Chaire de Physique de la Matière Condensée

(Some) Thermoelectric Properties of `Strongly-Correlated' Quantum Systems

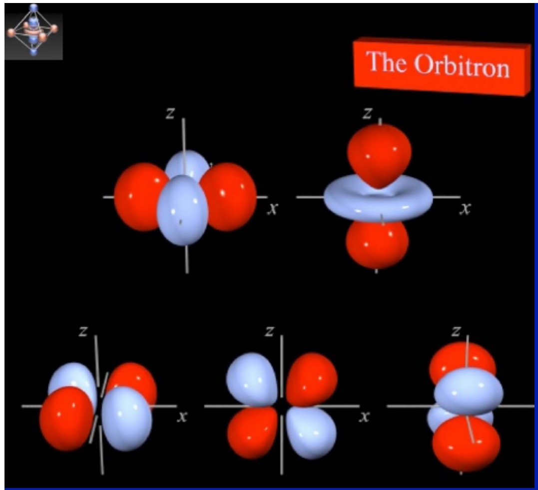
Antoine Georges – Cargèse 2012

OUTLINE

- I. Thermopower S(T) of materials/models with strong electron correlations
- 1. Brief introduction to strongly correlated materials
- 2. Key questions/puzzles about transport and thermopower in oxides
- 3. Some formalism: Kubo formula, DMFT
- 4. S(T) in Fermi-Liquid regime
- 5. S(T) in `bad metal' regime: Heikes and beyond
- [II. A proposal to investigate thermoelectric effects with ultra-cold atomic gases (C.Grenier, C.Kollath & AG arXiv:1209.3942)]

1. Materials with strong electron correlations: some examples

(geared at thermoelectrics)

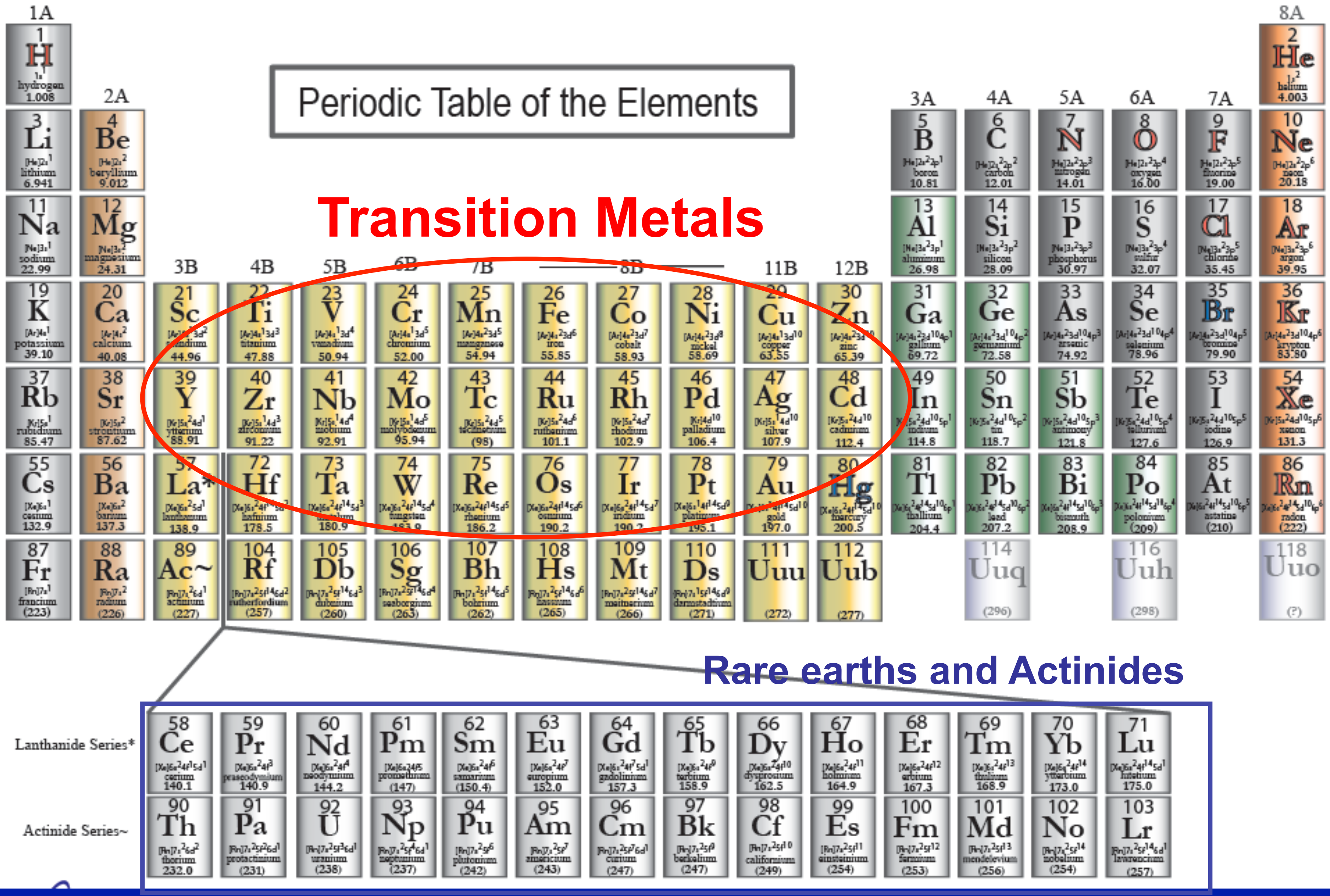


Strongly correlated materials: ***the suspects are the localized orbitals !***

* d- or f- orbitals are quite close to nucleus
(particularly 3d and 4f, for orthogonality reasons)

They do not behave as regular band-forming orbitals (e.g sp-bonding) and retain atomic-like aspects
→ Electrons "hesitate" between localized and itinerant behaviour !

Materials: transition-metals and their oxides, rare-earth/actinides and their compounds, but also some organic materials

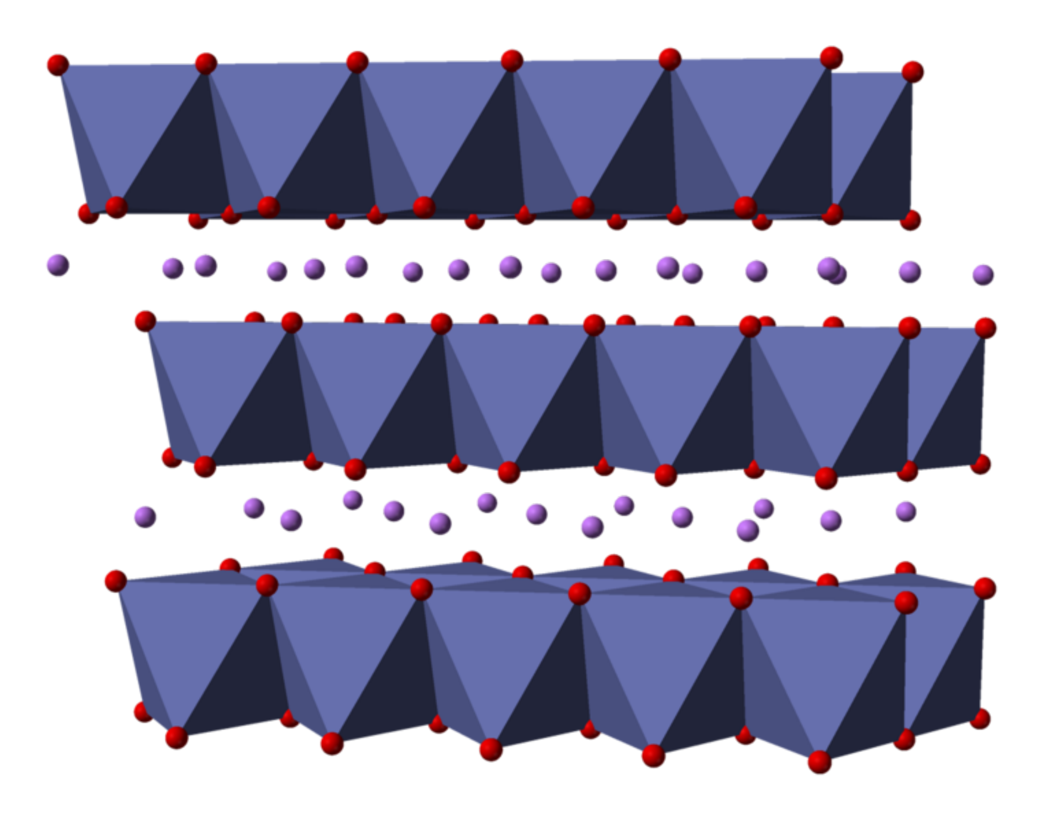

Periodic Table of the Elements
Transition Metals
Rare earths and Actinides
1A
2A
3B
4B
5B
6B
7B
8B
11B
12B
3A
4A
5A
6A
7A
8A
1 H hydrogen 1.008
2 He helium 4.003
3 Li lithium 6.941
4 Be beryllium 9.012
5 B boron 10.81
6 C carbon 12.01
7 N nitrogen 14.01
8 O oxygen 16.00
9 F fluorine 19.00
10 Ne neon 20.18
11 Na sodium 22.99
12 Mg magnesium 24.31
13 Al aluminum 26.98
14 Si silicon 28.09
15 P phosphorus 30.97
16 S sulfur 32.07
17 Cl chlorine 35.45
18 Ar argon 39.95
19 K potassium 39.10
20 Ca calcium 40.08
21 Sc 44.96
22 Ti titanium 47.88
23 V vanadium 50.94
24 Cr chromium 52.00
25 Mn manganese 54.94
26 Fe iron 55.85
27 Co cobalt 58.93
28 Ni nickel 58.69
29 Cu copper 63.55
30 Zn zinc 65.39
31 Ga gallium 69.72
32 Ge germanium 72.58
33 As arsenic 74.92
34 Se selenium 78.96
35 Br bromine 79.90
36 Kr krypton 83.80
37 Rb rubidium 85.47
38 Sr strontium 87.62
39 Y yttrium 88.91
40 Zr zirconium 91.22
41 Nb niobium 92.91
42 Mo molybdenum 95.94
43 Tc technetium (98)
44 Ru ruthenium 101.1
45 Rh rhodium 102.9
46 Pd palladium 106.4
47 Ag silver 107.9
48 Cd cadmium 112.4
49 In indium 114.8
50 Sn tin 118.7
51 Sb antimony 121.8
52 Te tellurium 127.6
53 I iodine 126.9
54 Xe xenon 131.3
55 Cs cesium 132.9
56 Ba barium 137.3
57 La* lanthanum 138.9
72 Hf hafnium 178.5
73 Ta 180.9
74 W tungsten
75 Re rhenium 186.2
76 Os osmium 190.2
77 Ir iridium
78 Pt platinum
79 Au gold 197.0
80 Hg mercury 200.5
81 Tl thallium 204.4
82 Pb lead 207.2
83 Bi bismuth 208.9
84 Po polonium (209)
85 At astatine (210)
86 Rn radon (222)
87 Fr francium (223)
88 Ra radium (226)
89 Ac~ actinium (227)
104 Rf rutherfordium (257)
105 Db dubnium (260)
106 Sg seaborgium (263)
107 Bh bohrium (262)
108 Hs hassium (265)
109 Mt meitnerium (266)
110 Ds darmstadtium (271)
111 Uuu (272)
112 Uub (277)
114 Uuq (296)
116 Uuh (298)
118 Uuo (?)
Lanthanide Series*
58 Ce cerium 140.1
59 Pr praseodymium 140.9
60 Nd neodymium 144.2
61 Pm promethium (147)
62 Sm samarium (150.4)
63 Eu europium 152.0
64 Gd gadolinium 157.3
65 Tb terbium 158.9
66 Dy dysprosium 162.5
67 Ho holmium 164.9
68 Er erbium 167.3
69 Tm thulium 168.9
70 Yb ytterbium 173.0
71 Lu lutetium 175.0
Actinide Series~
90 Th thorium 232.0
91 Pa protactinium (231)
92 U uranium (238)
93 Np neptunium (237)
94 Pu plutonium (242)
95 Am americium (243)
96 Cm curium (247)
97 Bk berkelium (247)
98 Cf californium (249)
99 Es einsteinium (254)
100 Fm fermium (253)
101 Md mendelevium (256)
102 No nobelium (254)
103 Lr lawrencium (257)

Some Transition-Metal Oxides which may be useful Thermoelectrics

Cobalt oxides: Na_xCoO_2 Terasaki et al. PRB 1997

Na ions in between layers →

Triangular CoO_2-layers →



Note similarities to $LiCoO_2$ batteries

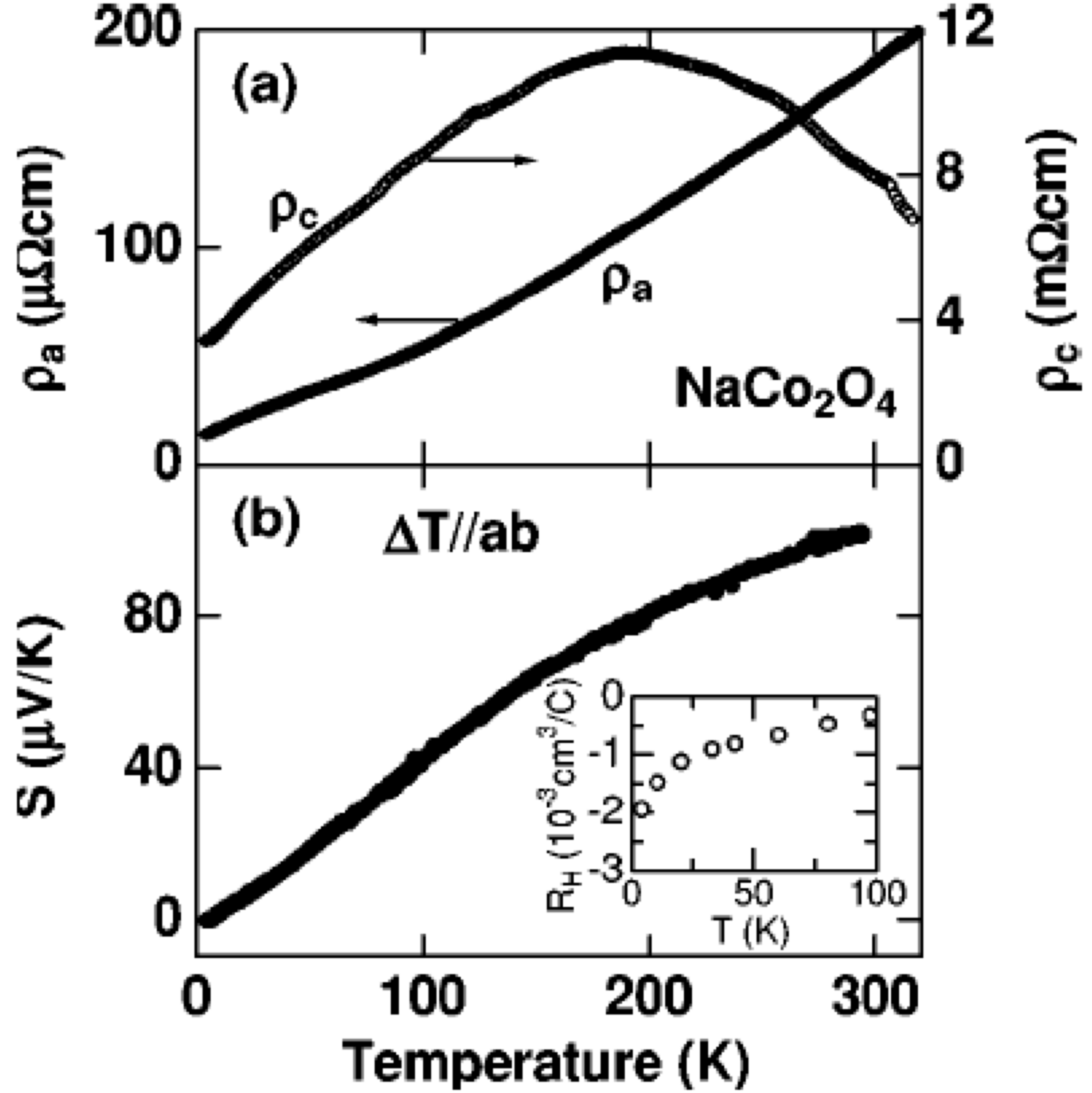


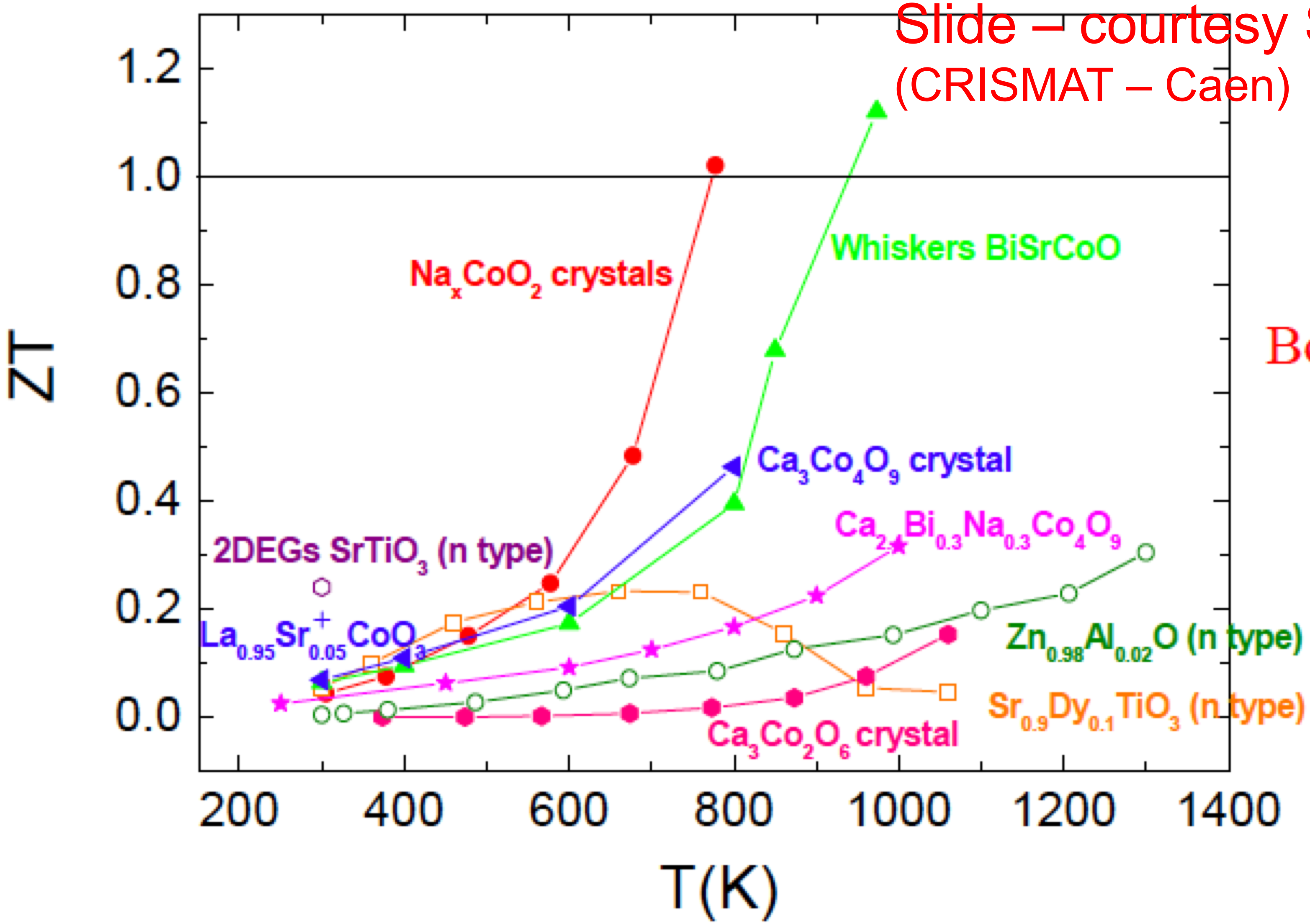
FIG. 2. (a) In-plane (ρ_a) and out-of-plane (ρ_c) resistivity of $NaCo_2O_4$ single crystals. (b) In-plane thermoelectric power (S) of $NaCo_2O_4$ single crystal. The inset shows the in-plane Hall coefficient (R_H) of $NaCo_2O_4$ single crystal.

