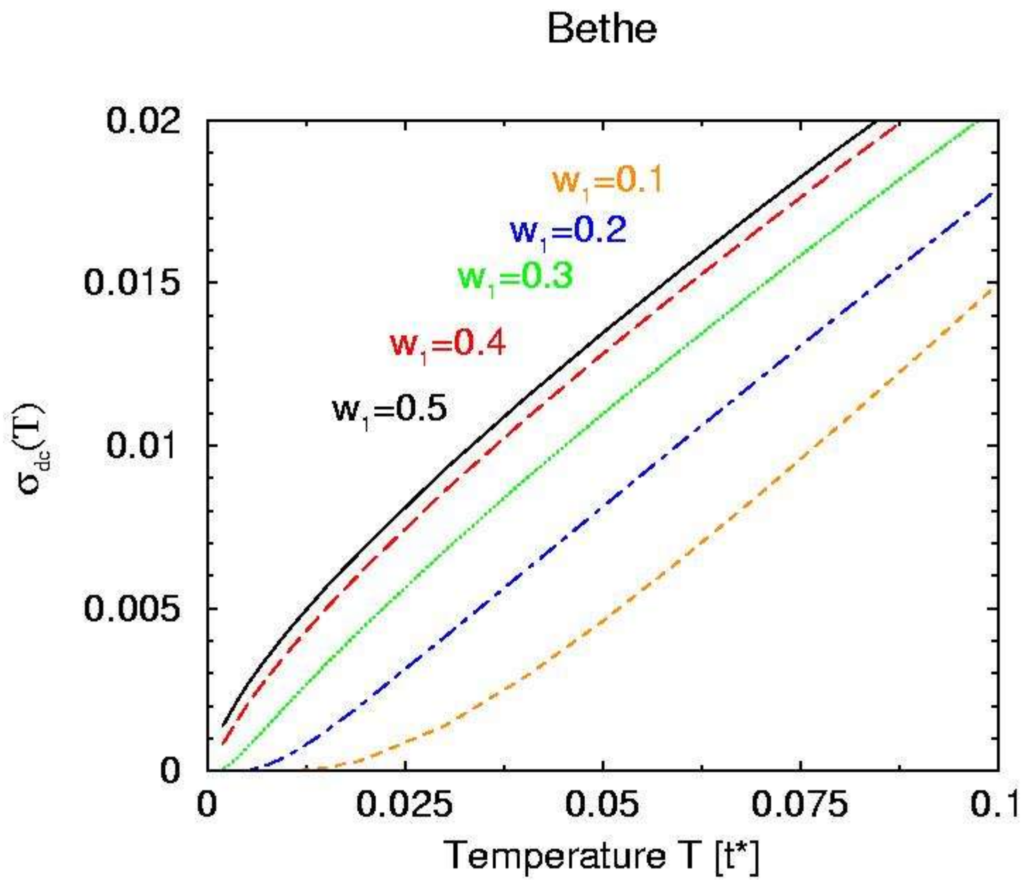
- Relaxation times for $U = 3.0$ $\quad \tau(\omega)=\int d\epsilon\, \rho(\epsilon) A^2(\omega,\epsilon)$
- Relaxation time defines behavior of L_{ij} and transport
- HC – relaxation time is finite outside the band (affects high T results)
- HC – relaxation time is power law "inside" the band gap (low T)
- HC – gap states have exponentially large lifetime, and can contribute significantly into current

