``Enseigner la recherche en train de se faire''

COLLÈGE DE FRANCE 1530

Chaire de
Physique de la Matière Condensée

Seconde partie:
Quelques questions liées au transport dans les matériaux à fortes corrélations électroniques

Les mercredis dans l'amphithéâtre Maurice Halbwachs
11, place Marcelin Berthelot 75005 Paris
Cours à 14h30 - Séminaire à 15h45

Cycle 2011-2012
Partie II: 30/05, 06/06,13/06/2012

Antoine Georges

Séance du 13 juin 2012

- **Séminaires : 15h45 et 16h45 –**

Sriram Shastry (University of Caifornia, Santa Cruz)

- ***1. "Simple insights into the Thermopower of correlated matter"***

  2. "Extremely correlated Fermi liquids"

OUTLINE of the 3 lectures

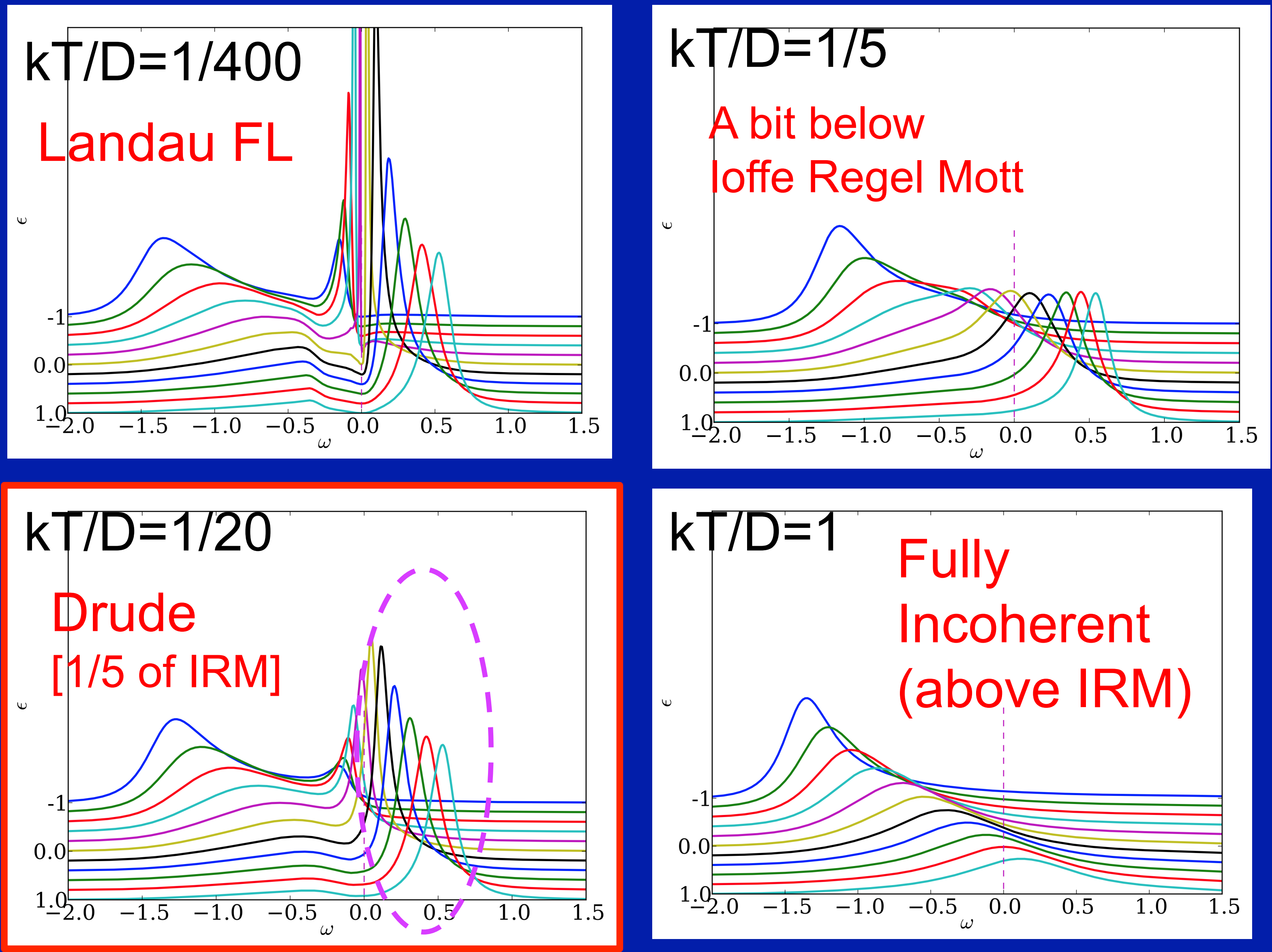
- May, 30: Phenomenology, simple theory background. Mainly raise questions.
- June, 6: Answer some of these questions for a doped Mott insulator (simplest 1-site DMFT description, recent results)
- June, 13 : A few remarks on thermolectric power (Seebeck coefficient)

→ Not really a lecture on thermoelectrics ! [Here Seebeck as probe]
→ `Hors d'oeuvre' / `Mise en bouche' for next year's lectures (march-april 2013) on thermoelectrics

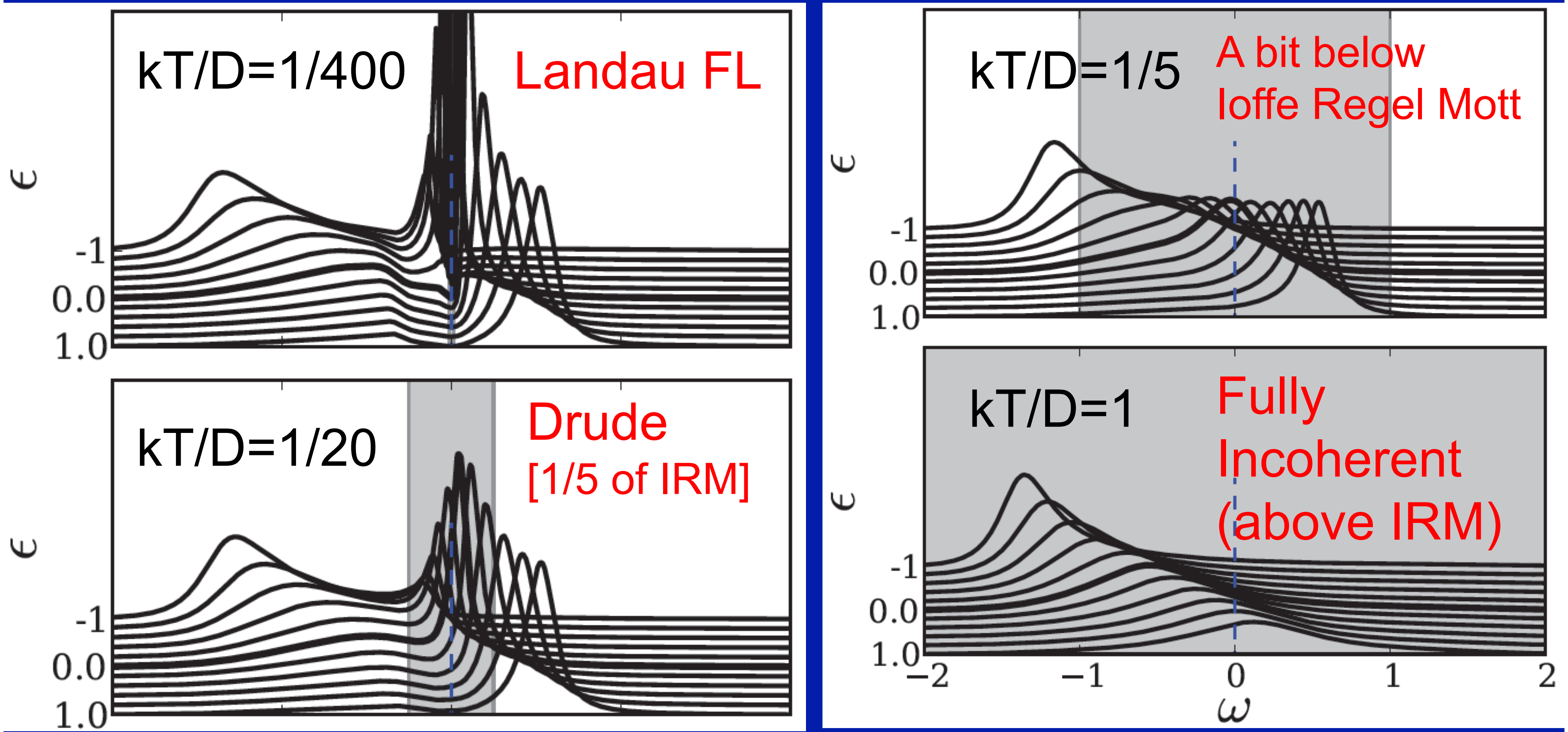
Three transport regimes (previous lectures)