TABLE I. Various physical parameters for $NaCo_2O_4$ and Bi_2Te_3 (Ref. 6) at 300 K. ρ, S, and μ are resistivity, thermoelectric power, and mobility, respectively. Note that ρ and S of $NaCo_2O_4$ are the in-plane data.

Parameters	Unit	$NaCo_2O_4$	Bi_2Te_3
ρ	mΩ cm	0.2	1
$\|S\|$	μV/K	100	200
S^2/ρ	μW/K^2 cm	50	40
μ	cm^2/V s	13	150

Performances thermoélectriques des oxydes

Slide – courtesy Sylvie Hebert
(CRISMAT – Caen)



Oxydes :
Bonne stabilité
chimique
à haute T
et sous air

NaxCoO2 _ Fujita : JJAP 40, 4644 (2001); SrTiO3 _ Muta : J. Alloys and compounds 350, 292 (2003); Ca2.4Bi0.3Na0.3Co4O9 _ Xu : APL80, 3760 (2002); Whiskers BiSrCoO _ Funahashi : APL81, 1459 (2002); Ca3Co2O6 _ Mikami : JAP94, 10 (2003); 2DEGs(SrTiO3) _ Ohta : Nature Materials 6, 129 (2007); Ca3Co4O9 crystal _ Shikano : APL 82, 1851 (2003); LaSrCoO _ Androulakis : APL84, 1099 (2004); ZnAlO _ Ohtaki : JAP79, 1816 (1996)

WHY ?

- Oxides have spin and orbital degrees of freedom
- Hence possibly large entropy !
- However : at low-T in Fermi Liquid regime, S(T) constrained by Pauli principle S(T) ~ T → small
- Need to beat Pauli principle: heat above `quantum degeneracy'
- BUT in such a way that i) resistivity does not become too large and ii) avoid phases with spin/ orbital long-range order (frustration helps)

Hence, fundamental issue:

Understand crossover
from the low-T Fermi-Liquid regime
all the way to
the hi-T `Bad Metal' regime where
quasiparticle (QP) excitations no longer exist

In the following, address this issue in two ways:

- Illustrate by some experimental results on oxides
- Discuss a model calculation: hole-doped single-band Hubbard model (doped Mott insulator)

2. Illustrate this on THE best documented oxide: Sr_2RuO_4

`The Helium 3 of transition-metal oxides !'

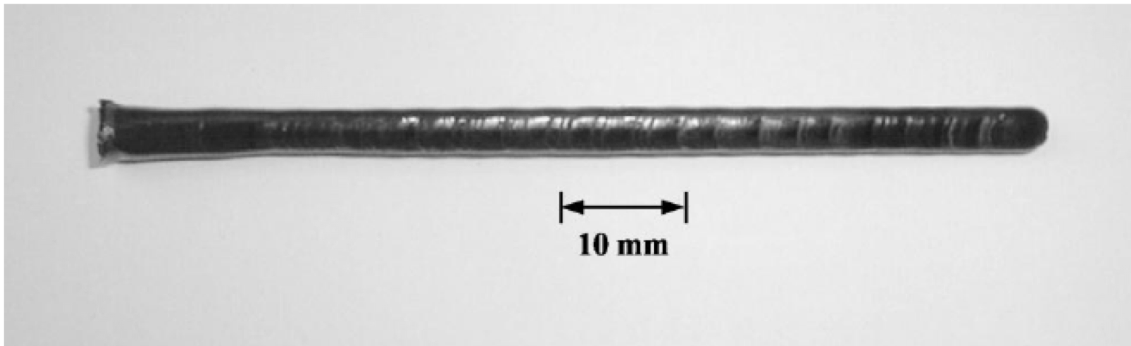
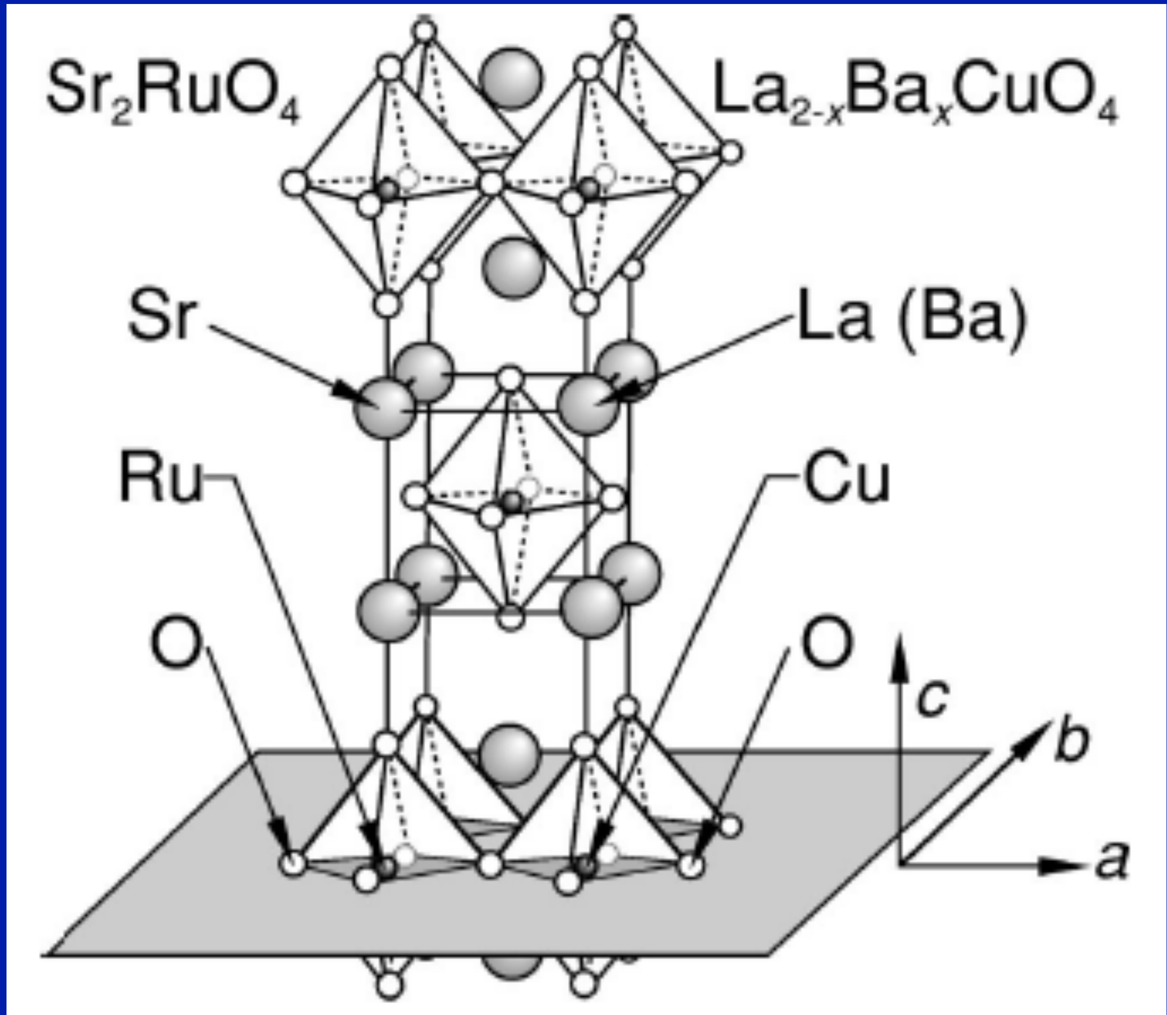


FIG. 4. A single crystal of Sr_2RuO_4 (Mao, Maeno, and Fukuzawa, 2000).

Review: McKenzie and Maeno, Rev Mod Phys 75, 657 (2003)

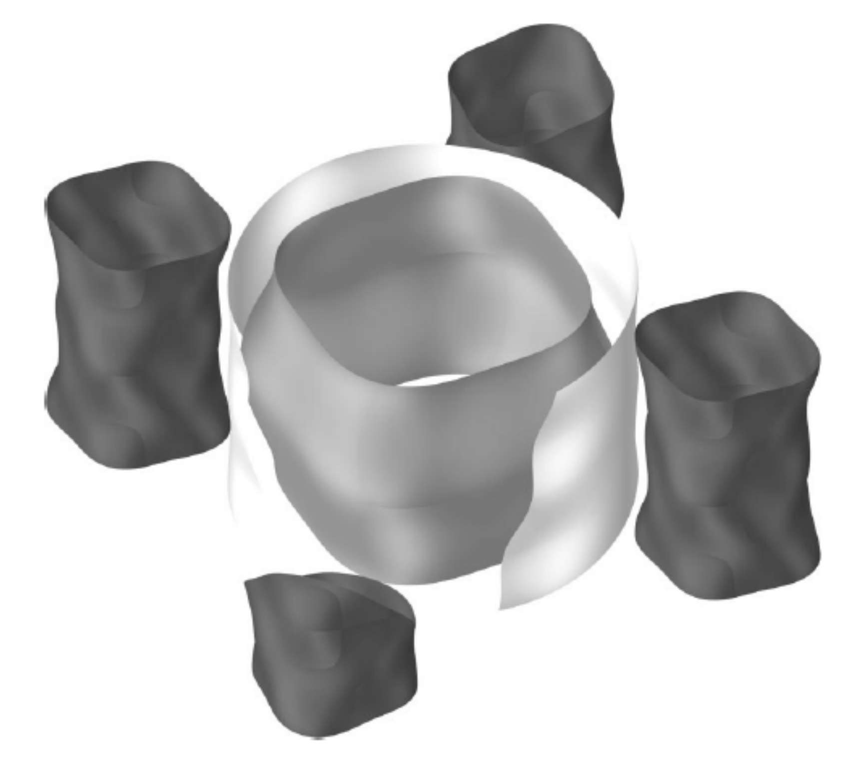
- Has been investigated with basically all techniques in the experimentalist's toolbox
- 4d-row structural analogue of La_2CuO_4



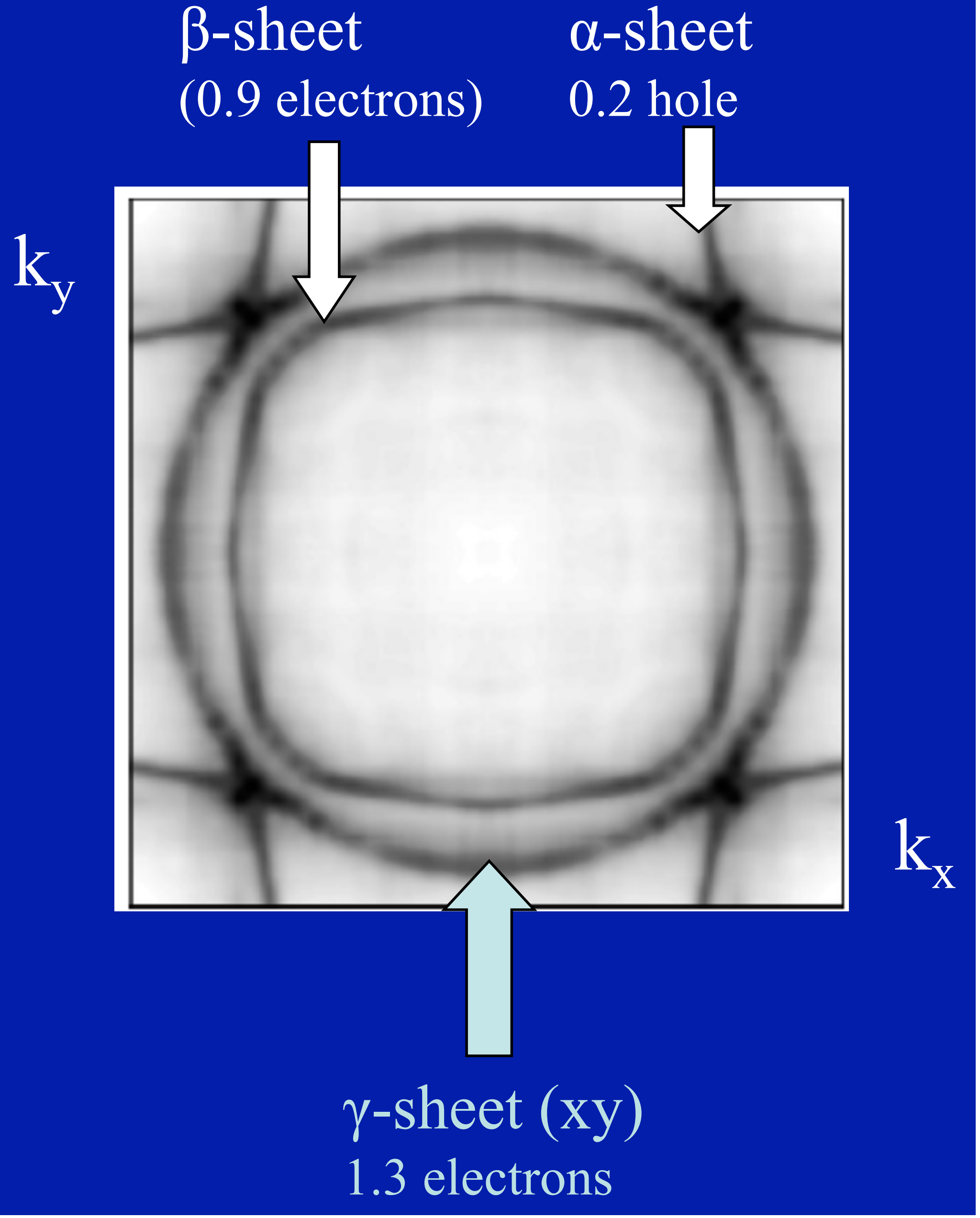
Basic electronic structure of Sr_2RuO_4

Fermi surface and orbital populations from dHvA:

FS from quantum oscillations:



FS from photoemission:

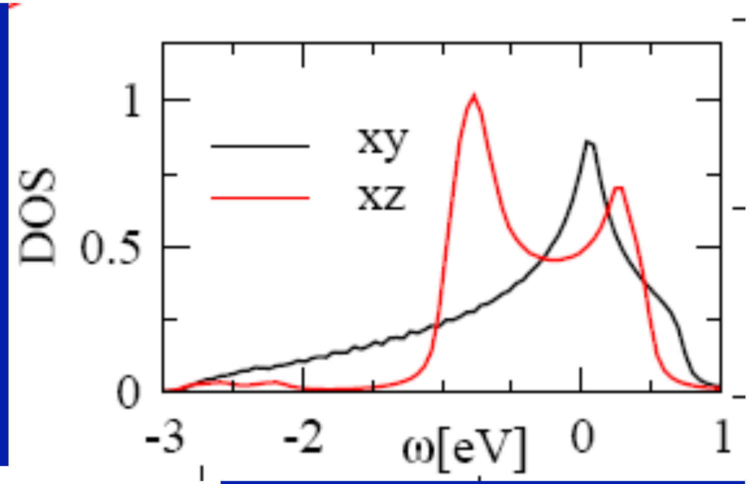


Bands (DFT-LDA)

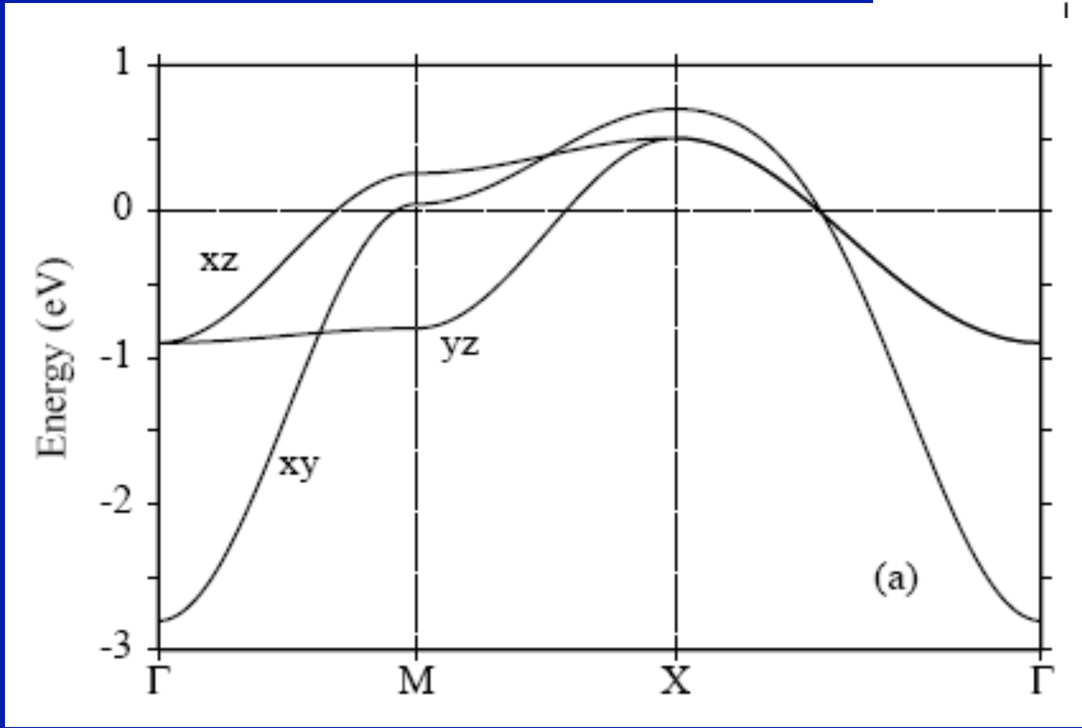
4 electrons

in t_{2g} ={dxy,dyz,dxz}

subset of d-orbitals



~ 1.5 eV
(xz,yz)