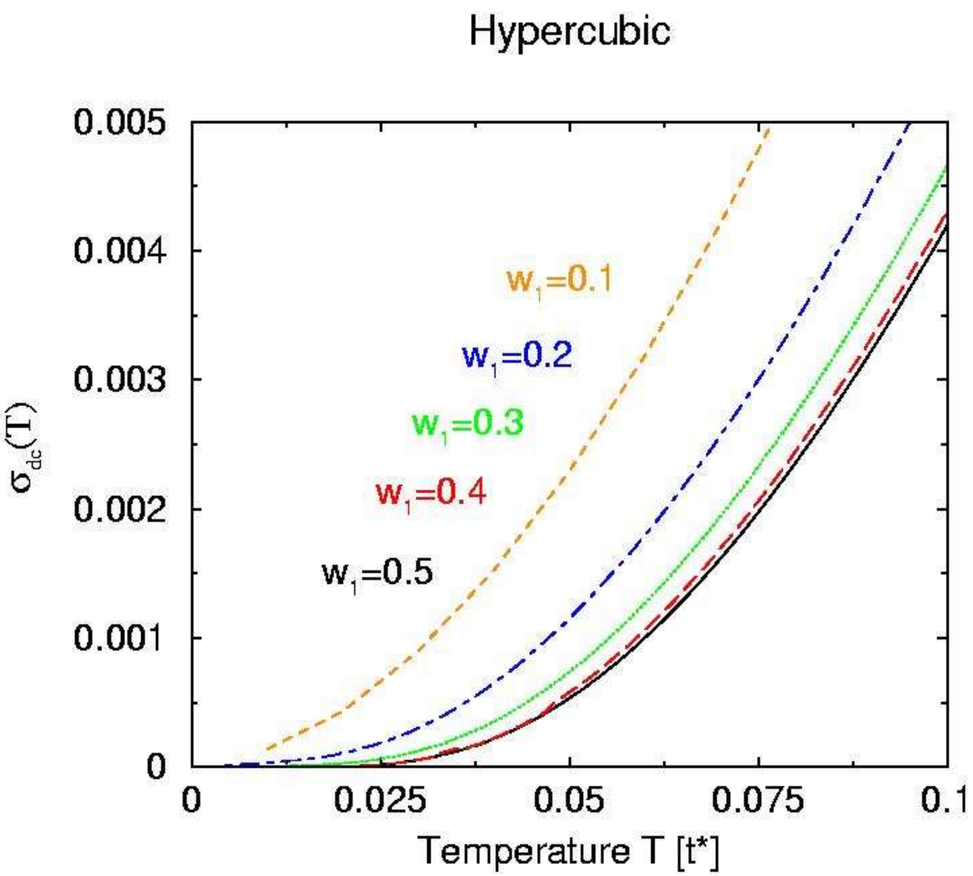
Example: bulk charge transport



- dc conductivity for $U = 2.0$
- HC and Bethe have both gap and pole for w_1=0.5, otherwise Bethe is an insulator (with no pole), HC is still a metal
- No significant differences, except low T (blow up)

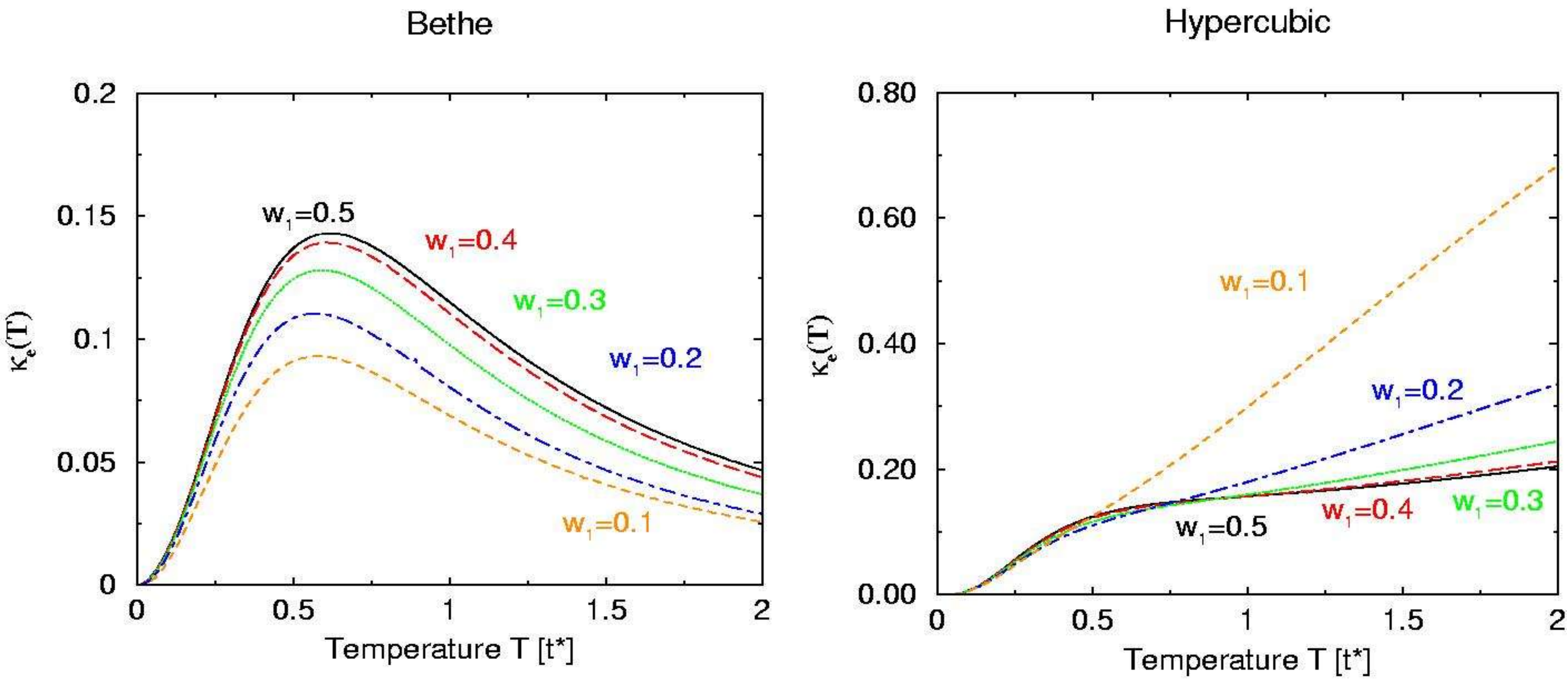
Example: bulk charge transport (low T)