- $T<T_{FL}$: Fermi Liquid regime with long-lived coherent quasiparticles ($T_{FL} \sim 0.05\ \delta.D$)
- $T_{FL}< T < T_{IRM}$ Metallic resistivity. In this regime, quasiparticles are still present but with a shorter lifetime than Landau's. Optics has a low-frequency peak. Drude description of transport applies
- $T>T_{IRM}$ `Pseudo-metallic' resistivity in excess of IRM value. No quasiparticles. Doped lower Hubbard band. Optics ~ flat.

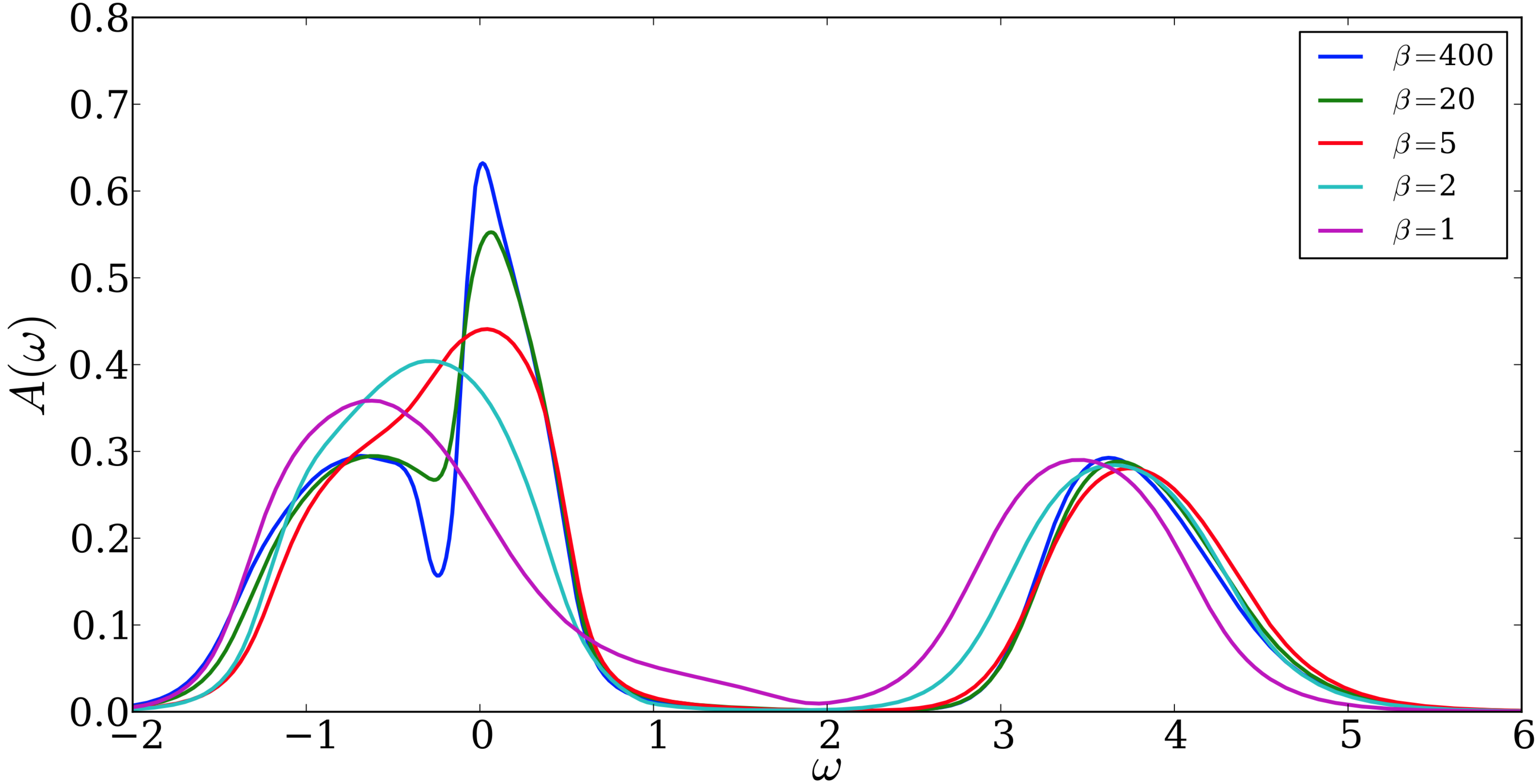
Landau quasiparticles → Drude quasiparticles



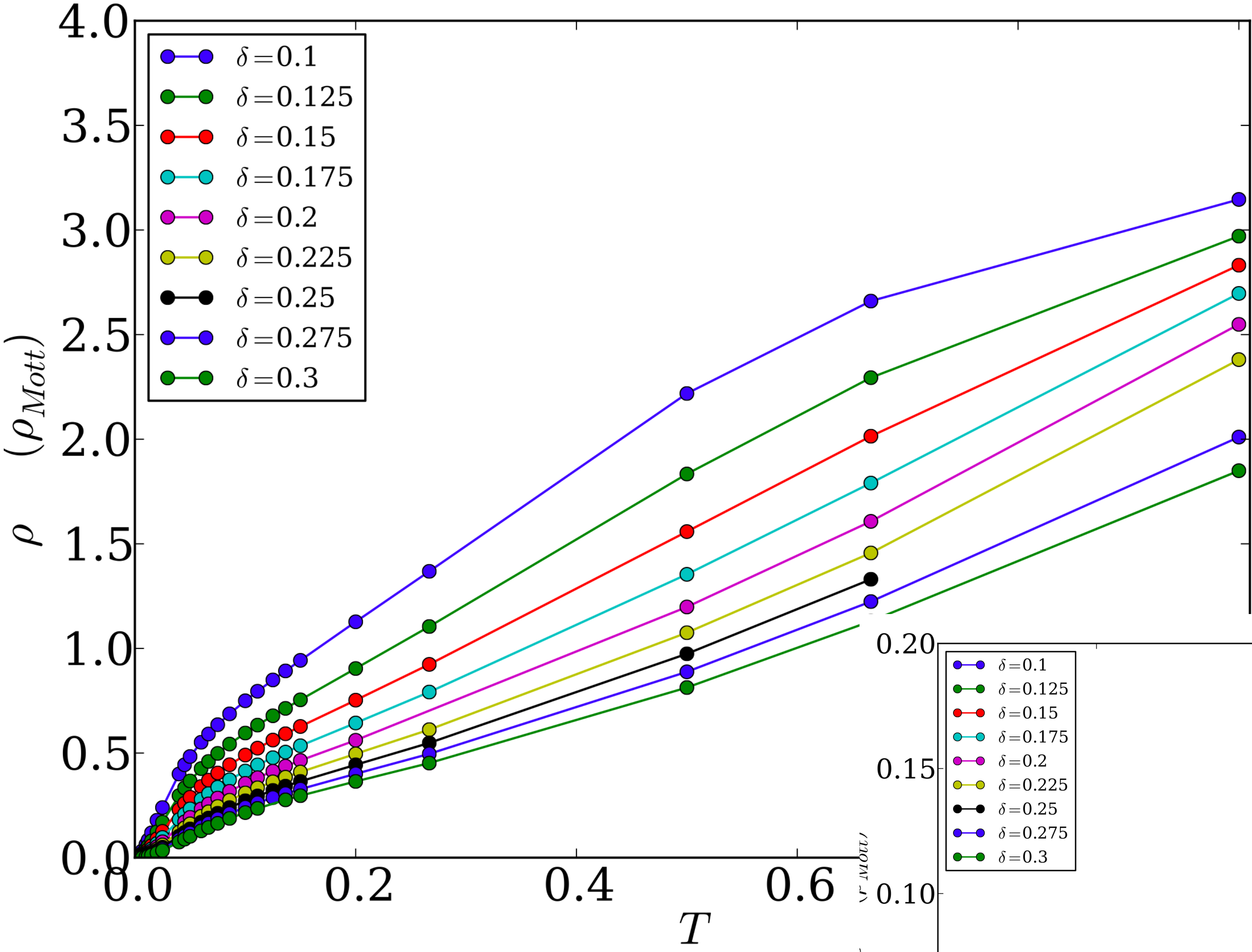
Which excitations contribute to dc transport ?