~ 3.6 eV
(xy band)

But kinetic energies of all bands comparable

Low-T state of ruthenates: a Fermi liquid

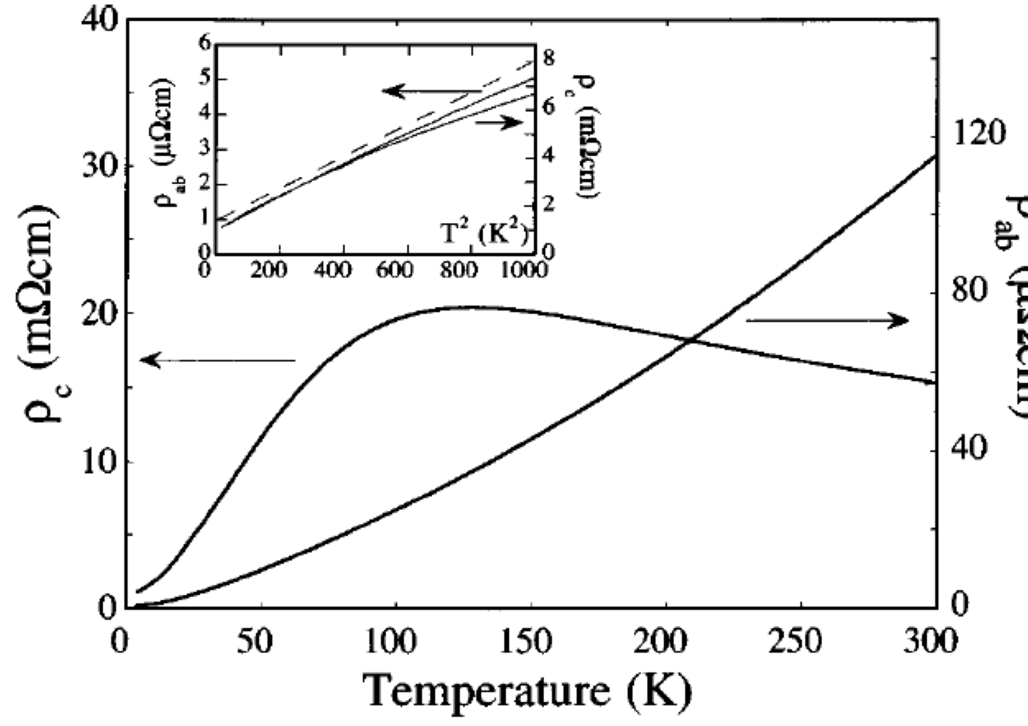
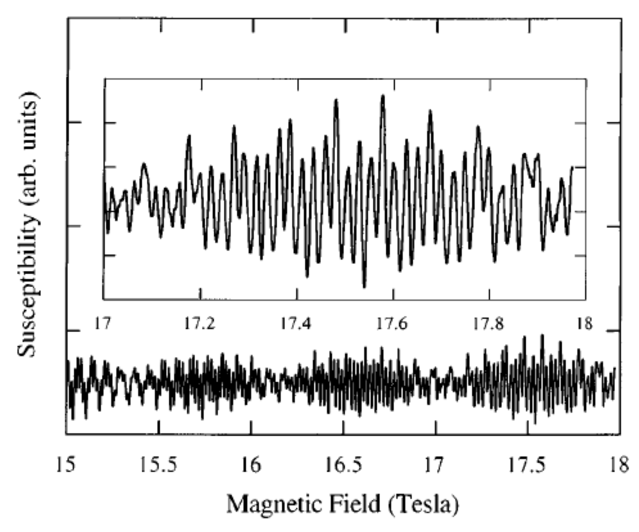


FIG. 1. Zero-field $\rho_{ab}(T)$ and $\rho_c(T)$ of Sr_2RuO_4. The inset shows $\rho_c(T)$ and $\rho_{ab}(T)$ below 32 K plotted against T^2. The dashed line is a guide to the eye.



~ T^2 up to about T_{FL} ~ 20K

When is a quasiparticle description legitimate ?
Ioffe-Regel [1960], Mott [1972]

→ When the mean-free path of the charge carriers is larger than ~ the Fermi wavelength

$$l = v_F \tau_D \quad , \quad v_F = \frac{\hbar k_F}{m} \quad , \quad \sigma_{dc} = \frac{e^2}{\hbar} \frac{n}{k_F} l$$

Quasi-2D (layered) geometry:

$$n = \frac{N}{\Omega} = 2.\frac{1}{8\pi^3}.\pi k_F^2 \frac{2\pi}{c_0}$$

c_0: c-axis lattice spacing

3D isotropic geometry: $$n = 2.\frac{1}{8\pi^3}.\frac{4}{3}\pi k_F^3$$

MIR (cont'd):

Quasi 2D: $$\boxed{\sigma_{dc} = \frac{e^2}{\hbar}\frac{1}{c_0}\frac{k_F l}{2\pi}}$$

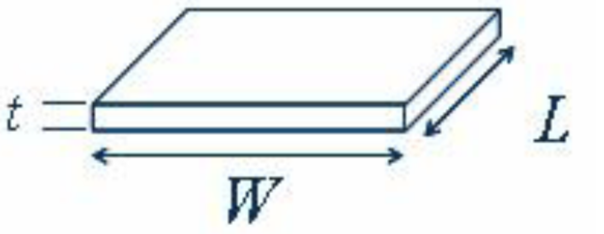
3D isotropic: $$\sigma_{dc} = \frac{e^2}{\hbar}\frac{1}{3\pi^2}k_F^2 l$$

MIR criterion – 2D – 1FS sheet

$$k_F l = 1 \rightarrow \rho_M = \frac{h}{e^2}c_0 = 0,25\,\mathrm{m}\Omega.\mathrm{cm} \times c_0[\mathrm{nm}]$$

MIR limit corresponds to sheet resistance = resistance h/e^2 quantum per layer

$$R = \rho\frac{L}{tW} = R_s\frac{L}{W}$$



Higher-T regime: gradual crossover to `bad metal'

ab-plane:

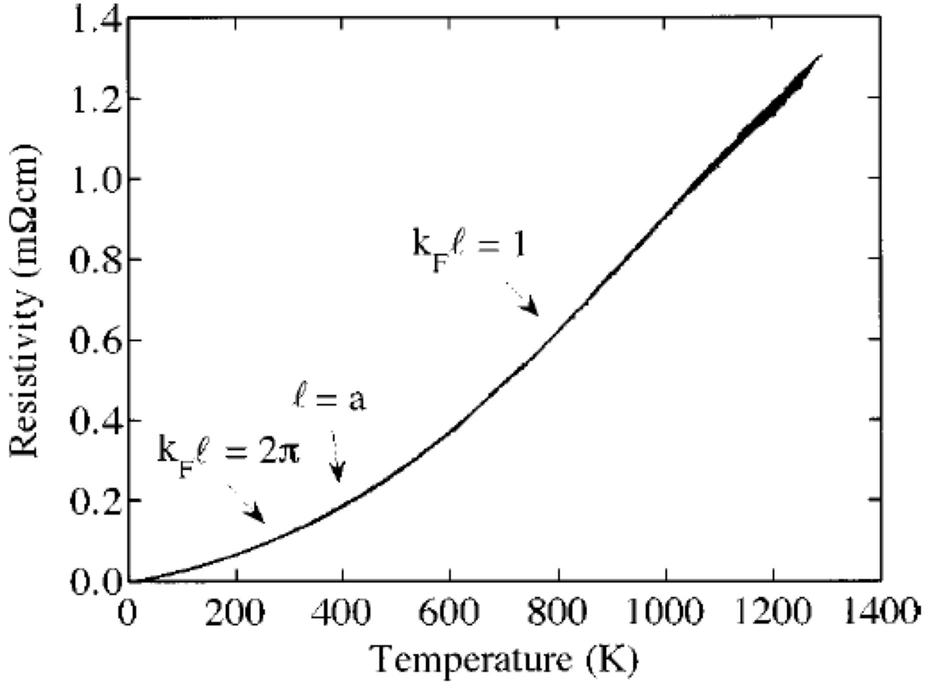


FIG. 1. The in-plane resistivity of Sr_2RuO_4 from 4 to 1300 K. Three criteria for the Mott-Ioffe-Regel limit are marked on the graph, and there is no sign of resistivity saturation, so Sr_2RuO_4 is a "bad metal" at high temperatures, even though it is known to be a very good metal at low temperatures.

Resistivity
does cross MIR value

Nothing dramatic is seen
in ρ upon crossing MIR

$\rho \ll \rho_M$ at $T \sim T_{FL}$
Hence large regime of T
with non-T^2 (non FL) transport
but still `good' metal

Tyler, Maeno, McKenzie
PRB 58 R10107 (1998)

Thermopower of Sr_2RuO_4

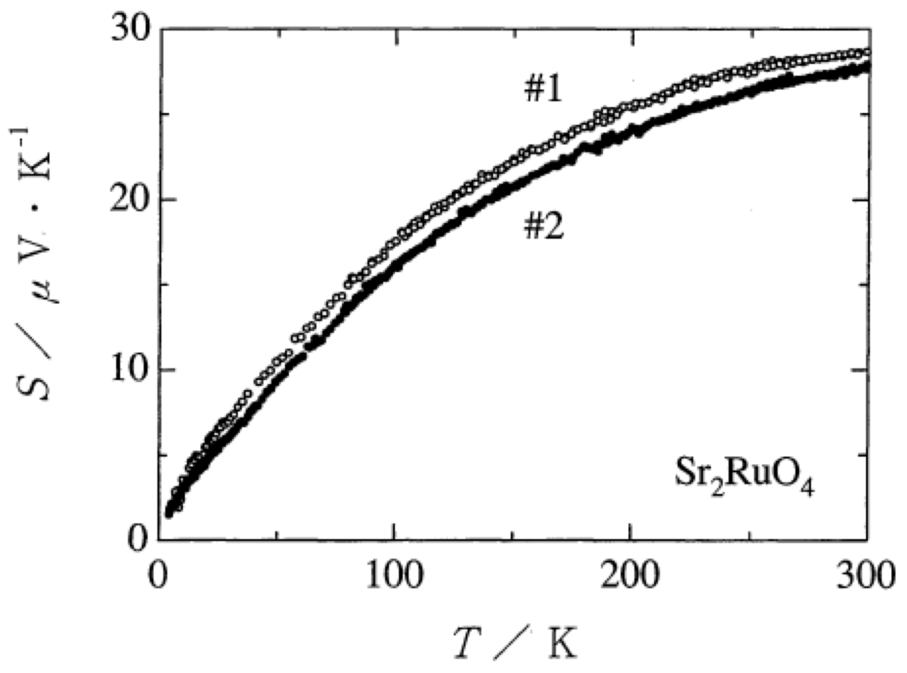


Fig. 1. Thermopower of Sr_2RuO_4 measured along the two-dimensional *ab* plane. Open and closed circles are for samples 1 and 2, respectively.

< 300K: Yoshino et al, 1996

Keawprak et al, 2008
300K < T < 1000K

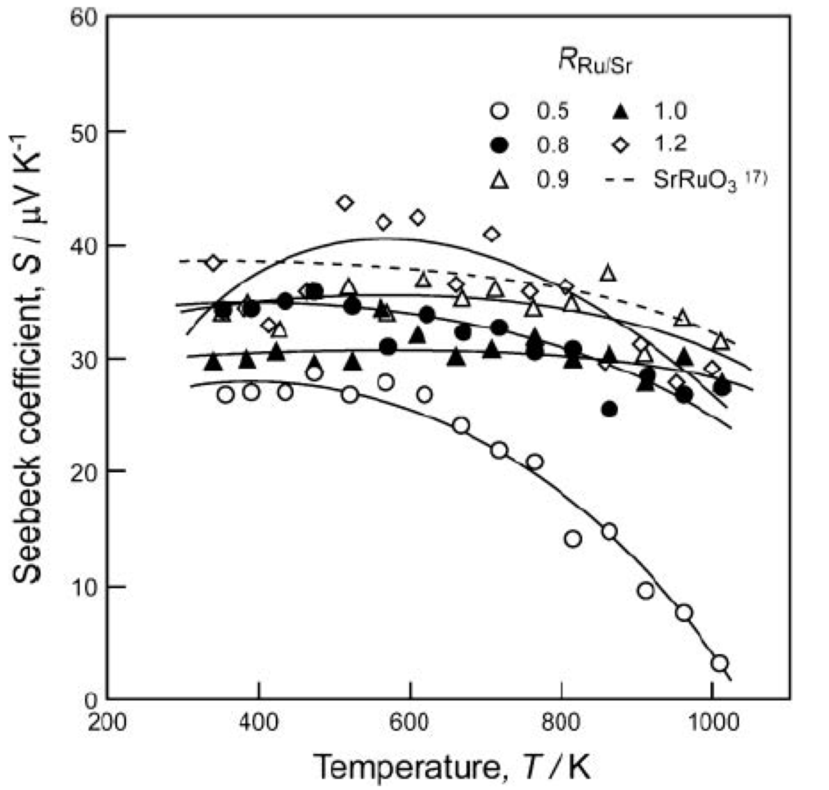


Fig. 8 Temperature dependence of Seebeck coefficient of Sr-Ru-O compounds.

Summary: often observed facts in SCES (e.g. oxides) and Questions

- Fermi Liquid behavior applies only below a low scale T_{FL}, much smaller than bare electronic scales. In this regime $S(T) \sim T$

Question: what controls slope of T-linear term ?

- Resistivity at high temperature T can reach large values, in excess of the Mott-Ioffe-Regel (MIR) value → `bad-metal' behavior at hi-T ($T>T_{MIR}$)

Questions: Are QPs gone then ? Thermopower at hi-T ?

- Intermediate regime $T_{FL}<T<T_{MIR}$

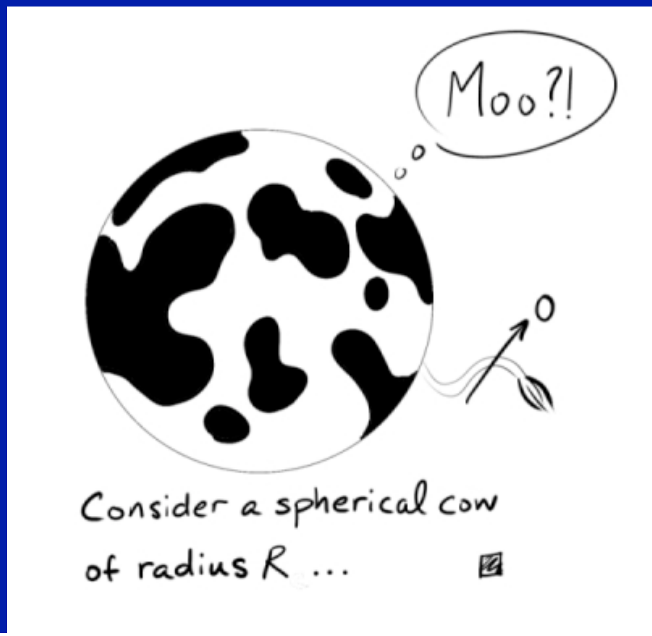
QPs but not Landau ?

Can complex T- dependence of S(T) be calculated understood ?

3. Answers to these questions in `simple' model case

Single-band Hubbard model, doped, large U/t
i.e. hole-doped Mott insulator

$$H = -t \sum_{\langle ij \rangle} (c_i^\dagger c_j + h.c.) + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



Key players

[X.Deng, J.Mravlje, R.Zitko; with : M.Ferrero, G.Kotliar, AG]

arXiv:1210.1769

Xiaoyu Deng, Jernej Mravlje, Rok Zitko

École Polytechnique, College de France, J.Stefan Institute

Thanks also to: Nigel Hussey, Sriram Shastry, C.Berthod, Dirk van der Marel

Theoretical Formalism

- Kubo formula
- Many-body effects: non-perturbative calculation of self-energy Σ using Dynamical Mean-Field Theory (DMFT)
- Σ(ω) is energy-dependent but k-independent (local) → vertex corrections can be neglected
- Numerical Renormalisation Group a la Wilson and/or Quantum Monte Carlo to obtain Σ(ω)

Seebeck from Kubo :

Relating entropy current to energy current:

$$Tds = dE - \mu dn \Rightarrow Tj_s = j_E - \mu j_n$$

Using particle & energy densities and equations of motion:

$$j_n = \sum_{kq\sigma} v_k c^\dagger_{k\sigma} c_{k+q\sigma}$$

$$j_E = \sum_{kq\sigma} v_k \frac{\partial c^\dagger_{k\sigma}}{\partial \tau} c_{k+q\sigma}$$

As before for conductivity, relate transport coefficients to correlators <j j>, <j j_E>, <j_E j_E>

At the end of the day…

Conductivity: Seebeck:

$$\sigma_{dc} = e^2 \beta A_0 \ , \quad S = -\frac{k_B}{e}\frac{A_1}{A_0}$$

$$A_n = \frac{2\pi}{\hbar}\int d\omega\,(\beta\omega)^n f(\omega) f(-\omega) \int d\epsilon\,\Phi(\epsilon) A(\epsilon,\omega)^2$$

Two key observations:

-The Seebeck is a RATIO, such as R_H . Scattering is not requested to get a non-zero S (although scattering rate does not entirely cancel, actually – see below) . In other words: a uniform entropy current can exist without entropy production (ds/dt=0)

- The Seebeck coefficient involves an odd moment (A_1) <u>and hence is very sensitive to the particle-hole asymmetry</u>

In these expressions:

- Transport function contains information about **BARE** velocities:

$$\Phi_{\mu\nu}(\epsilon) = \int_{\mathrm{BZ}} \frac{d^d k}{(2\pi)^d} \frac{\partial \epsilon_{\vec{k}}}{\partial k_\mu} \frac{\partial \epsilon_{\vec{k}}}{\partial k_\nu} \delta(\epsilon - \epsilon_{\vec{k}}) \quad ,$$

$$\Phi(\epsilon) = \frac{1}{d} \sum_\mu \Phi_{\mu\mu}(\epsilon)$$

- 1-particle spectral function encodes many-body effects:

$$A(\epsilon_k, \omega) = -\frac{1}{\pi} \mathrm{Im} \frac{1}{\omega + \mu - \epsilon_k - \Sigma(\omega)}$$