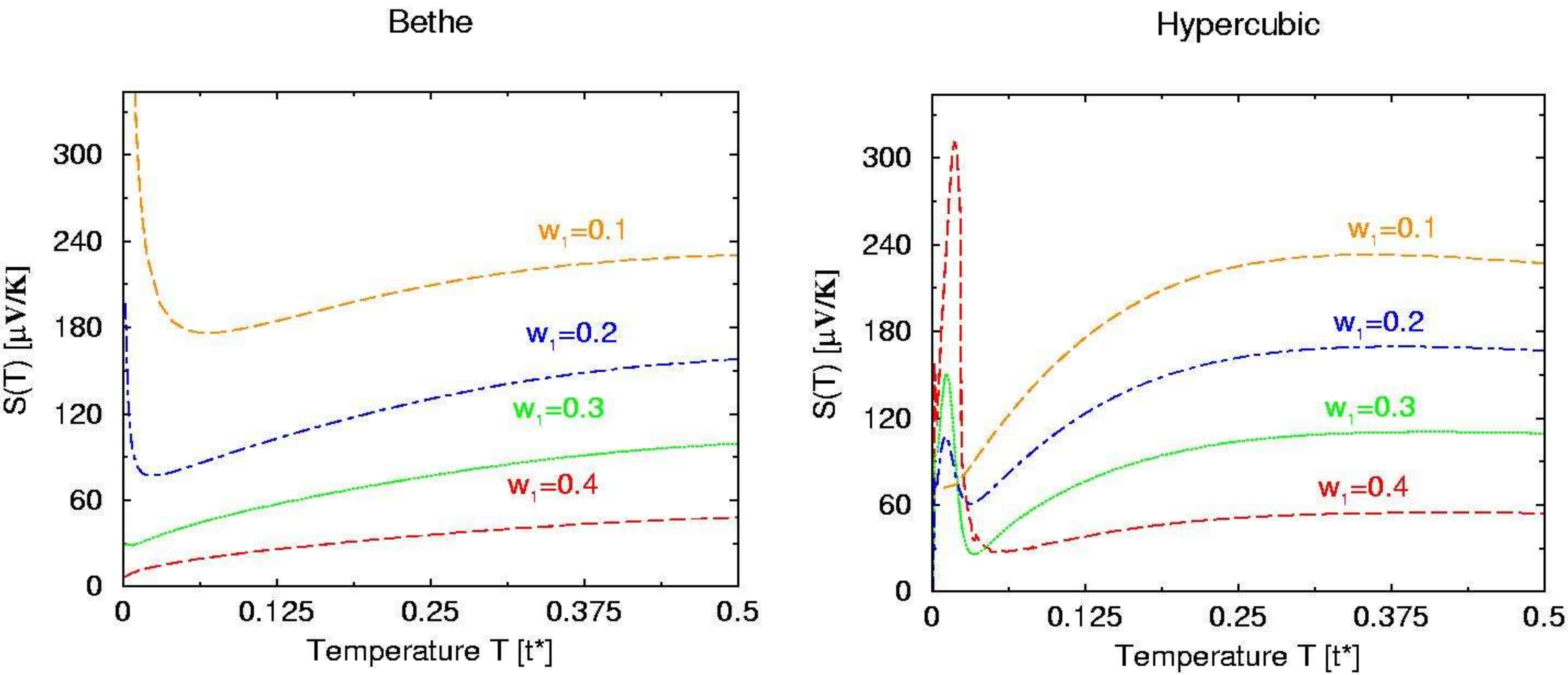
- dc conductivity for $U = 2.0$
- HC and Bethe have both gap and pole for w_1=0.5, otherwise Bethe is an insulator (with no pole), HC is still a metal
- At T=0 HC – exponentially small σ, Bethe - σ=0