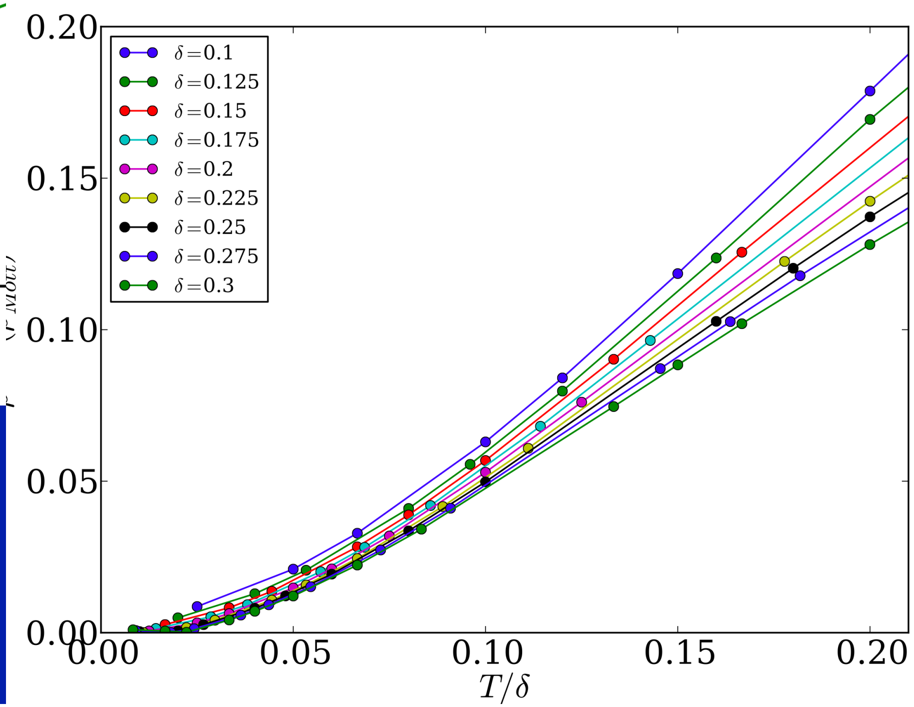
Total DOS



Clear 3-peak structure way above T_{FL}



4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
0.0
0.2
0.4
0.6
$\rho \quad (\rho_{Mott})$
T
$\delta = 0.1$
$\delta = 0.125$
$\delta = 0.15$
$\delta = 0.175$
$\delta = 0.2$
$\delta = 0.225$
$\delta = 0.25$
$\delta = 0.275$
$\delta = 0.3$



0.20
0.15
0.10
0.05
0.00
0.00
0.05
0.10
0.15
0.20
T/δ
$\delta = 0.1$
$\delta = 0.125$
$\delta = 0.15$
$\delta = 0.175$
$\delta = 0.2$
$\delta = 0.225$
$\delta = 0.25$
$\delta = 0.275$
$\delta = 0.3$

Ex: Ruthenates (remember: 3 FS sheets)

ab-plane:

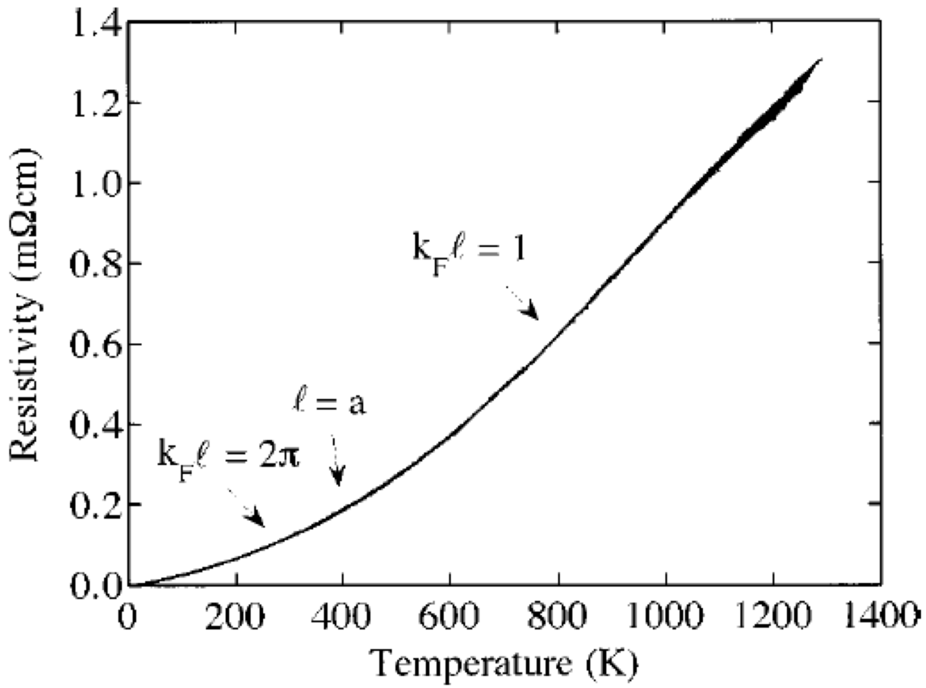


FIG. 1. The in-plane resistivity of Sr_2RuO_4 from 4 to 1300 K. Three criteria for the Mott-Ioffe-Regel limit are marked on the graph, and there is no sign of resistivity saturation, so Sr_2RuO_4 is a ‘‘bad metal’’ at high temperatures, even though it is known to be a very good metal at low temperatures.