NB: full frequency-dependent optical conductivity

$$\mathrm{Re}\,\sigma_{\mu\nu}(\vec{q} = \vec{0}, \omega) \; = \; = \frac{2\pi e^2}{\hbar} \int d\omega' \frac{f(\omega') - f(\omega' + \omega)}{\omega} \int d\epsilon \, \Phi_{\mu\nu}(\epsilon) \, A(\epsilon, \omega') A(\epsilon, \omega' + \omega)$$

Transport function for quasi-2D free electrons :

$$\Phi(\epsilon) = \frac{1}{2}\int_{-\pi/c_0}^{+\pi/c_0} \frac{dk_z}{2\pi} \int \frac{dk_x dk_y}{4\pi^2} \left(\frac{\hbar^2}{m}\right)^2 (k_x^2 + k_y^2)\, \delta\left[\epsilon - \frac{\hbar^2}{2m}(k_x^2 + k_y^2)\right],$$

$$\Phi(\epsilon) = \frac{\epsilon}{2\pi c_0}$$

Hence, the IRM limit is naturally expressed in terms of $\Phi(\epsilon_F)/\epsilon_F$

Drude, quasi-2D:

$$\boxed{\sigma_{dc} = \frac{e^2}{\hbar} \frac{\Phi(\epsilon_F)}{\epsilon_F} (k_F l)}$$

UNITS

Energy, Temperature, Frequency:

½ bandwidth D (=1). Think of D= 1eV = 12000 K

Note: $\beta \equiv \dfrac{D}{k_B T}$

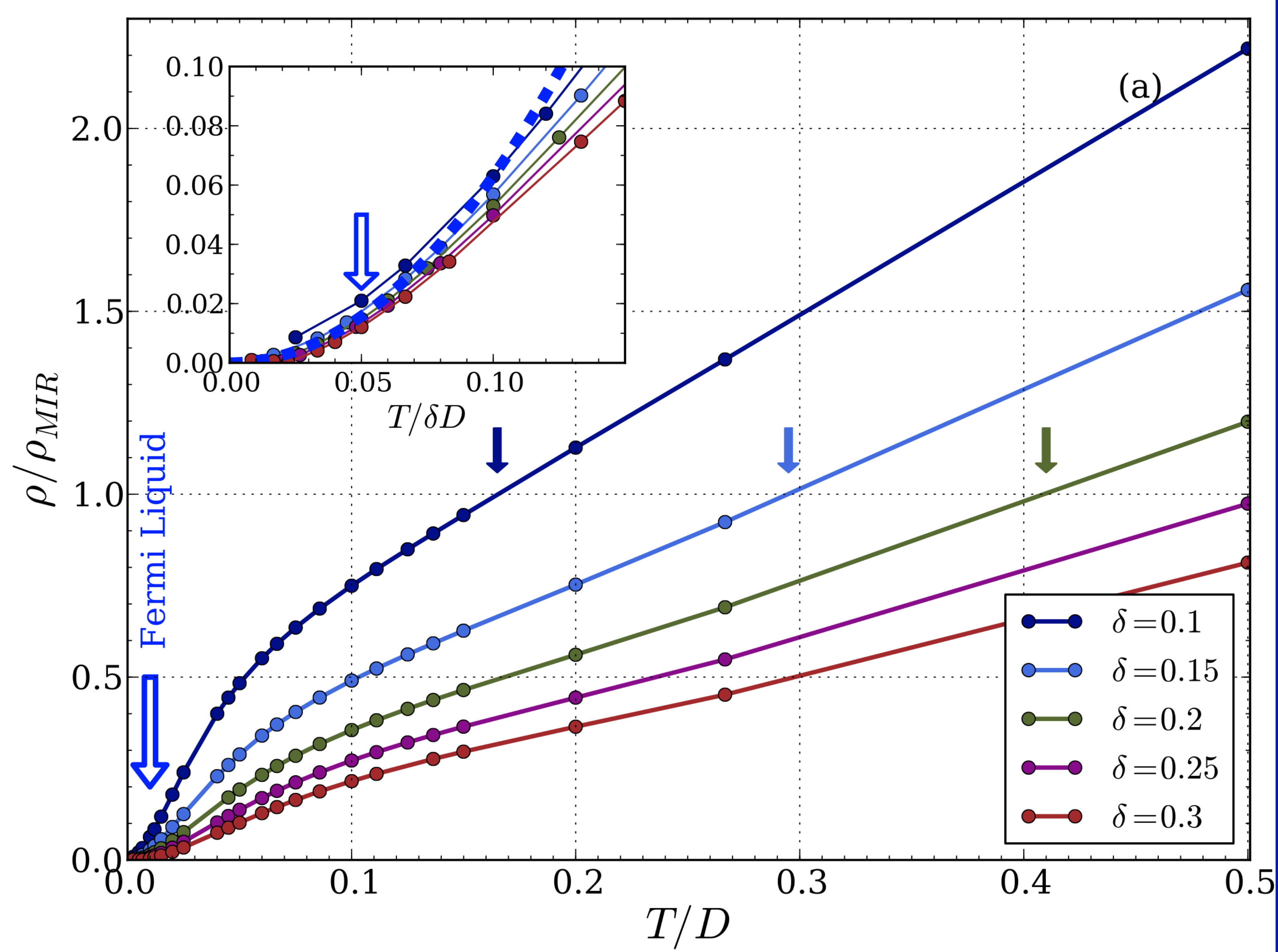
Resistivity

Ioffe-Regel-Mott value $\sigma_M = \dfrac{e^2}{\hbar} \dfrac{\Phi(\epsilon_F)}{\epsilon_F} \quad , \quad (k_F l = 1)$

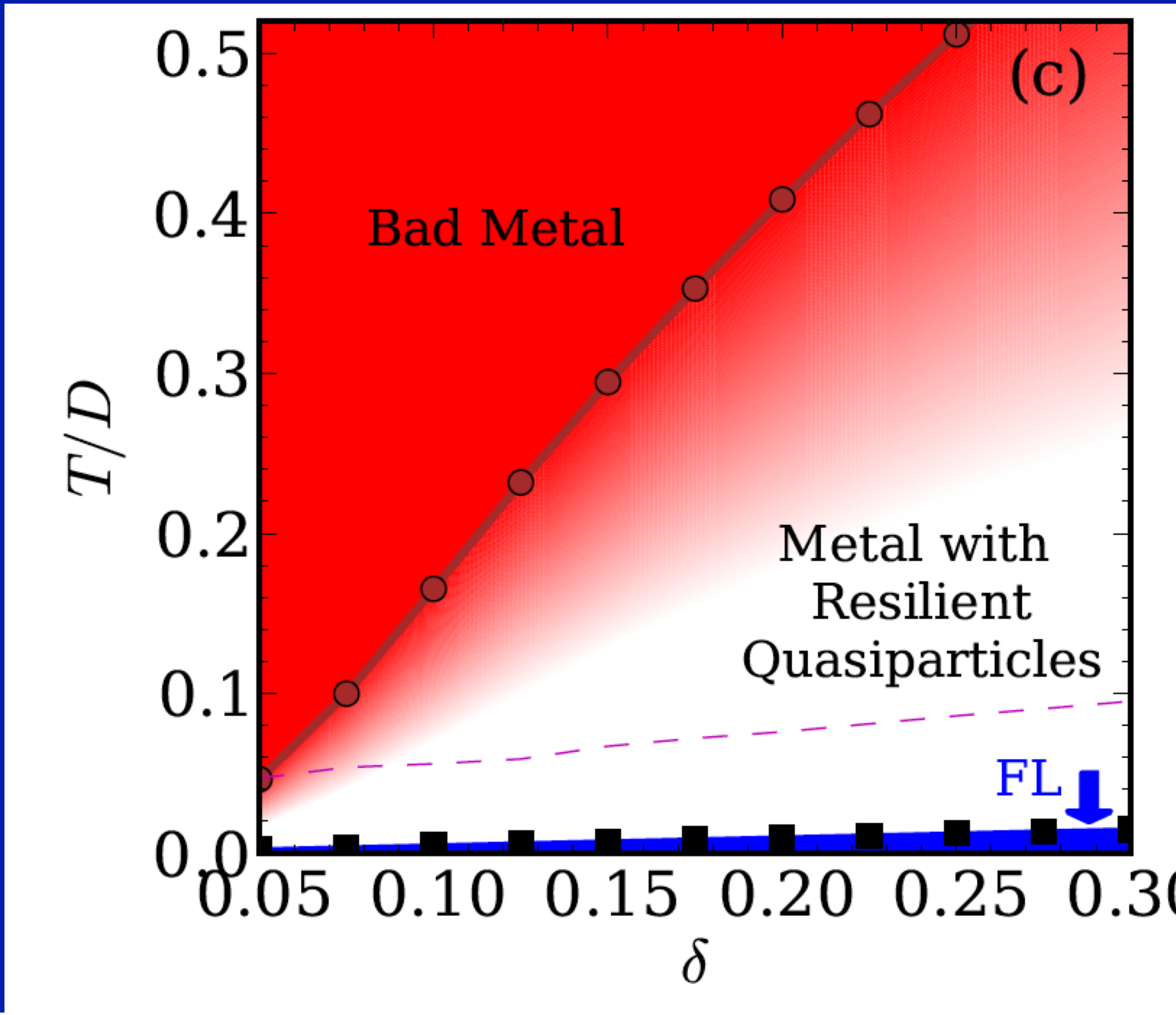
Most calculations shown for U/D=4 (> Mott MIT ~ 3)

OVERVIEW OF T-dependence:
Resistivity and Seebeck

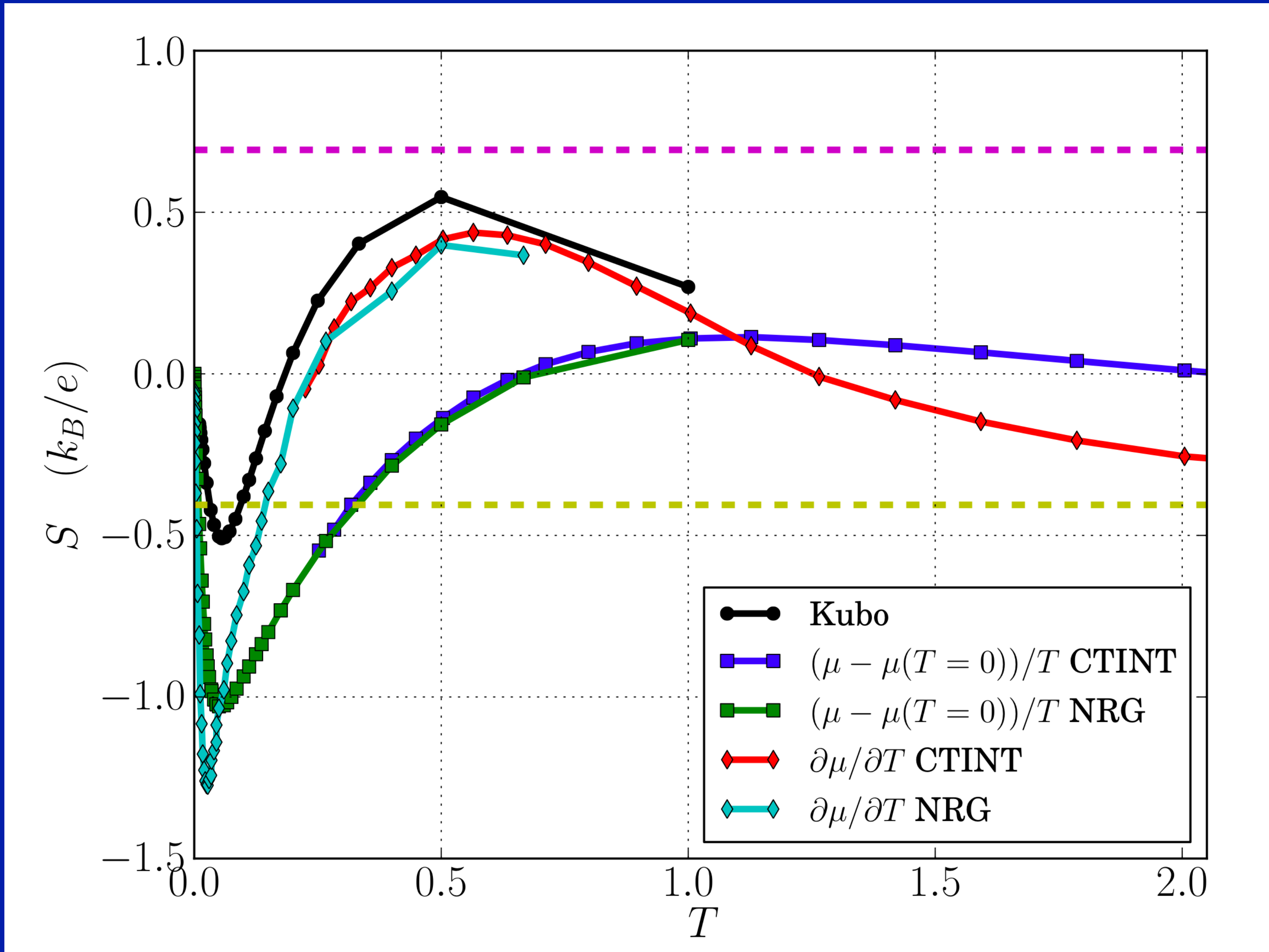
Overview of resistivity vs. T

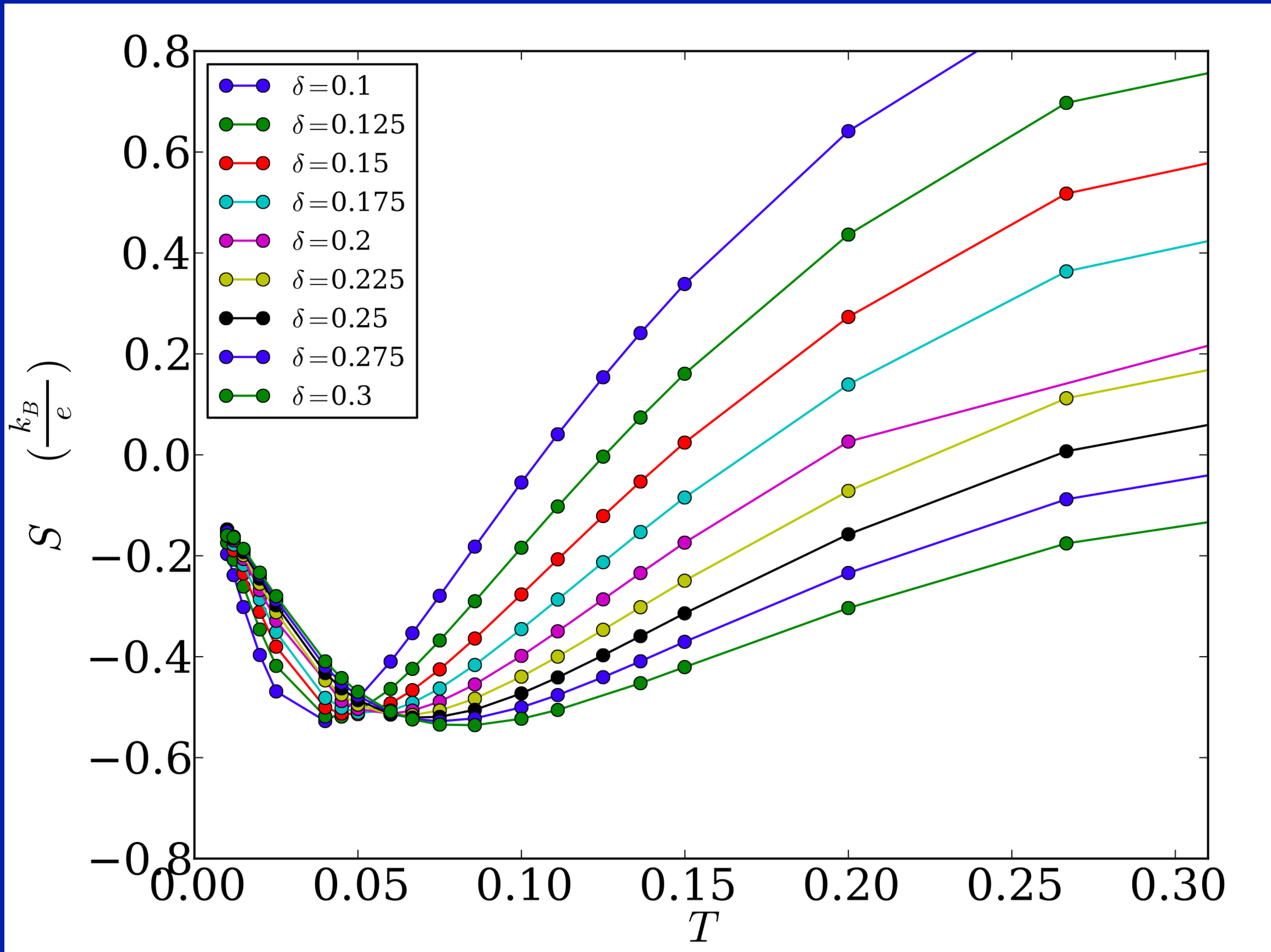


Three transport regimes :



Seebeck (look mostly at black curve)





$\delta=0.1$
$\delta=0.125$
$\delta=0.15$
$\delta=0.175$
$\delta=0.2$
$\delta=0.225$
$\delta=0.25$
$\delta=0.275$
$\delta=0.3$
$S \quad \left(\frac{k_B}{e}\right)$
T
0.8
0.6
0.4
0.2
0.0
−0.2
−0.4
−0.6
−0.8
0.00
0.05
0.10
0.15
0.20
0.25
0.30

A. The Fermi Liquid Regime

Quasiparticles excitations characterized by:

- Enhanced effective mass $m^*/m \sim 1/\delta$ (as compared to band mass)

  → The Brinkman-Rice phenomenon
- Small quasiparticle spectral weight $Z \sim \delta$ (Within local FLT: $m^*/m = 1/Z$)
- Short lifetime
- Low coherence scale above which FL theory no longer applies

All encoded in the low-frequency behaviour of self-energy:

$$\mathrm{Re}\Sigma(\omega + i0^+) = \Sigma_0 + (1 - \frac{1}{Z})\omega + \cdots$$

$$-\,\mathrm{Im}\Sigma(\omega + i0^+) = \frac{c}{D}\left[\omega^2 + (\pi T)^2\right]$$