Electronic thermal conductivity



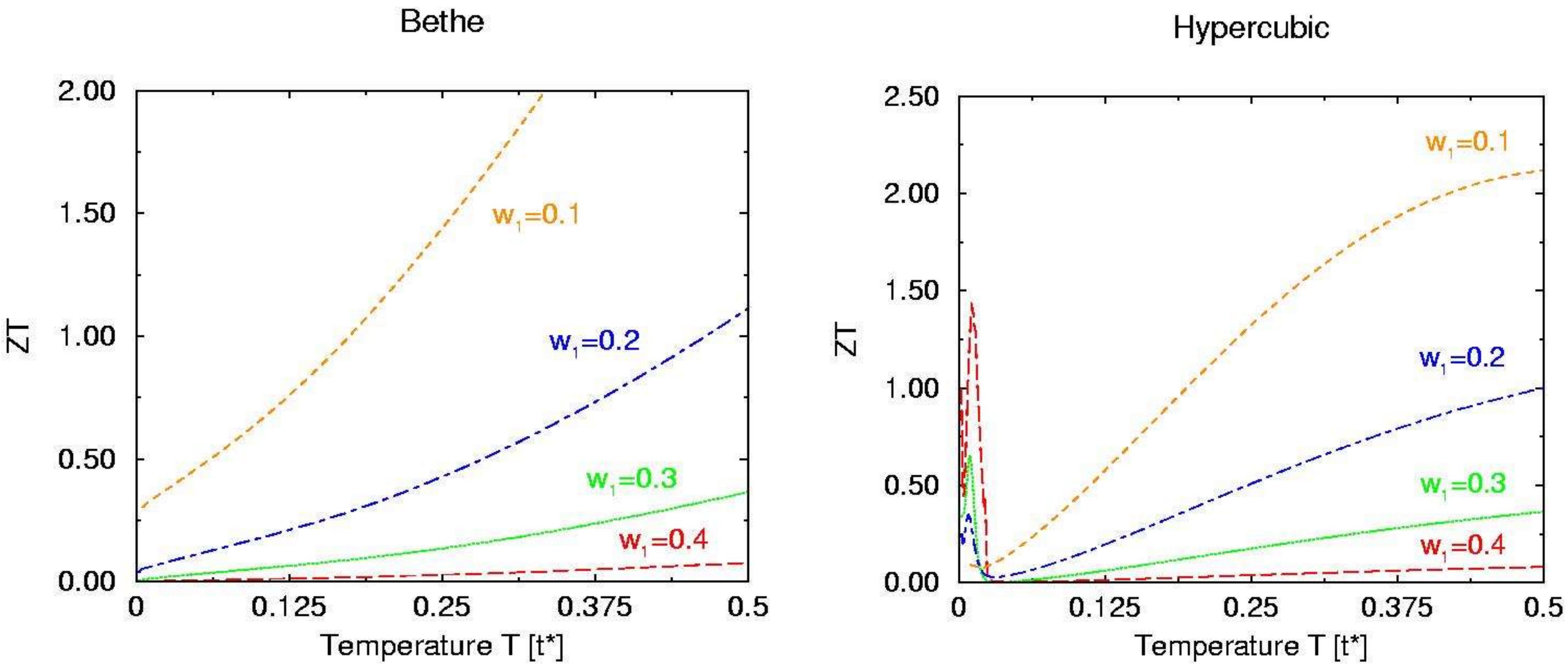
- Thermal conductivity (electronic part) for $U = 2.0$
- HC and Bethe have both gap and pole for w_1=0.5, otherwise Bethe is an insulator (with no pole), HC is still a metal
- HC is linearly increasing at high T – consequence of $\tau(\omega)$ behavior

Thermopower



- Thermopower for $U = 2.0$
- At half-filling thermopower is zero
- Bethe – S diverges at $T \rightarrow 0$ (linear response breaks down), HC – S peaks, then goes to zero as $T \rightarrow 0$
- Potential for thermoelectric applications