- resistivity does cross IRM value

- Nothing dramatic is seen in ρ upon crossing IRM

Tyler,Maeno, McKenzie
PRB 58 R10107 (1998)

Outline of todays' lecture

- Basics of thermoelectric effects/coefficients
- Seebeck from Kubo
- DMFT results: S(T) for doped Hubbard in the 3 temperature regimes
- Low-T behaviour and key importance of particle-hole asymmetry
- High-T behaviour: Heike's formula(s) vs. Kelvin relation

Thermoelectric effects: basics

Grand-canonical potential $\Omega(T,\mu) = -k_B T \ln Z_G$

Particle-number and Entropy: $s = -\frac{\partial \Omega}{\partial T}|_\mu \;,\; n = -\frac{\partial \Omega}{\partial \mu}|_T$

Particle and entropy currents: linear response

$$j_n = -\mathcal{L}_{11} \nabla \mu - \mathcal{L}_{12} \nabla T$$
$$j_s = -\mathcal{L}_{21} \nabla \mu - \mathcal{L}_{22} \nabla T$$

Onsager's relation: $\mathcal{L}_{12} = \mathcal{L}_{21}$

Electrical and heat currents:

Heat $\delta Q = T\,ds \Rightarrow j_Q = T\,j_s$

Potential drop $\nabla\mu = q\,\nabla V = -q\vec{E}$

El. current:

$$j_e = q\,j_n \quad (q \equiv -e)$$

$$j_e = q^2\,\mathcal{L}_{11}\vec{E} - q\mathcal{L}_{12}\nabla T$$

$$j_Q = Tq\,\mathcal{L}_{21}\vec{E} - T\mathcal{L}_{22}\nabla T$$

Ashcroft-Mermin's notations: $L_{11} = q^2\mathcal{L}_{11}\ ,\ L_{22} = T\mathcal{L}_{22}$

$$L_{12} = q\mathcal{L}_{12}\ ,\ L_{21} = Tq\mathcal{L}_{21}$$

Electrical conductivity: $\nabla T = 0 \Rightarrow \sigma = q^2\mathcal{L}_{11} = L_{11}$

Thermal conductivity: $j_n = 0 \Rightarrow j_Q = \kappa(-\nabla T)$

(no particle current)

$$\kappa = T\left[\mathcal{L}_{22} - \frac{\mathcal{L}_{12}\mathcal{L}_{21}}{\mathcal{L}_{11}}\right]$$

Two thermoelectric effects

1. Seebeck effect: thermal gradient induces a voltage drop between the two ends of a conductor

$$j_e = 0 \Rightarrow \vec{E} = S\,\vec{\nabla}T \quad , \quad S \equiv \frac{\mathcal{L}_{12}}{q\mathcal{L}_{11}}$$

2. Peltier effect: electrical current induces heat current

$$\nabla T = 0 \Rightarrow j_Q = \Pi\, j_e \quad , \quad \Pi \equiv T\,\frac{\mathcal{L}_{21}}{q\mathcal{L}_{11}}$$

Kelvin's relation (consequence of Onsager): $\Pi = T\,S$

<u>The Seebeck coefficient S measures the entropy per charge flow:</u>

$$j_s = S\,j_e - \frac{\kappa}{T}\nabla T$$

(eliminating μ)

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 $\langle j\ j \rangle$, $\langle j\ j_E \rangle$, $\langle j_E\ j_E \rangle$

At the end of the day…

(neglecting vertex → exact in DMFT)

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

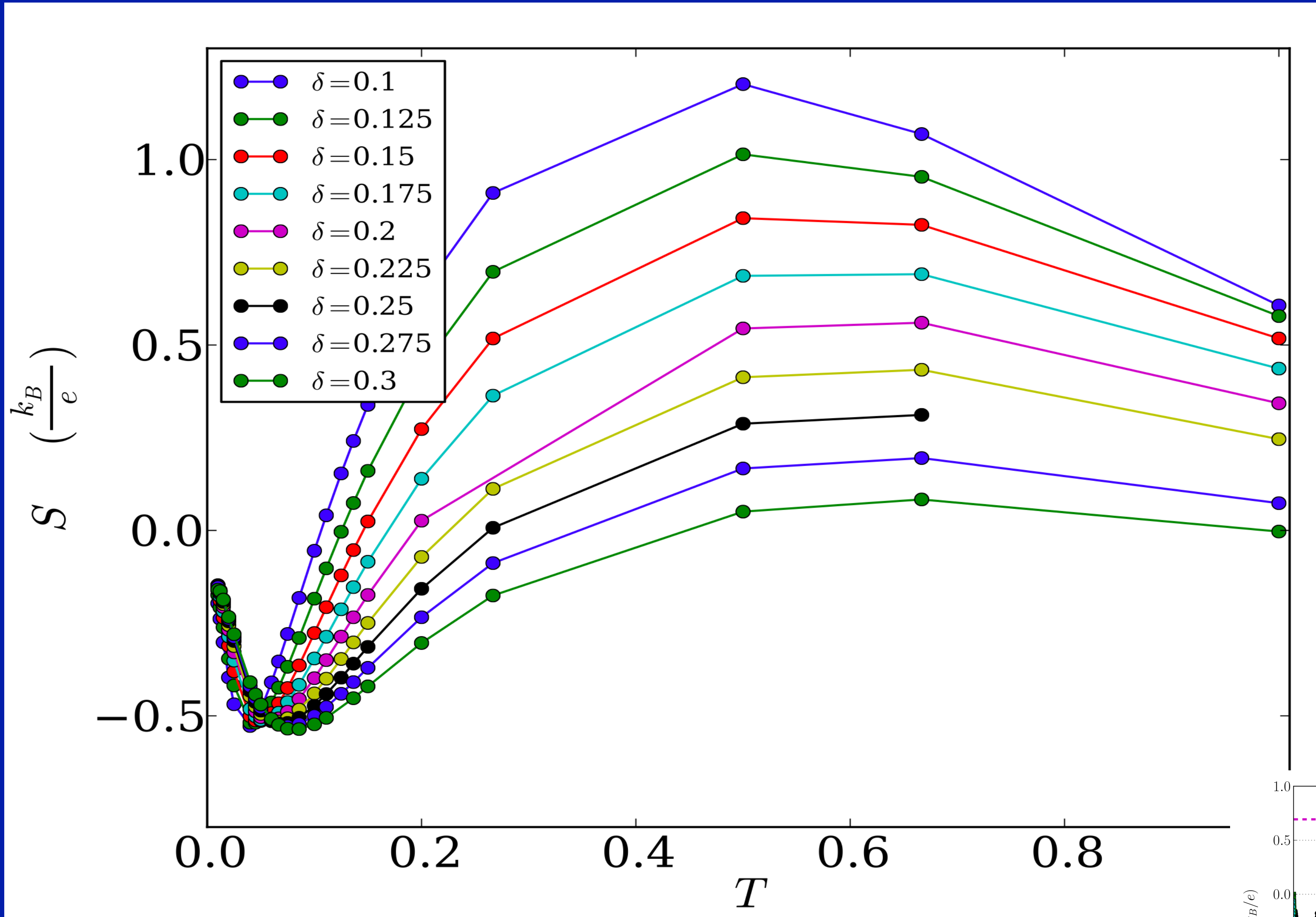
$$A_n = \frac{2\pi}{\hbar} \int d\omega \, (\beta\omega)^n f(\omega) f(-\omega) \int d\epsilon \, \Phi(\epsilon) A(\epsilon,\omega)^2$$

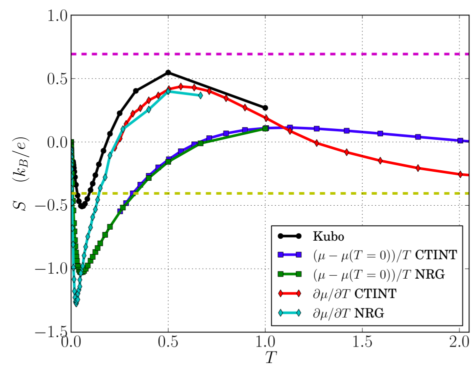
Two key observations:

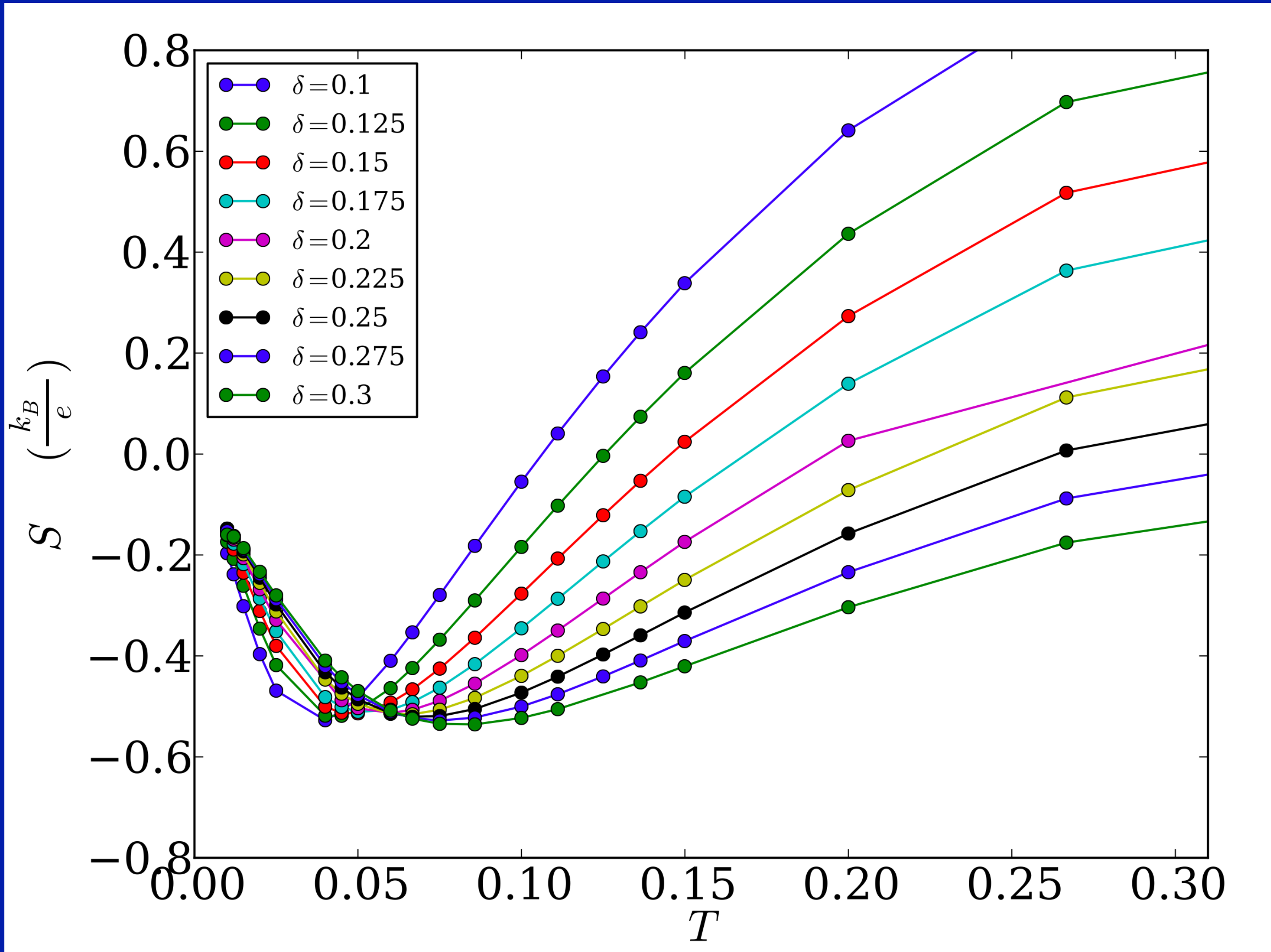
-The Seebeck is a RATIO, such as R_H . Scattering is not requested to get a non-zero S (although scattering rate does not entirely cancel, actually – see below) . In other words: a uniform entropy current can exist without entropy production (ds/dt=0)

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

Seebeck from DMFT, doped Hubbard

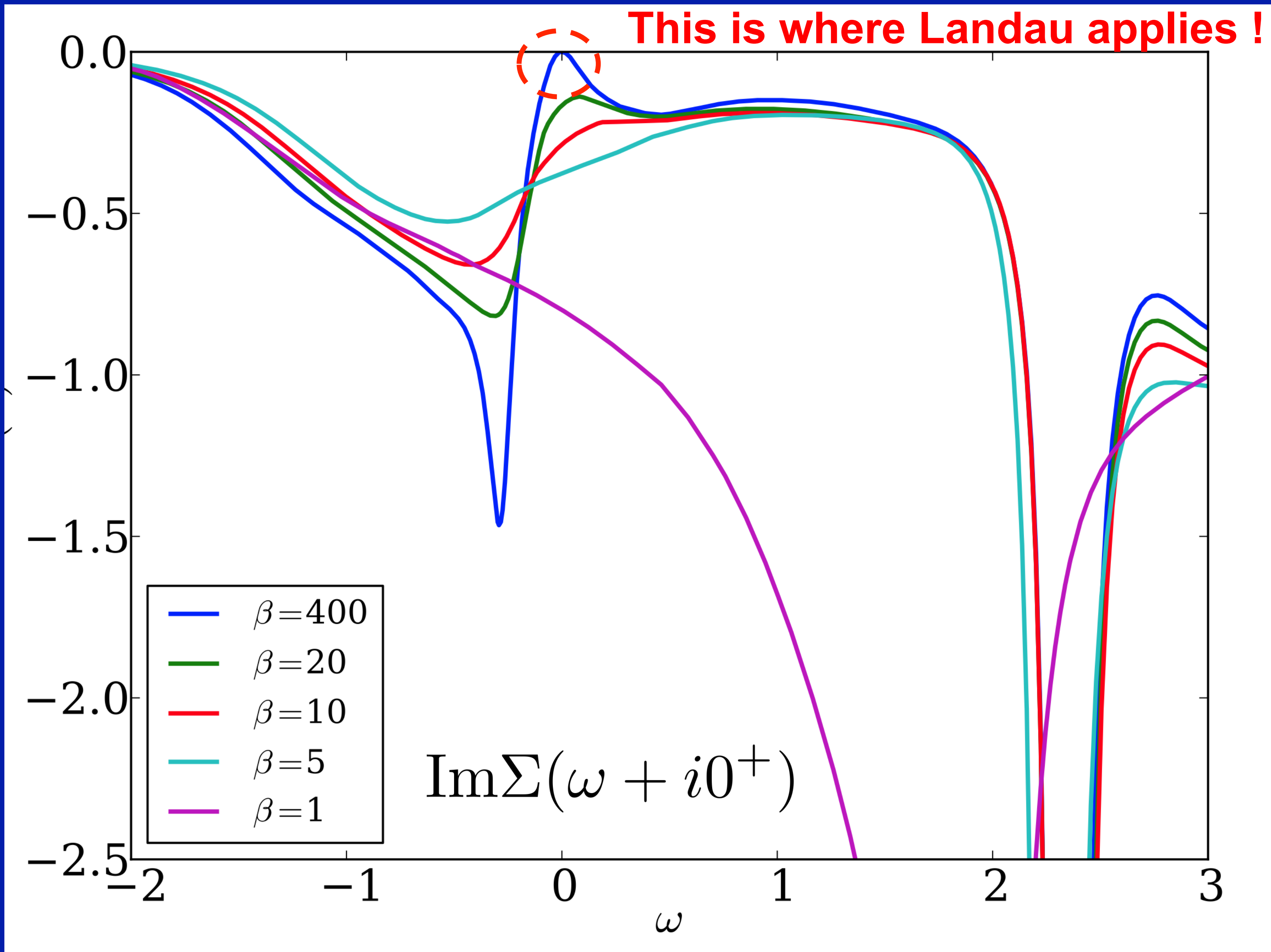






0.8
0.6
0.4
0.2
0.0
−0.2
−0.4
−0.6
−0.8
0.00
0.05
0.10
0.15
0.20
0.25
0.30
T
$S \quad \left(\frac{k_B}{e}\right)$
$\delta = 0.1$
$\delta = 0.125$
$\delta = 0.15$
$\delta = 0.175$
$\delta = 0.2$
$\delta = 0.225$
$\delta = 0.25$
$\delta = 0.275$
$\delta = 0.3$