← Landau

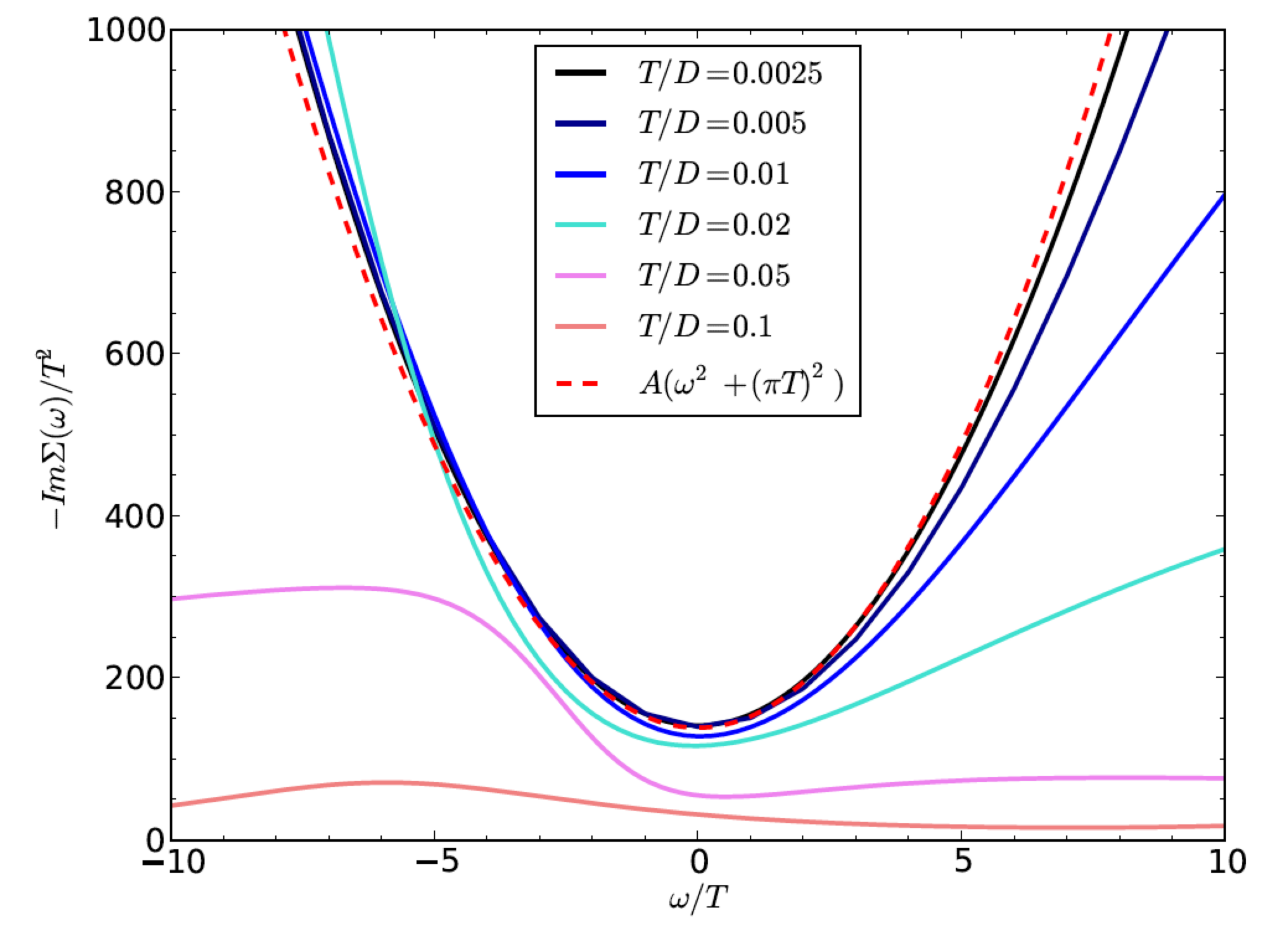
Luttinger theorem (large FS):

$$\mu - \Sigma_0(T = 0) = \mu_{U=0}(n) \equiv \epsilon_F$$

QP spectral weight : Z ~ doping level δ

Short lifetime: close to MIT, c ~ $1/Z^2$: a single energy-scale and 1-parameter scaling form

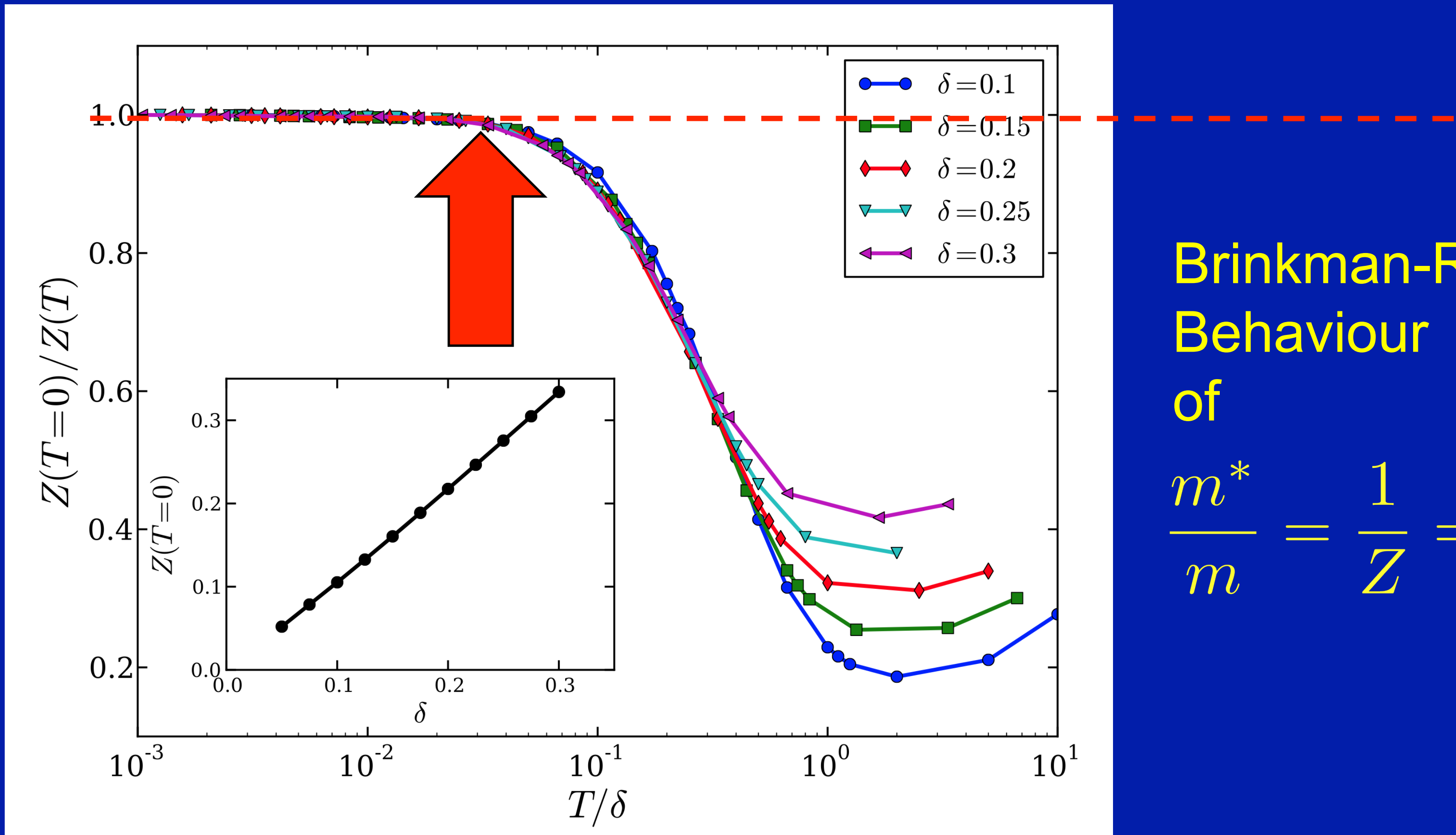
In FL regime: beautiful scaling of the self-energy onto $\omega^2 + (\pi T)^2$



-> Precise identification of FL scale T_{FL}

From T-dep of ``effective mass''

$Z(0)/Z(T)$ = const. for $T < T_{FL}$



Brinkman-Rice Behaviour of

$$\frac{m^*}{m} = \frac{1}{Z} = \frac{1}{\delta}$$

Fermi Liquid scale (U/D=4)

$$\boxed{T_{\mathrm{FL}}/D \simeq 0.05\,\delta}$$

* A very low scale
(as compared to bare electronic scales) !
e.g. D=1eV, δ=10% → 60 K

* Proportional to doping level
but <u>much lower</u> than \`Brinkman-Rice' scale ~ δD
(by 1/20)

Resistivity in the FL regime: analytics

Low ω,T scaling form of scattering rate:

$$-\,\mathrm{Im}\Sigma/D = a\left[\left(\frac{\omega}{\pi\delta}\right)^2 + \left(\frac{T}{\delta}\right)^2\right] + \cdots$$

$$a(U/D=4) \simeq 5.5$$

Note: BOTH
T AND ω
dependence
Is important
(Also in Lorentz ratio)

$$\frac{\rho(T)}{\rho_M} = 1.22a\left(\frac{T}{\delta D}\right)^2 + \cdots \simeq 0.017\left(\frac{T}{T_{FL}}\right)^2$$

$$\rho(T_{FL}) << \rho_M$$

Much lower than MIR value at T_{FL}

Note: Z~δ drops out from A/γ^2 = NON-UNIVERSAL constant
`Kadowaki Woods' 1986, TM Rice 1968
cf. N.Hussey JPSJ 74 (2005) 1107; B.Powell et al. Nature Physics 2009

Seebeck: the dominant linear low-T behaviour involves **corrections to Fermi Liquid theory !**
[Particle-hole asymmetry of the scattering rate]

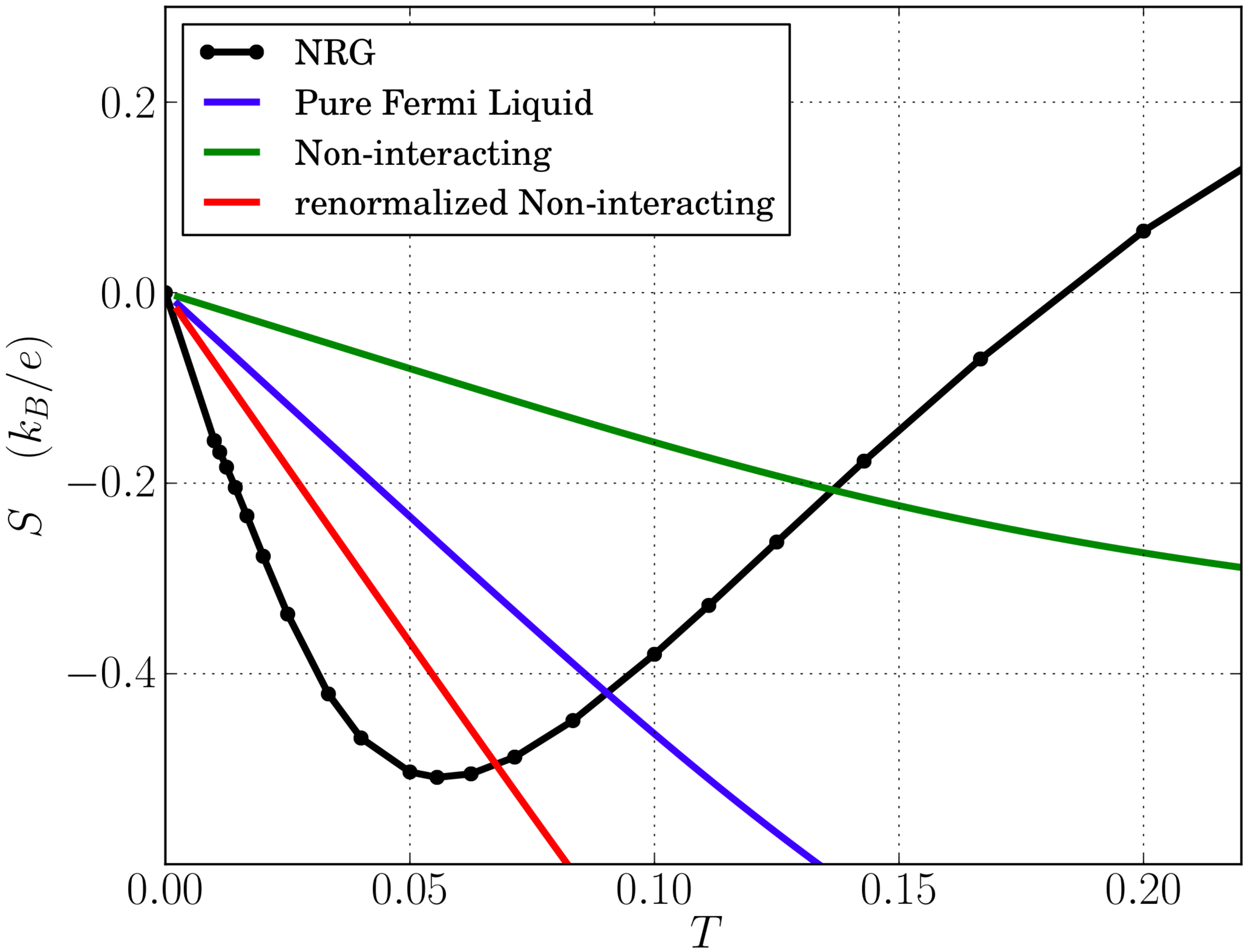
(Haule and Kotliar, arXiv:0907.0192) in "Properties and Applications of Thermoelectric Materials", Edited by V. Zlatic and A.C. Hewson, Springer

$$\Sigma''(\omega) = \Sigma^{(2)}(\omega) + \Sigma^{(3)}(\omega) + \cdots$$

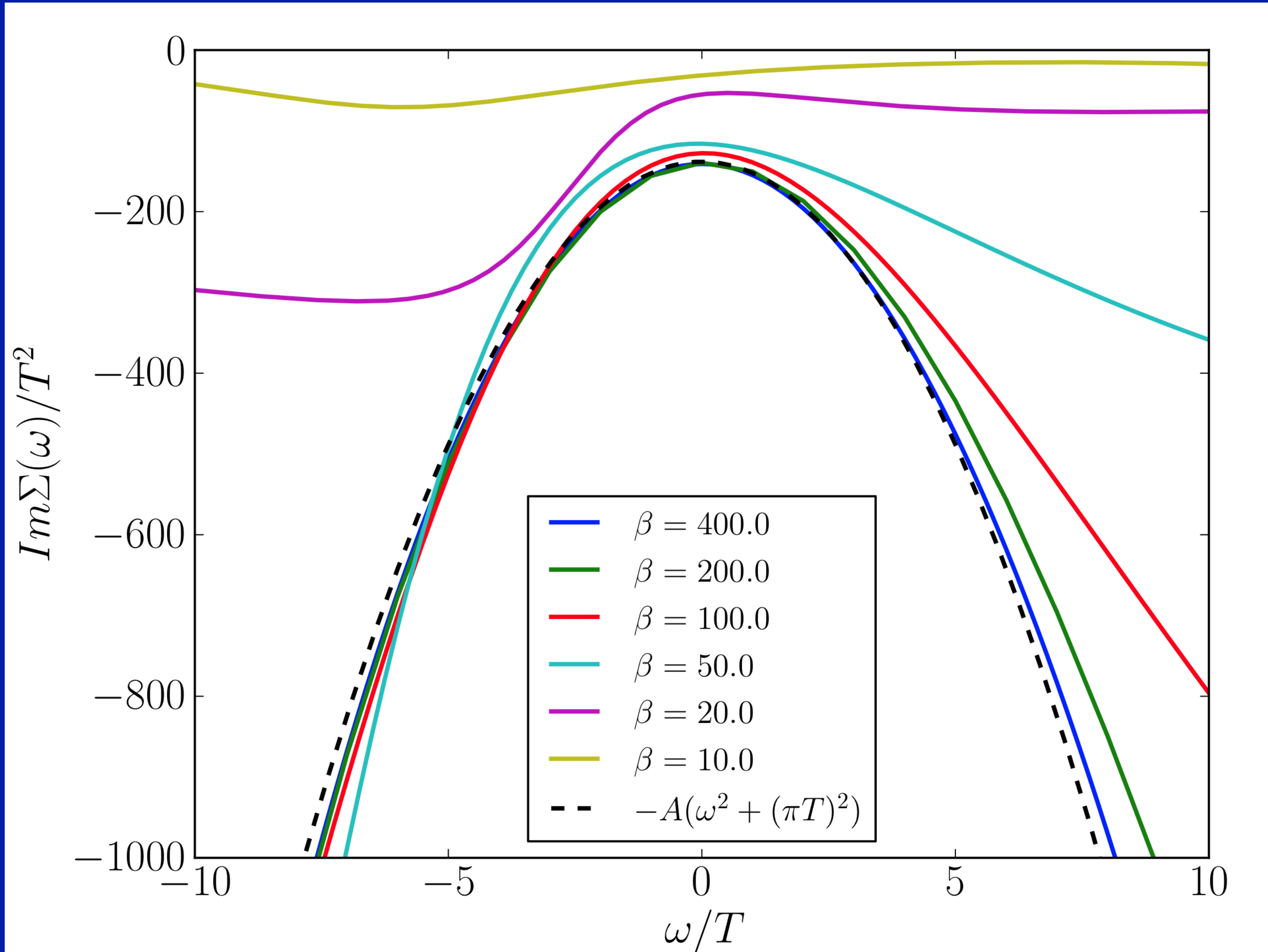
$$\Sigma^3(\omega) = \frac{(a_1\omega^3 + a_2\,\omega T^2)}{Z^3}$$

$$E_n^k = \int_{-\infty}^{\infty} \frac{x^n dx}{4\cosh^2(x/2)[1+(x/\pi)^2]^k}$$

$$S = \boxed{-\frac{k_B}{|e|}\frac{k_B T}{Z}}\left[\boxed{\frac{\Phi'(\mu_0)}{\Phi(\mu_0)}\frac{E_2^1}{E_0^1}} - \boxed{\frac{a_1 E_4^2 + a_2 E_2^2}{\gamma_0 E_0^1}}\right]$$


NRG
Pure Fermi Liquid
Non-interacting
renormalized Non-interacting
0.2
0.0
−0.2
−0.4
0.00
0.05
0.10
0.15
0.20
T
$S \quad (k_B/e)$