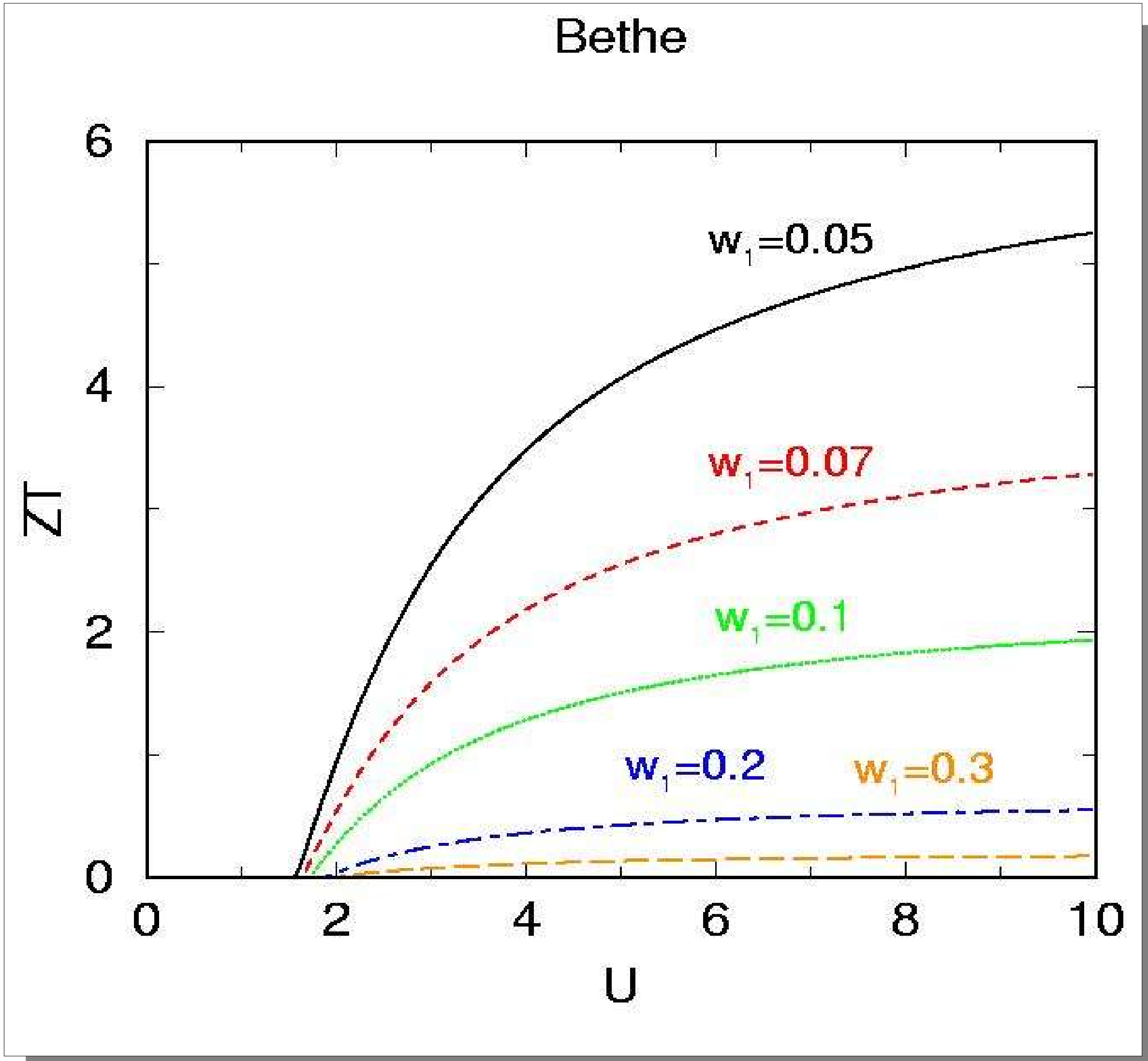
Thermoelectric figure-of-merit



- Thermoelectric figure-of-merit ($U = 2.0$), ZT > 1 needed for thermoelectric applications (freon-based refrigerators ZT~4)
- Large ZT's in both cases, different behavior, low T peak on HC
- Suggests use of correlated materials in thermoelectric devices
- How does ZT depend on U?

Zero temperature *ZT*

- Example: zero *T* *ZT* can be made arbitrarily high by increasing asymmetry
- At low *T* – small conductivity, even if *ZT* is large, high voltage would be needed to have cooling power
- Warning: at low *T* lattice component may be significant
- At high *T* lattice component is less significant, and *ZT* is still relatively high



Conclusions

- Effects of particle-hole asymmetry on the Mott transition in the infinite dimensional Falicov-Kimball model analyzed
- The scenarios of MIT are the same on hypercubic and Bethe lattices at half-filling only
- When the particle-hole symmetry is removed (as is often the case in real materials) the MIT and formation of the pole in the self-energy are unrelated on the Bethe lattice
- There is little or no difference in properties of a correlated insulator with or without a pole in the self-energy
- We conjecture that conclusions about the character of the MIT on the Bethe lattice will hold for other systems with finite bandwidth
- Transport properties also suggest that (within DMFT) the Bethe lattice is more realistic than hypercubic lattice
- Our calculations suggest that particle-hole asymmetric correlated materials may be useful in thermoelectric devices