1. Fermi Liquid Regime



This is where Landau applies !
0.0
−0.5
−1.0
−1.5
−2.0
−2.5
−2
−1
0
1
2
3
ω
β=400
β=20
β=10
β=5
β=1
ImΣ(ω + i0+)

Resistivity in the FL regime: analytics

Low ω,T scaling form of scattering rate:

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

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

→ On blackboard

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

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

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 <u>dominant</u> 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

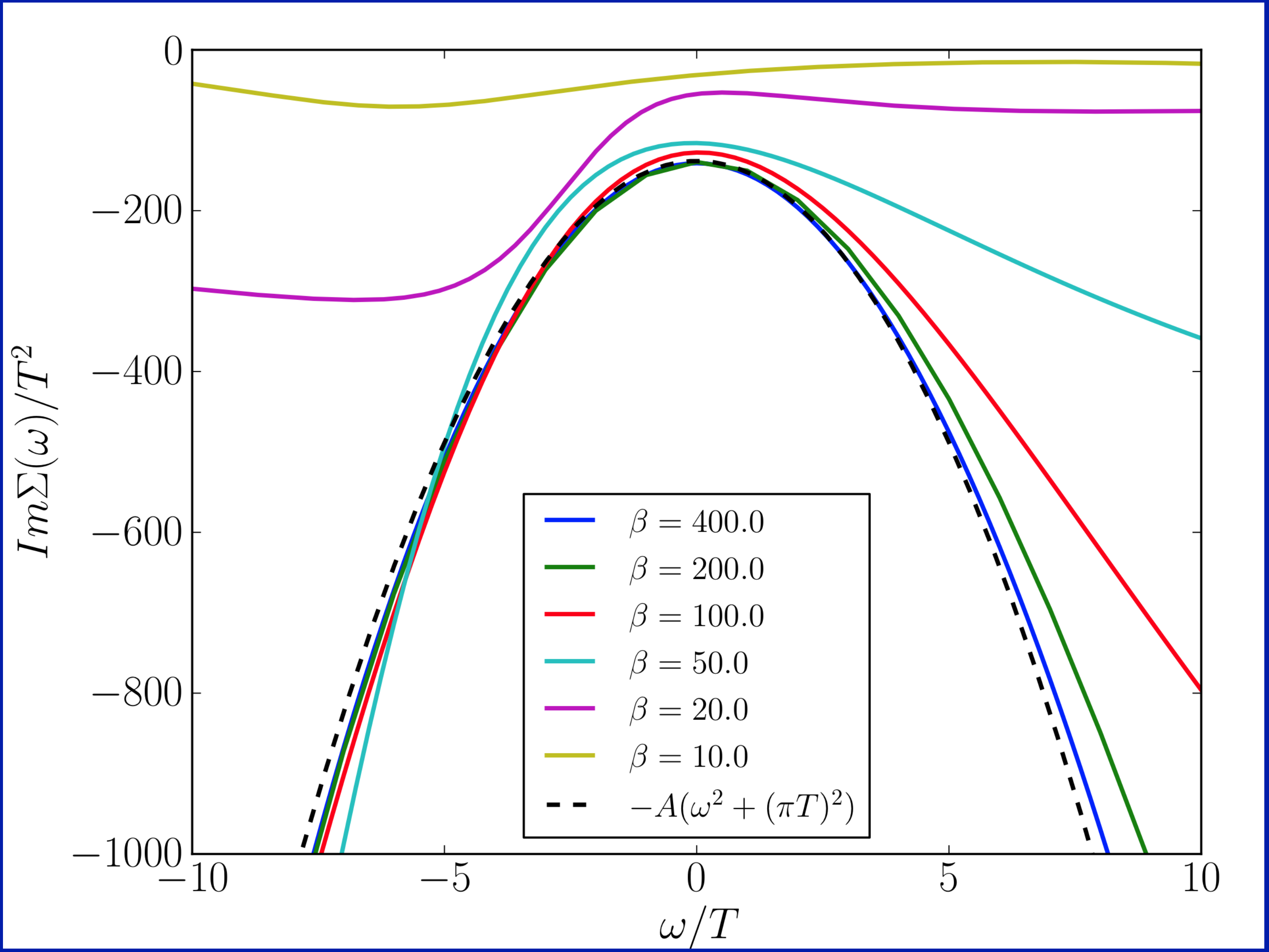
$$\Sigma''(\omega) = \Sigma^{(2)}(\omega) + \Sigma^{(3)}(\omega) + \cdots$$

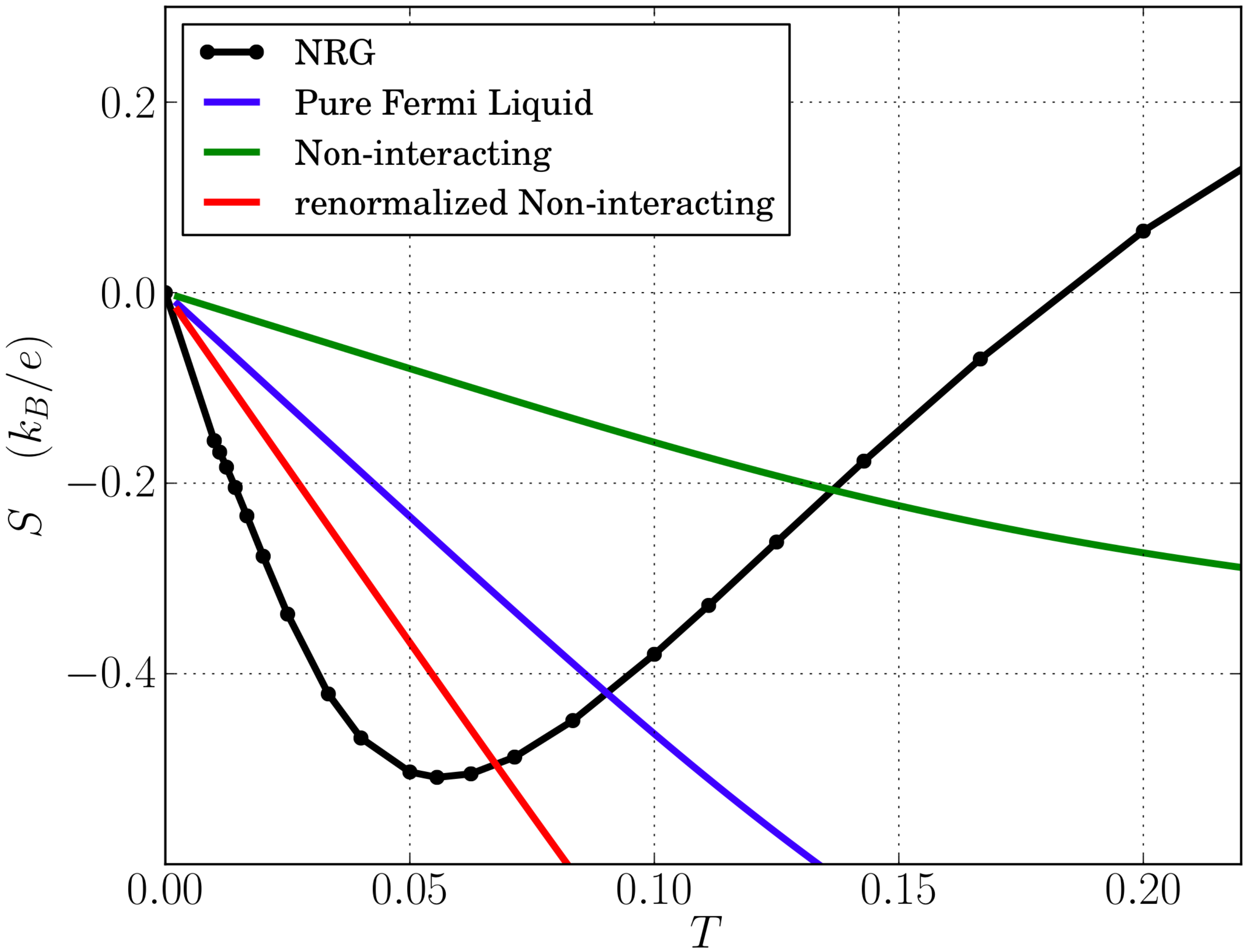
$$\Sigma^3(\omega) = \frac{(a_1\omega^3 + a_2\,\omega T^2)}{Z^3}$$