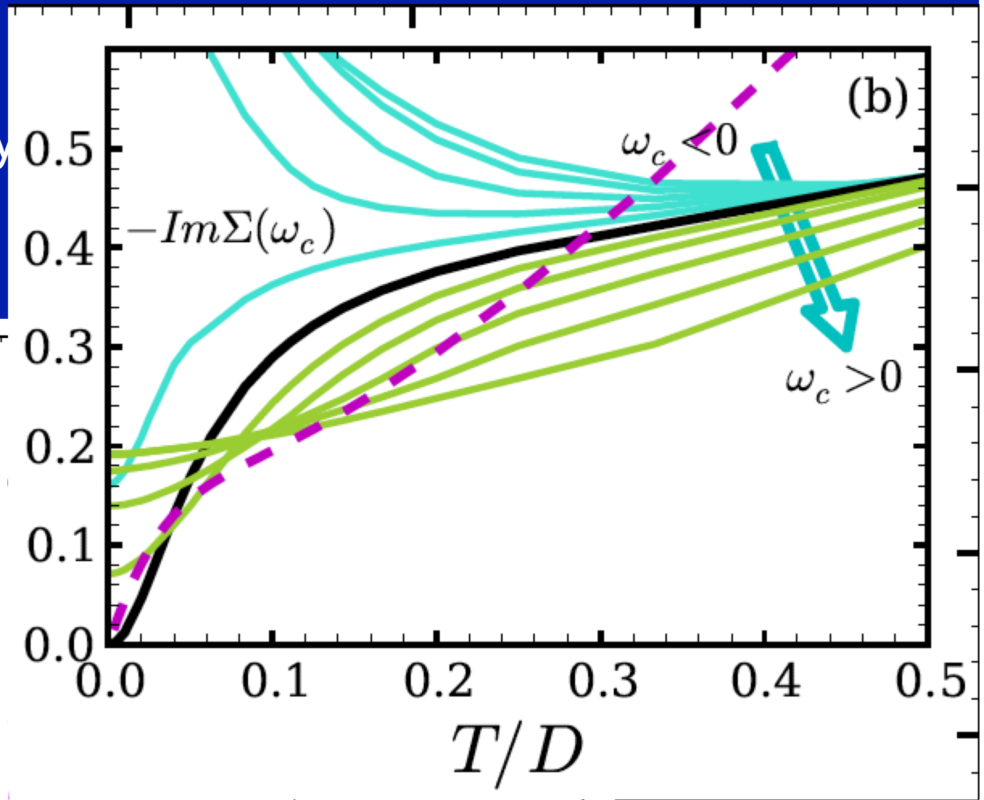
Particle-hole asymmetry of the scattering rate

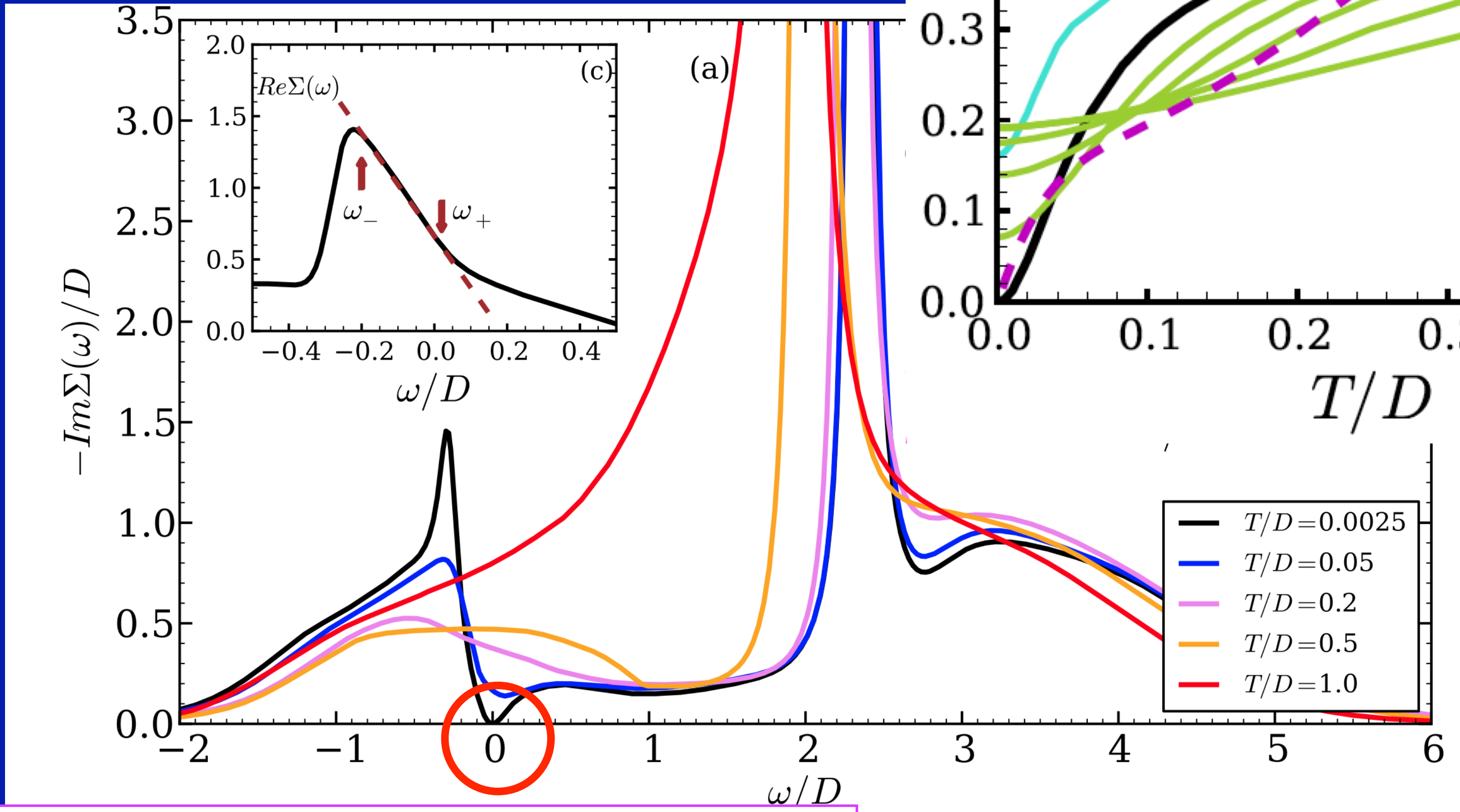


Self-energy on a broader scale: Beyond Landau FLT

Fixed ω, vs. T:

Scattering rate: particle-hole asyymetry
Longer lifetime for
electron-like excitations



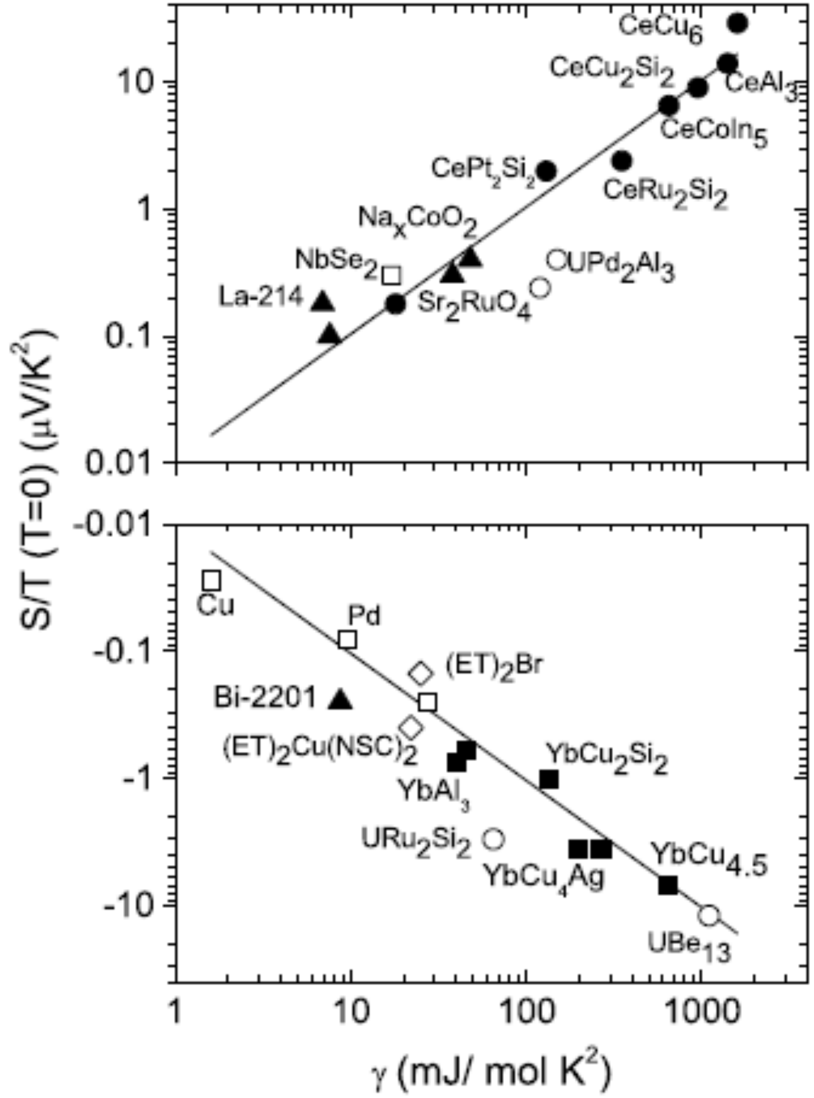


This is where Landau theory applies !

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER
J. Phys.: Condens. Matter **16** (2004) 5187–5198 PII: S0953-8984(04)80129-2

The Behnia-Jaccard-Flouquet law:
(wn=hen only Z matters)

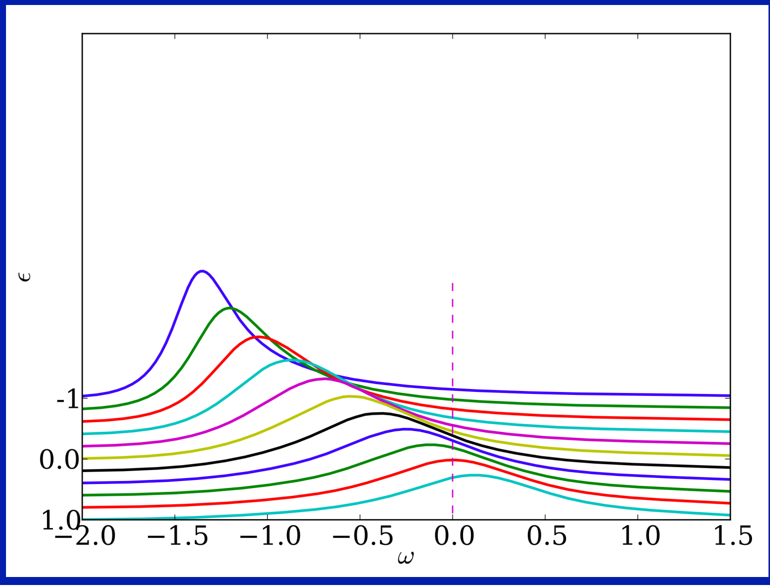
$$\frac{S/T}{\gamma}\ ,\quad (C=\gamma T+\cdots)$$

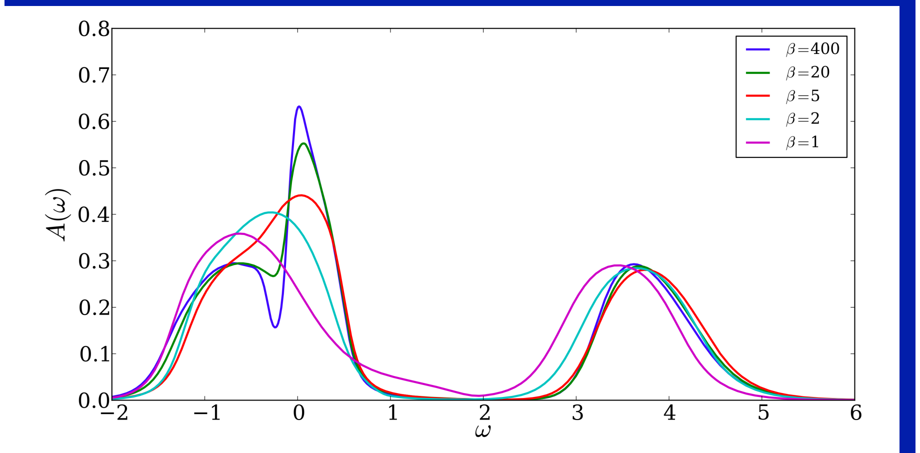


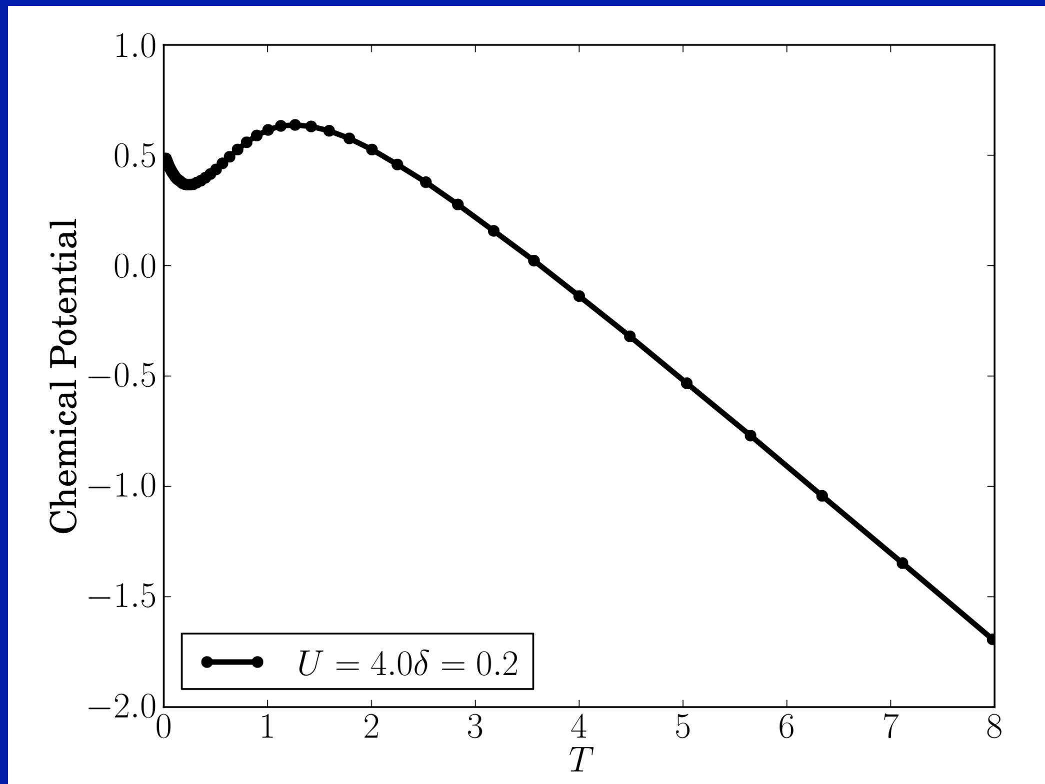
$$\frac{S}{\gamma T}=-\frac{3}{|e|}\frac{1}{D(\mu_0)}\left[\frac{\Phi'(\mu_0)}{\Phi(\mu_0)}\frac{E_2^1}{E_0^1}-\frac{a_1E_4^2+a_2E_2^2}{\gamma_0E_0^1}\right]$$

B. High-temperature regime(s):
Heikes limit(s)
[and `Kelvin formula

High temperatures: $T>T_{IRM}$ and beyond…
Incoherent regime – Hubbard band physics
~ classical carriers in a rigid band







Chemical potential is linear in T at very hi-T

Hi-T expansion of thermopower:

$$
\begin{aligned}
A_0 &= \pi N \zeta \left(\gamma_0 + \gamma_1 \beta \tau + \frac{1}{4} \gamma_2 \beta^2 [3\tau - 1] \right) \\
A_1 &= \pi N \zeta \left(-\alpha \gamma_0 + \gamma_1 \beta \left[1 - \alpha \tau \right] + \gamma_2 \beta^2 \left[\tau - \frac{\alpha}{4}(3\tau - 1) \right] \right) \\
A_2 &= \pi N \zeta \left(\alpha^2 \gamma_0 + \gamma_1 \beta \left[\alpha^2 \tau - 2\alpha \right] + \gamma_2 \beta^2 \left[1 - 2\alpha\tau + \frac{\alpha^2}{4}(3\tau - 1) \right] \right)
\end{aligned}
$$

Hence, all details of fermiology/bandstructure cancel out and a very simple hi-T limit holds:

PHYSICAL REVIEW B VOLUME 13, NUMBER 2 15 JANUARY 1976

Thermopower in the correlated hopping regime

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G. Beni
Bell Laboratories, Murray Hill, New Jersey 07974