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

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

Particle-hole asymetry of the scattering rate

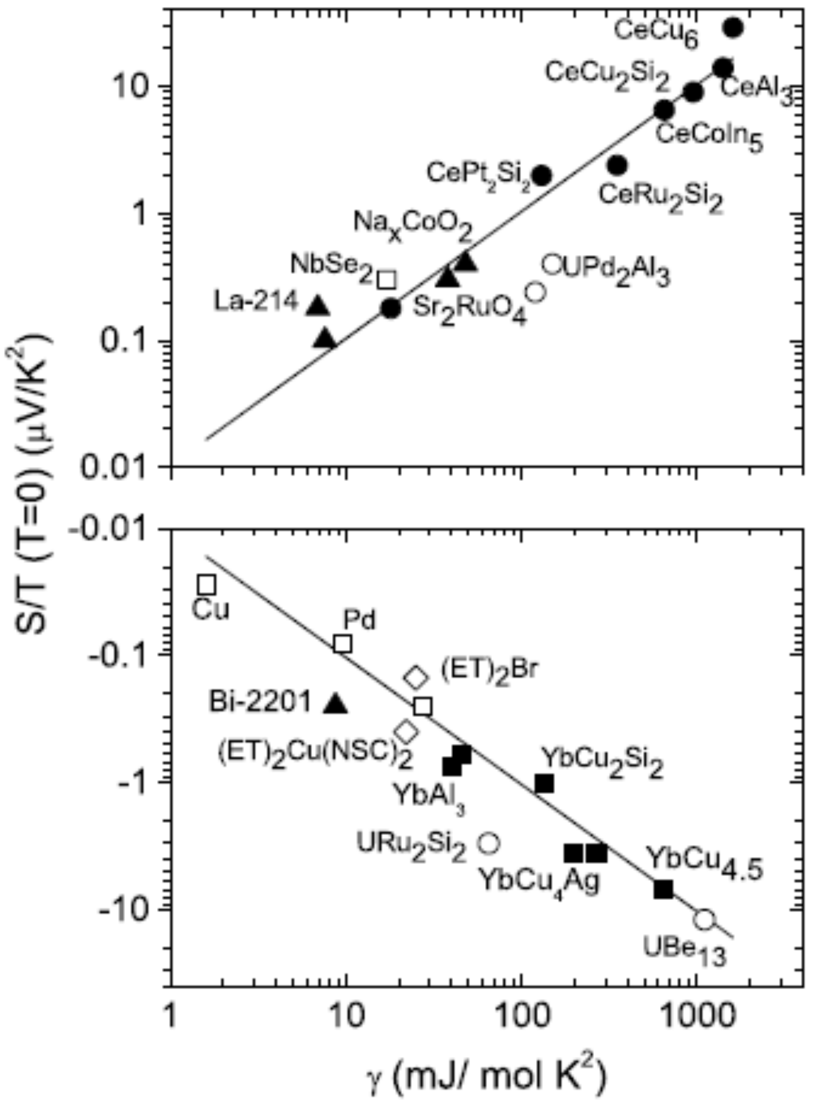




NRG
Pure Fermi Liquid
Non-interacting
renormalized Non-interacting
0.2
0.0
−0.2
−0.4
$S \quad (k_B/e)$
0.00
0.05
0.10
0.15
0.20
T

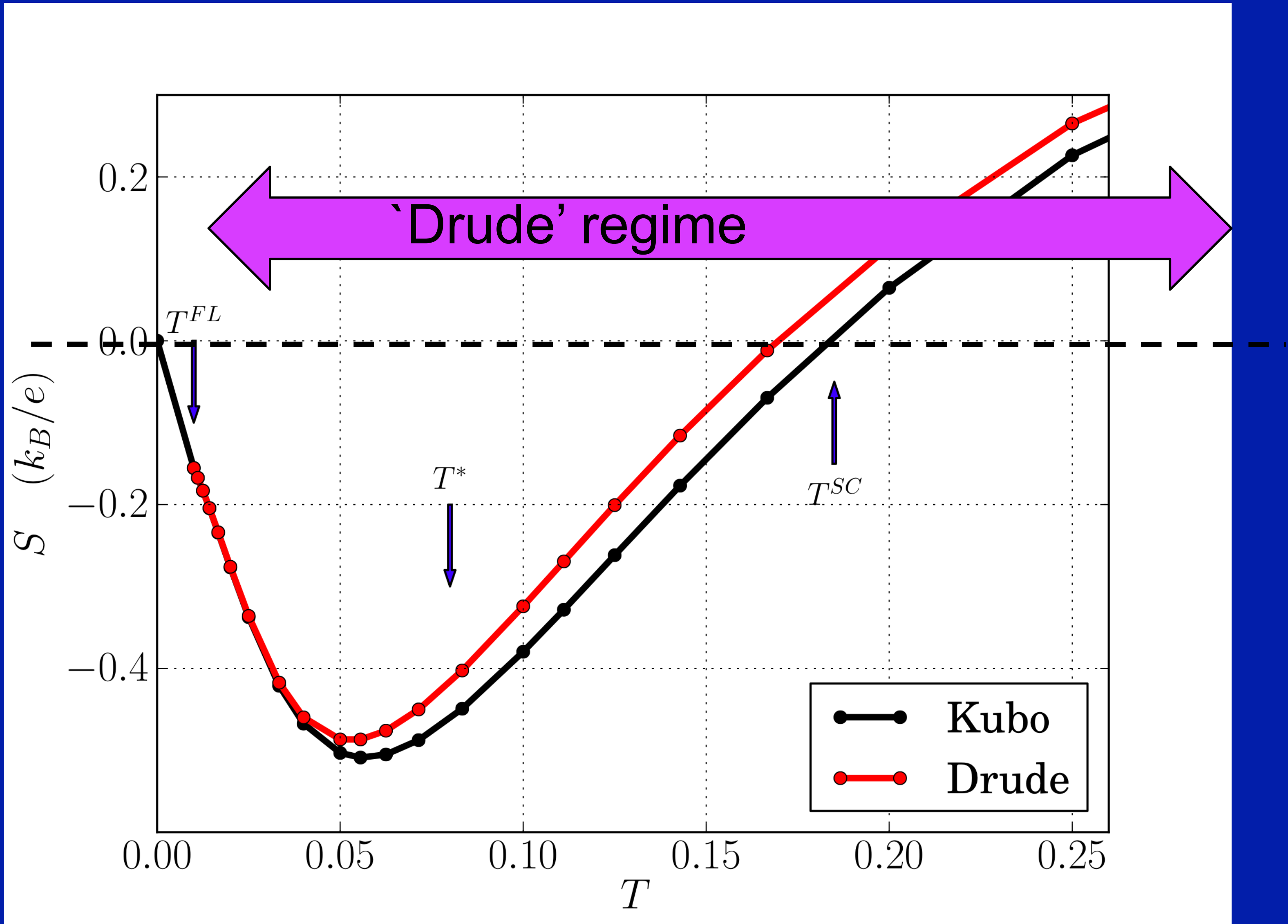
INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER
J. Phys.: Condens. Matter 16 (2004) 5187–5198 PII: S0953-8984(04)80129-2