(Received 16 June 1975)

$$S_\infty = + \frac{k_B}{e} \frac{\mu}{k_B T}$$

Thermodynamics: $T ds = dE - \mu dn \Rightarrow \frac{\mu}{T} = - \frac{\partial s}{\partial n} |_E$

$$S_\infty = - \frac{k_B}{e} \frac{\partial (s/k_B)}{\partial n} |_E$$

The two hi-T (Heikes) limits

1. D < T << U

$$p_0 + 2p_1 = 1\,,\; n = 2p_1 \Rightarrow p_0 = 1 - n\,,\; p_1 = n/2$$

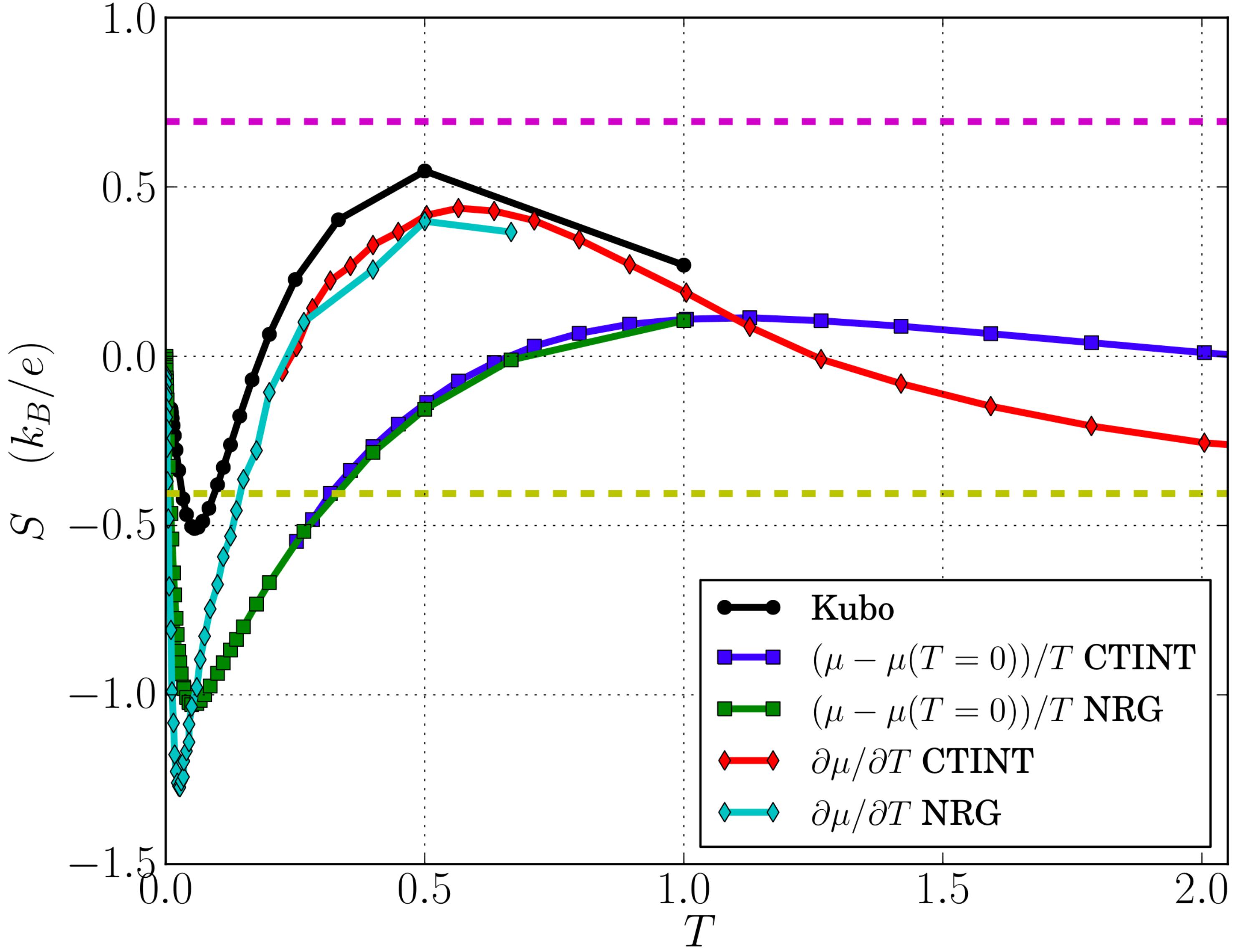
$$s/k = -(1-n)\ln(1-n) - n\ln n/2$$

$$\Rightarrow S_\infty^{(1)} = -\frac{k_B}{e}\ln\frac{2(1-n)}{n}$$

2. T > U

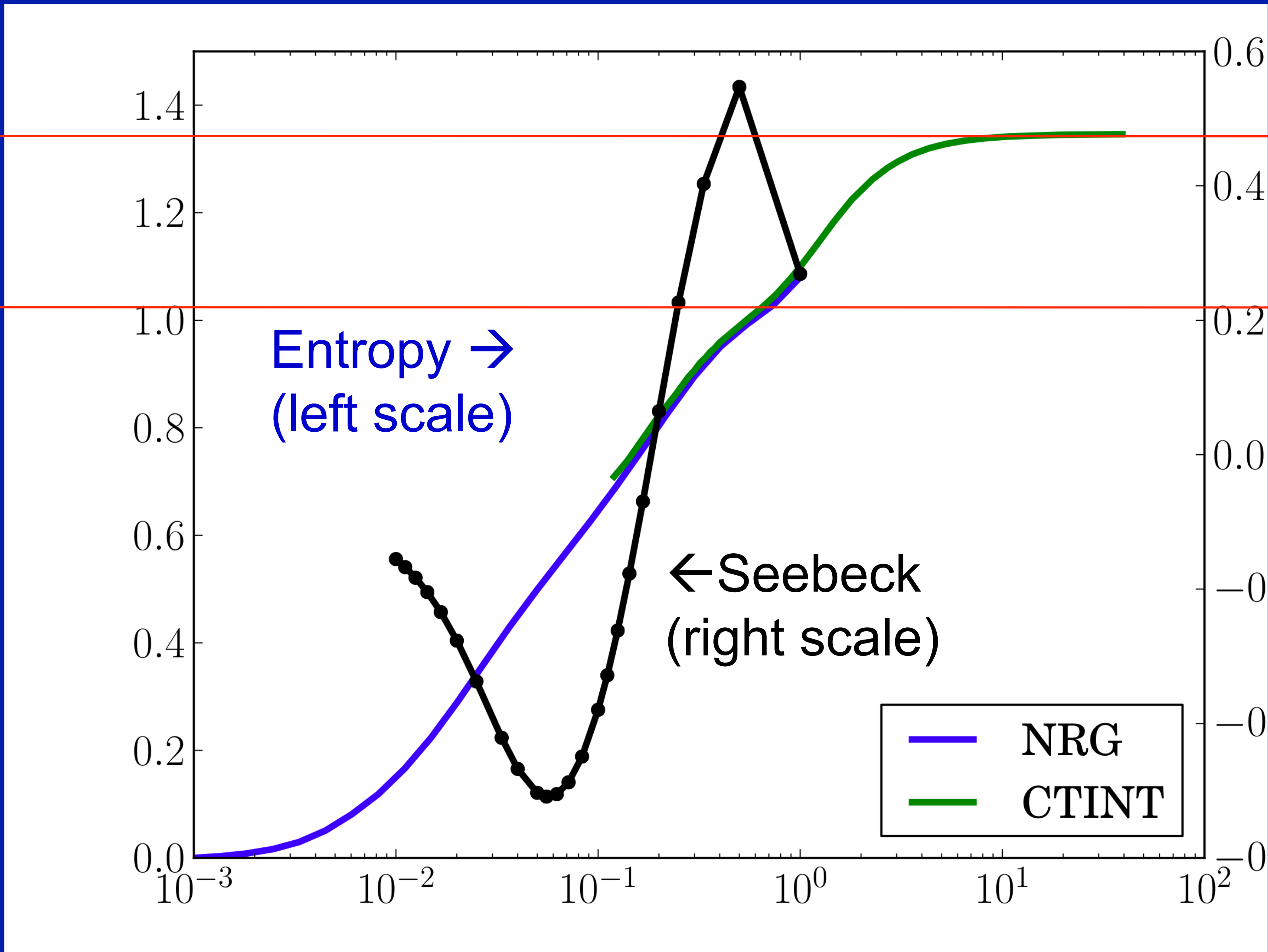
$$s/k = -2\left[\frac{n}{2}\ln\frac{n}{2} + \frac{1-n}{2}\ln\frac{1-n}{2}\right]$$

$$\Rightarrow S_\infty^{(2)} = +\frac{k_B}{e}\ln\frac{n}{2-n}$$

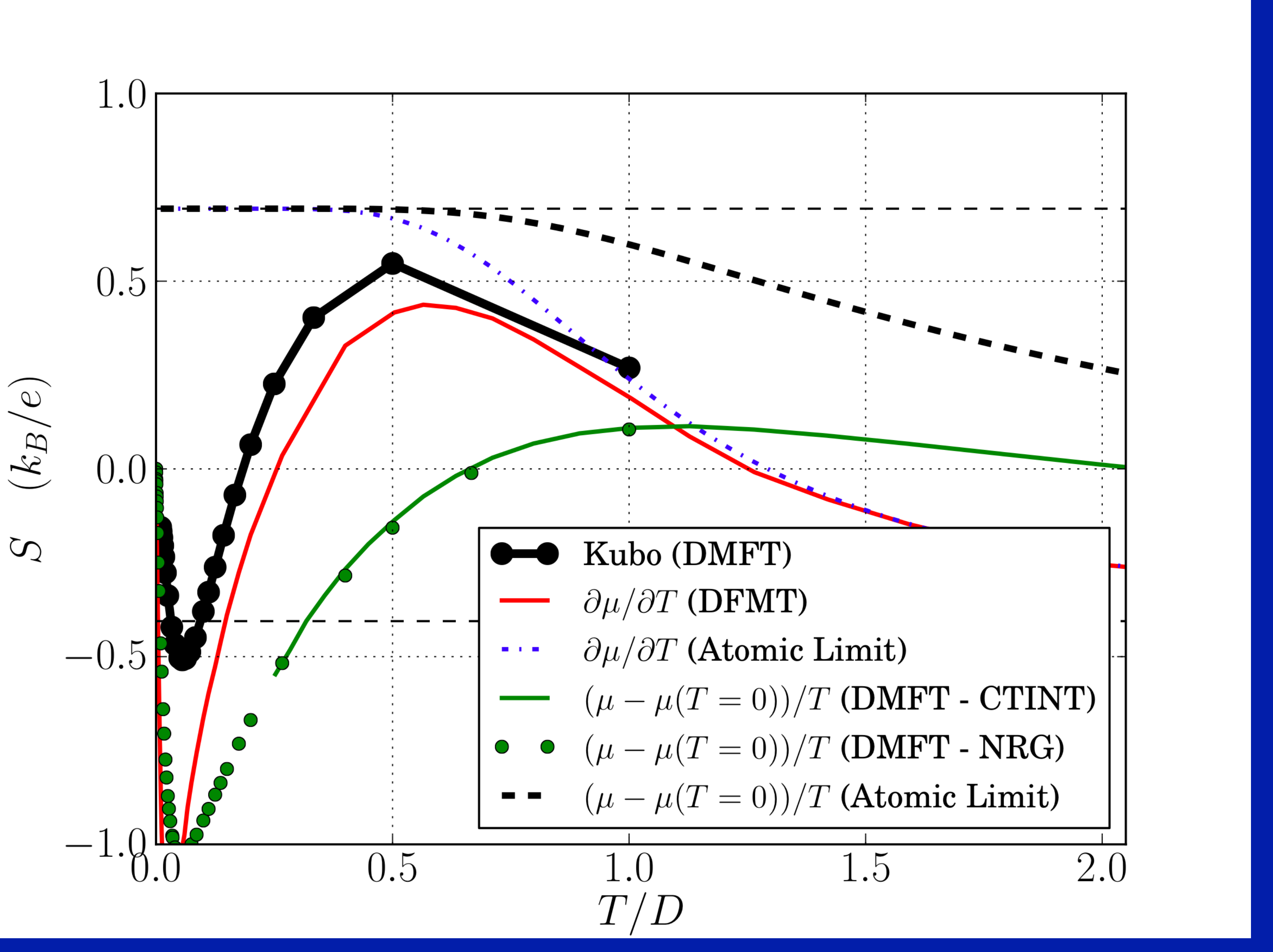


$S \quad (k_B/e)$
T
1.0
0.5
0.0
−0.5
−1.0
−1.5
0.0
0.5
1.0
1.5
2.0
Kubo
$(\mu - \mu(T=0))/T$ CTINT
$(\mu - \mu(T=0))/T$ NRG
$\partial\mu/\partial T$ CTINT
$\partial\mu/\partial T$ NRG

Seebeck and Entropy



“Kelvin formula” dμ/dT (cf. Shastry) is a good approximation (much better than Heike μ/T)

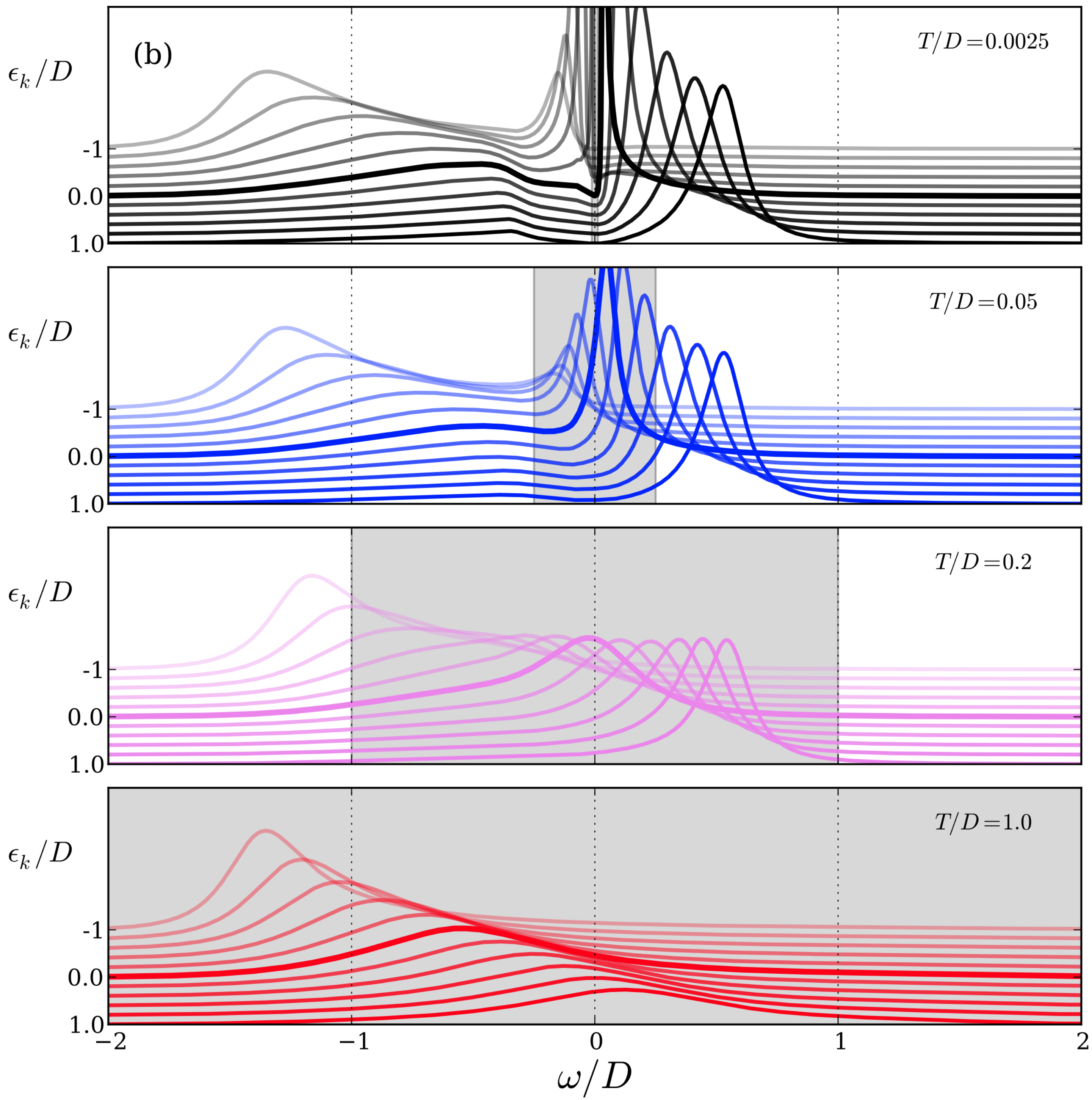


C. Intermediate regime

$T_{FL} < T < T_{MIR}$

Metal with

"Resilient quasiparticles"



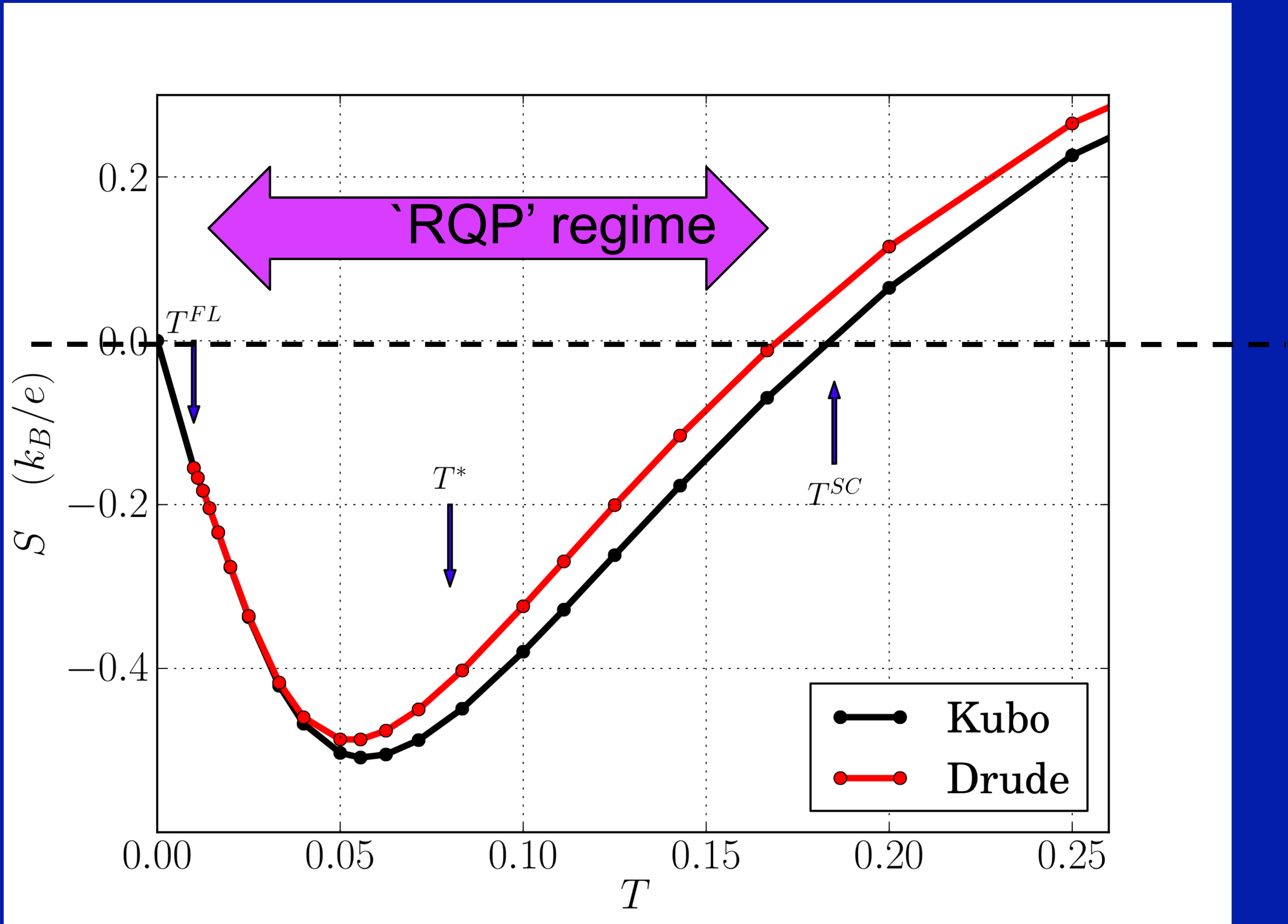
Landau FL

Resilient Non-Landau QPs

A bit below Ioffe Regel Mott

Fully Incoherent (above MIR)

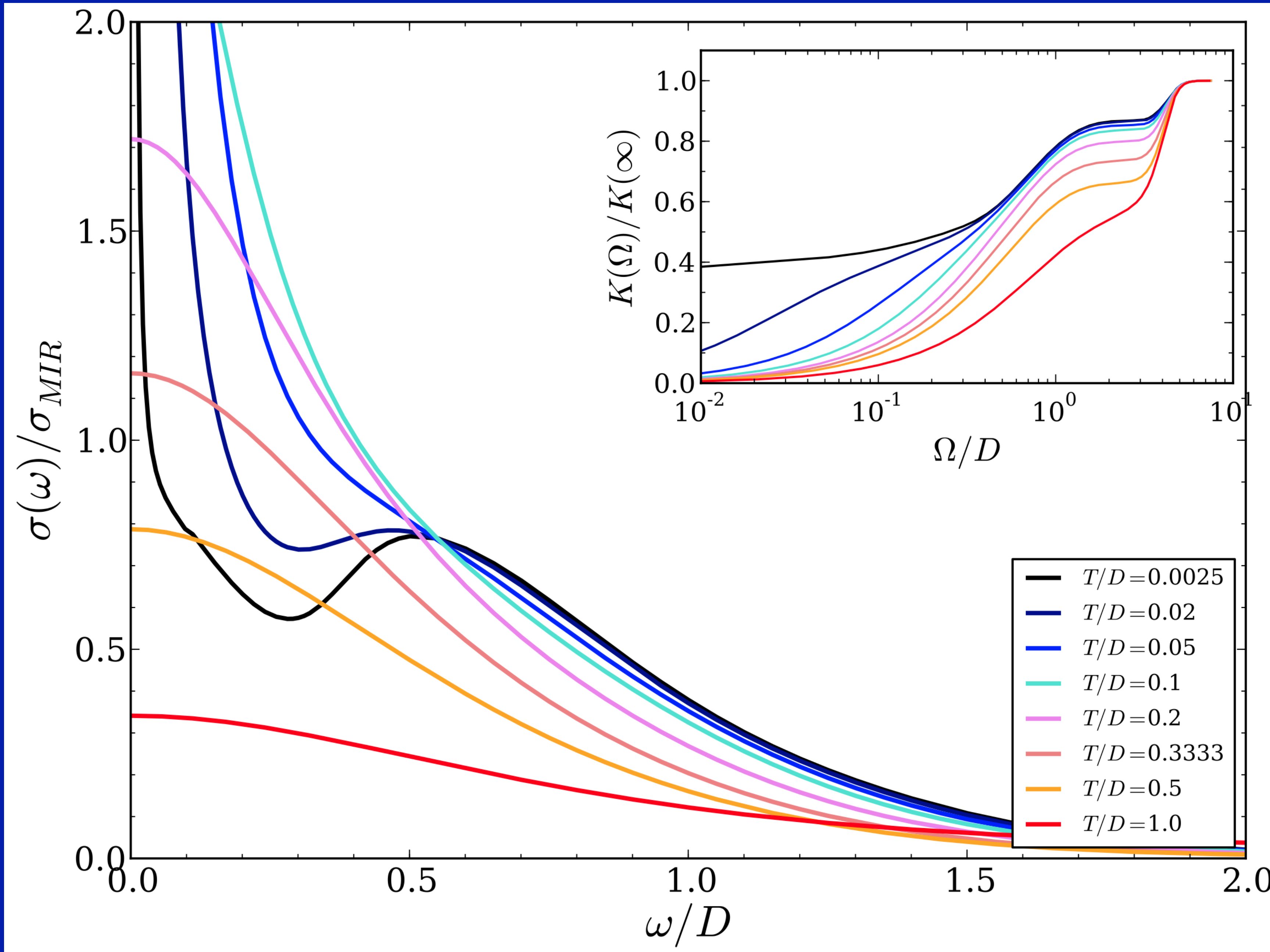
Seebeck in intermediate regime: minimum dominated by electron-like `resilient' quasiparticles



Signature of the the two crossovers (FL, MIR) in optical spectroscopy:

1. Merging of Drude peak and mid-infrared into broad peak at T_{FL}
2. Disappearance of QPs at MIR
3. Redistribution of sp.weight over very high energies at MIR, but involving only mid-infrared below T_{MIR}

cf. Hussey, Takenaka et al. LSCO PRB 2003
Hussey, Phil Mag
Gunnarsson RMP



$\sigma(\omega)/\sigma_{MIR}$
ω/D
$K(\Omega)/K(\infty)$
Ω/D
$T/D = 0.0025$
$T/D = 0.02$
$T/D = 0.05$
$T/D = 0.1$
$T/D = 0.2$
$T/D = 0.3333$
$T/D = 0.5$
$T/D = 1.0$

Back to real materials:
Towards predictive power for Seebeck from:

- Hi-T limits (Heikes/Kelvin)

→ Several authors

(e.g. Koshibae et al. PRB 2000)

- Electronic structure + many-body ``LDA+DMFT'' calculations

Main messages about thermopower

- Seebeck is a sensitive probe of the different transport regimes
- Particle-hole asymmetry crucial: not only of bandstructure <u>also of scattering rate</u>
- Fermi liquid theory insufficient even for dominant linear behaviour at low T !
- Progress in first-principle calculations of thermopower !

Take-home messages (cont'd):

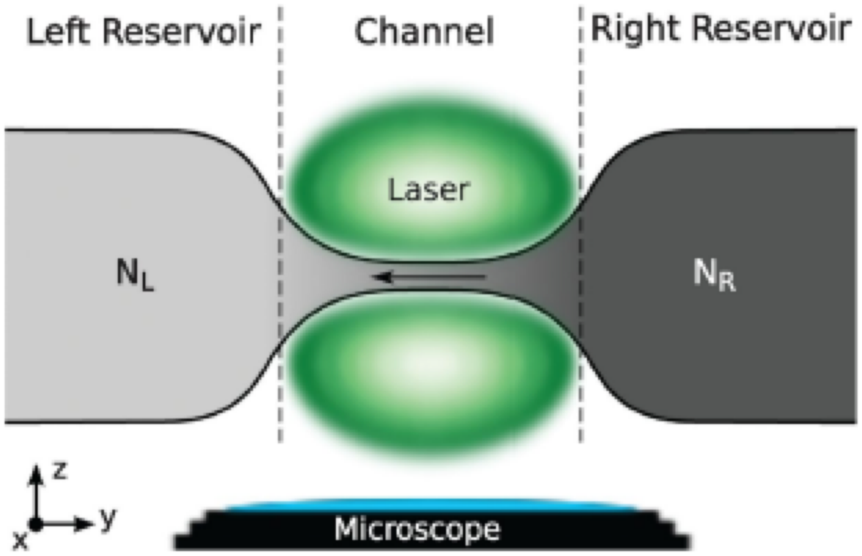
- Well-defined `resilient' QPs exist well above the range of validity of FL theory, all the way up to T_{MIR}
- They carry the current: no longer Landau's T^2, but smaller than MIR resistivity
- Clear spectroscopic signatures of their disappearance, eg in optics
- For a hole-doped MI: electron-like excitations above FS much longer-lived

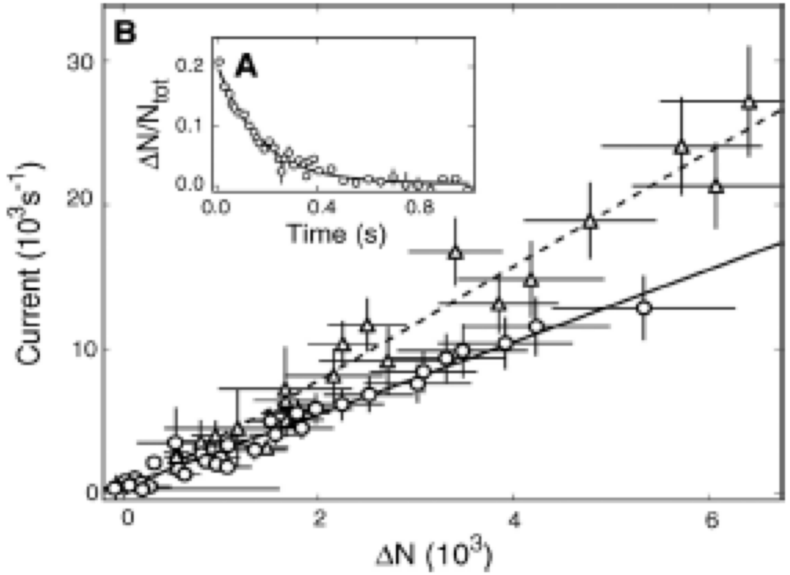
→ Explore the `dark side' of the FS !

In brief, as a dessert: Probing Thermoelectric Transport with Ultra-Cold Atoms

C.Grenier, C.Kollath & AG arXiv:1209.3942
Slides: courtesy C.Grenier
Acknowledgements: ETH cold atom group

Introduction - Experimental motivation





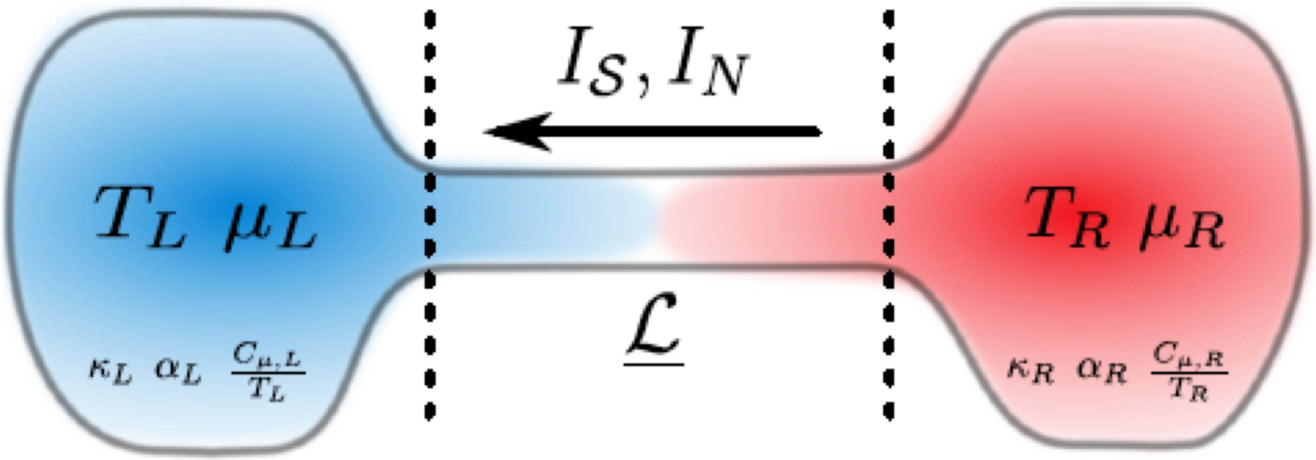
ETH, 2012 (Brantut et al., Science)

$\rightarrow$ Realization of a two terminal transport setup
Discharge of a mesoscopic capacitor (reservoirs) in a resistor (the conduction channel)

$\Rightarrow$ Simulation of mesoscopic physics with cold atoms

Question : Can this setup demonstrate offdiagonal transport ?

Transport setup



- Two terminal configuration
- Grand canonical ensemble : flow of entropy and particles between reservoirs
- Linear response coefficients in $\underline{\mathcal{L}}$

Transport equations and coefficients

Equations for particle number and temperature difference :

$$\boxed{\tau_0 \frac{d}{dt}\begin{pmatrix} \Delta N/\kappa \\ \Delta T \end{pmatrix} = -\underline{\Lambda}\begin{pmatrix} \Delta N/\kappa \\ \Delta T \end{pmatrix}, \underline{\Lambda} = \begin{pmatrix} 1 & -S \\ -\frac{S}{\ell} & \frac{L+S^2}{\ell} \end{pmatrix}.}$$

$\Rightarrow$ Discharge of a capacitor through a resistor, including thermal properties

Global timescale $\tau_0 = \frac{\mathcal{L}_{11}}{\kappa} \sim RC$

Effective transport coefficients :

$L \equiv \mathcal{L}_{22}/\mathcal{L}_{11} - (\mathcal{L}_{12}/\mathcal{L}_{11})^2 \sim R/TR_T \rightarrow$ Lorenz number

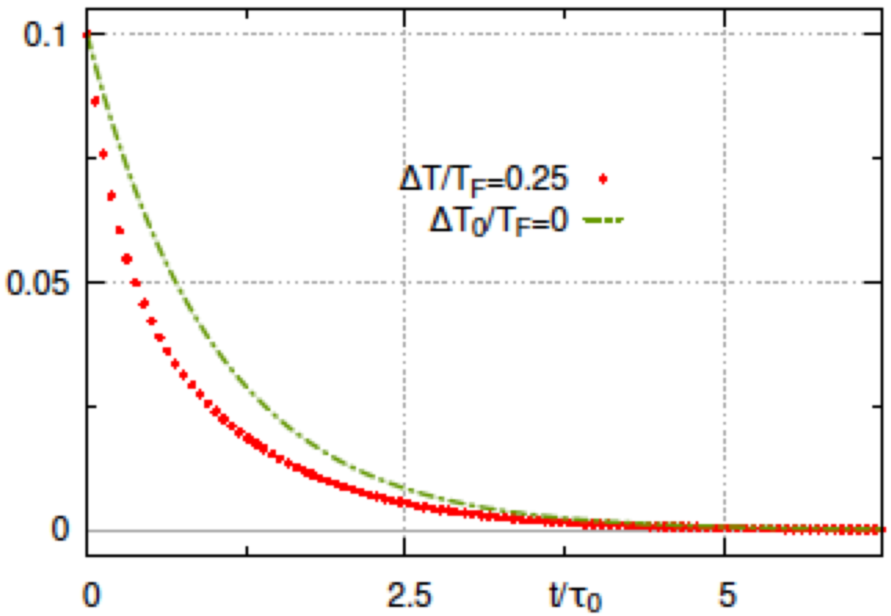
$\ell \equiv C_\mu/\kappa T - (\alpha/\kappa)^2 = C_N/\kappa T \rightarrow$ Charac. of reservoirs, analogue to L

$S \equiv \alpha/\kappa - \mathcal{L}_{12}/\mathcal{L}_{11} \rightarrow$ Total Seebeck coefficient

Solution to transport equations

$$\Delta N(t) = \underbrace{\left\{\frac{1}{2}\left[e^{-t/\tau_-} + e^{-t/\tau_+}\right] + \left[1 - \frac{L+S^2}{\ell}\right]\frac{e^{-t/\tau_-} - e^{-t/\tau_+}}{2(\lambda_+ - \lambda_-)}\right\}\Delta N_0}_{\text{Diagonal transport: exponential decrease}} +$$

$$\underbrace{\boxed{\frac{S\kappa}{\lambda_+ - \lambda_-}\left[e^{-t/\tau_-} - e^{-t/\tau_+}\right]\Delta T_0}}_{\text{Thermoelectric effect !}}, \tau_\pm^{-1} = \tau_0^{-1}\lambda_\pm$$



Particle imbalance with and without initial temperature difference

With $\frac{\Delta N_0}{N_0} = 10\%$

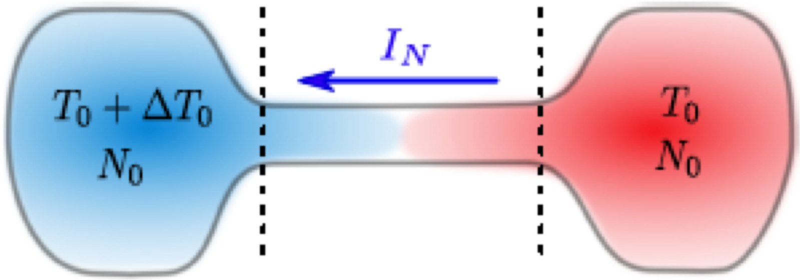
$\Rightarrow$ Hard to isolate thermoelectric contribution

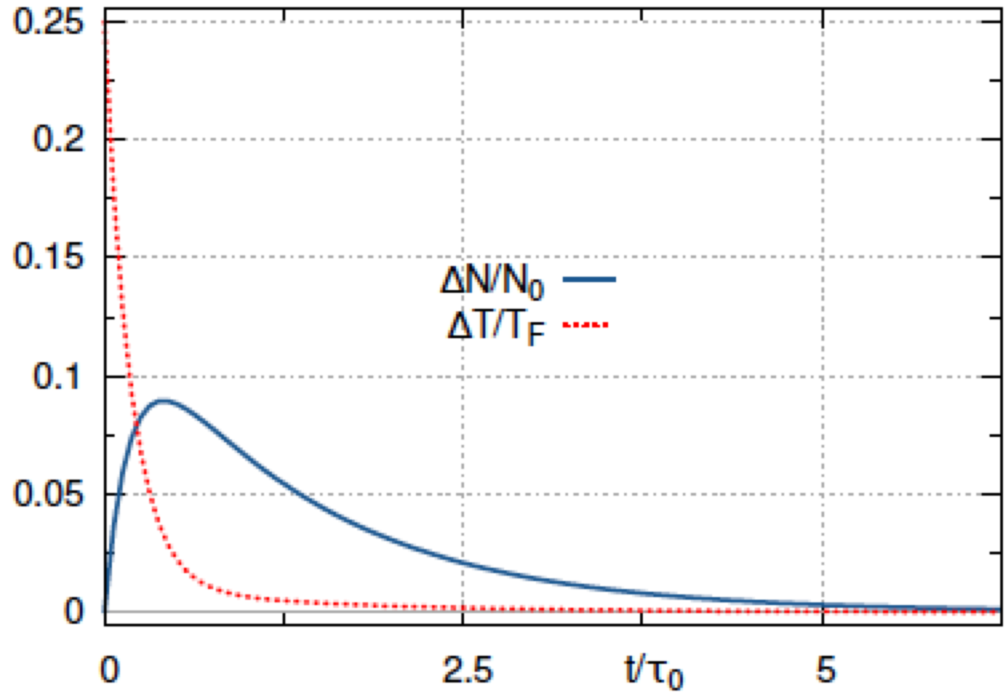
when exp. decrease is switched on

An appropriate setup

Two steps

i. Prepare reservoirs with equal particle number and different temperatures, with closed constriction
ii. Open the constriction and monitor particle number





Particles flow in both ways during temperature equilibration :
$\rightarrow$ Transient analogue of the Seebeck effect

What happens ?

$$\Delta N(t) = \frac{S\kappa}{\lambda_+ - \lambda_-}\left[e^{-t/\tau_-} - e^{-t/\tau_+}\right]\Delta T_0\,,\tau_\pm^{-1} = \tau_0^{-1}\lambda_\pm$$

with $\lambda_\pm = \frac{1}{2}\left(1 + \frac{L+S^2}{\ell}\right) \pm \sqrt{\frac{S^2}{\ell} + \left(\frac{1}{2} - \frac{L+S^2}{2\ell}\right)^2}$ and $S = \alpha/\kappa - \mathcal{L}_{12}/\mathcal{L}_{11}$.

i. Particle imbalance and current proportional to ΔT_0 and S

ii. Reservoir properties participate to the effect

iii. Sign of ΔN (and I_N) given by the sign of S

$$I_{\mathcal{S}} = SI_N - \underbrace{\sigma_{th}\Delta T}_{\text{Dominant}}$$

Entropy flows from hot to cold : 2nd principle ✓