The Behnia-Jaccard-Flouquet law:
S/Tγ



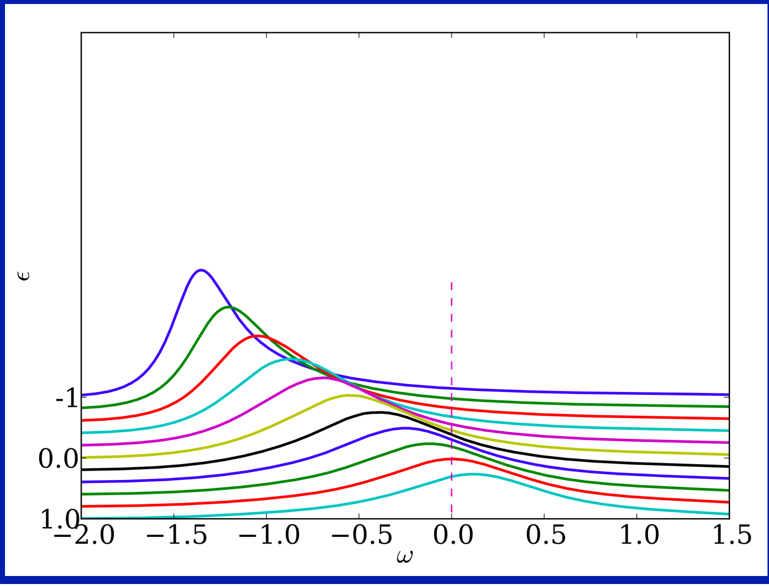
$$\frac{S}{\gamma T} = -\frac{3}{|e|}\frac{1}{D(\mu_0)}\left[\frac{\Phi'(\mu_0)}{\Phi(\mu_0)}\frac{E_2^1}{E_0^1} - \frac{a_1 E_4^2 + a_2 E_2^2}{\gamma_0 E_0^1}\right]$$

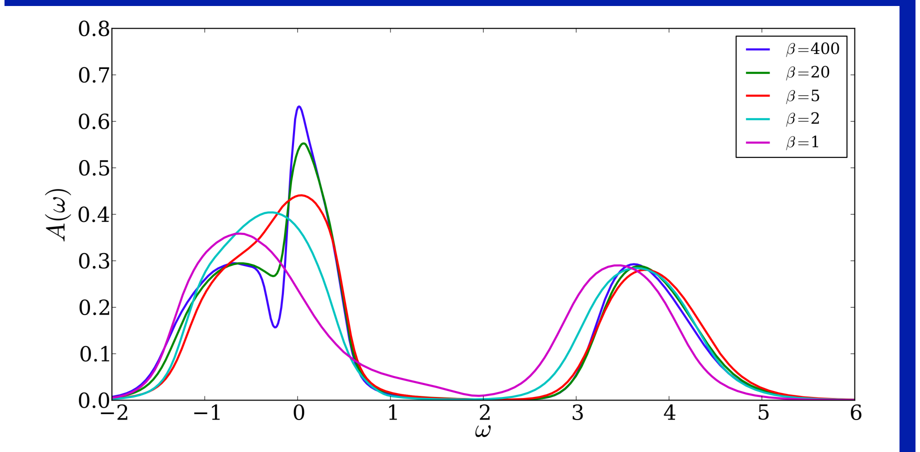
2. Seebeck in `Drude' regime: minimum dominated by electron-like Drude quasiparticles

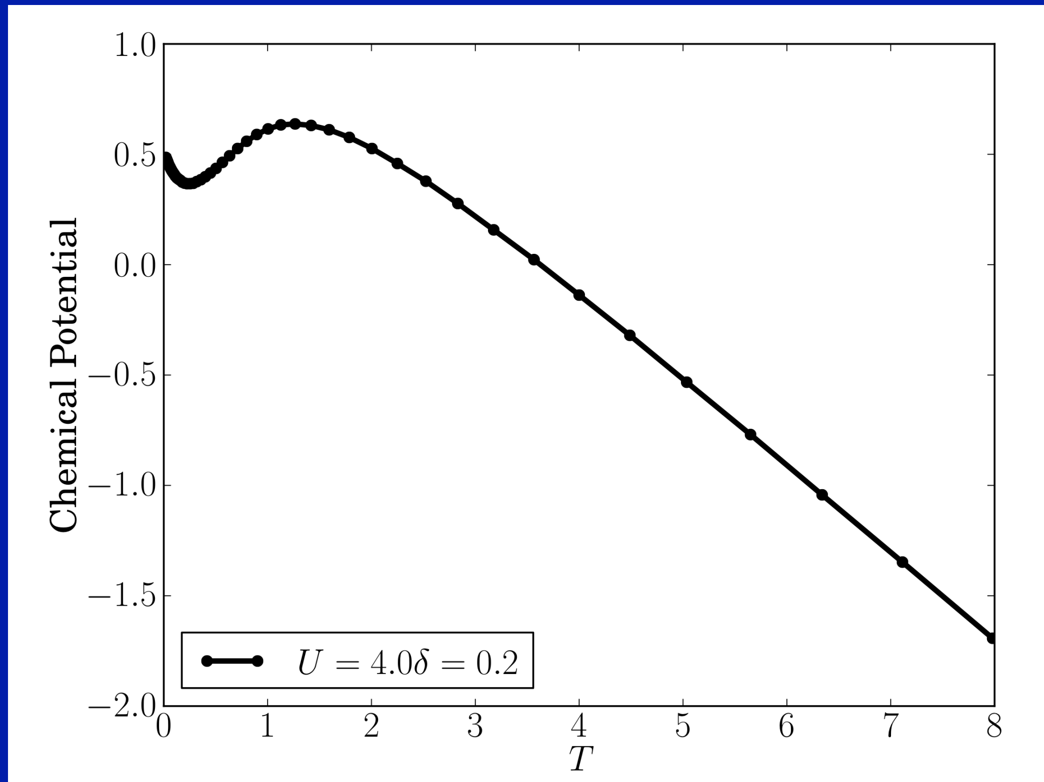


3. High-temperature regime(s):
Heike's limit(s) and Kelvin formula
(see also seminar by Sriram Shastry)

3. 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

$$\boxed{\alpha \equiv \beta\mu}$$

$$\tilde{\rho}(\omega, \epsilon) = \rho(\omega - \mu, \epsilon).$$

G.Palsson,
PhD thesis
Rutgers

Hence the coefficients A_n from §3.4 become[4]:

$$A_n = \frac{\pi N}{4} \int d\omega \frac{(\beta\omega - \alpha)^n}{\cosh^2(\frac{\beta\omega-\alpha}{2})} \int d\epsilon \tilde{\rho}^2(\omega, \epsilon)\Phi(\epsilon).$$

We now expand the hyperbolic cosine in Taylor series around $\beta = 0$.

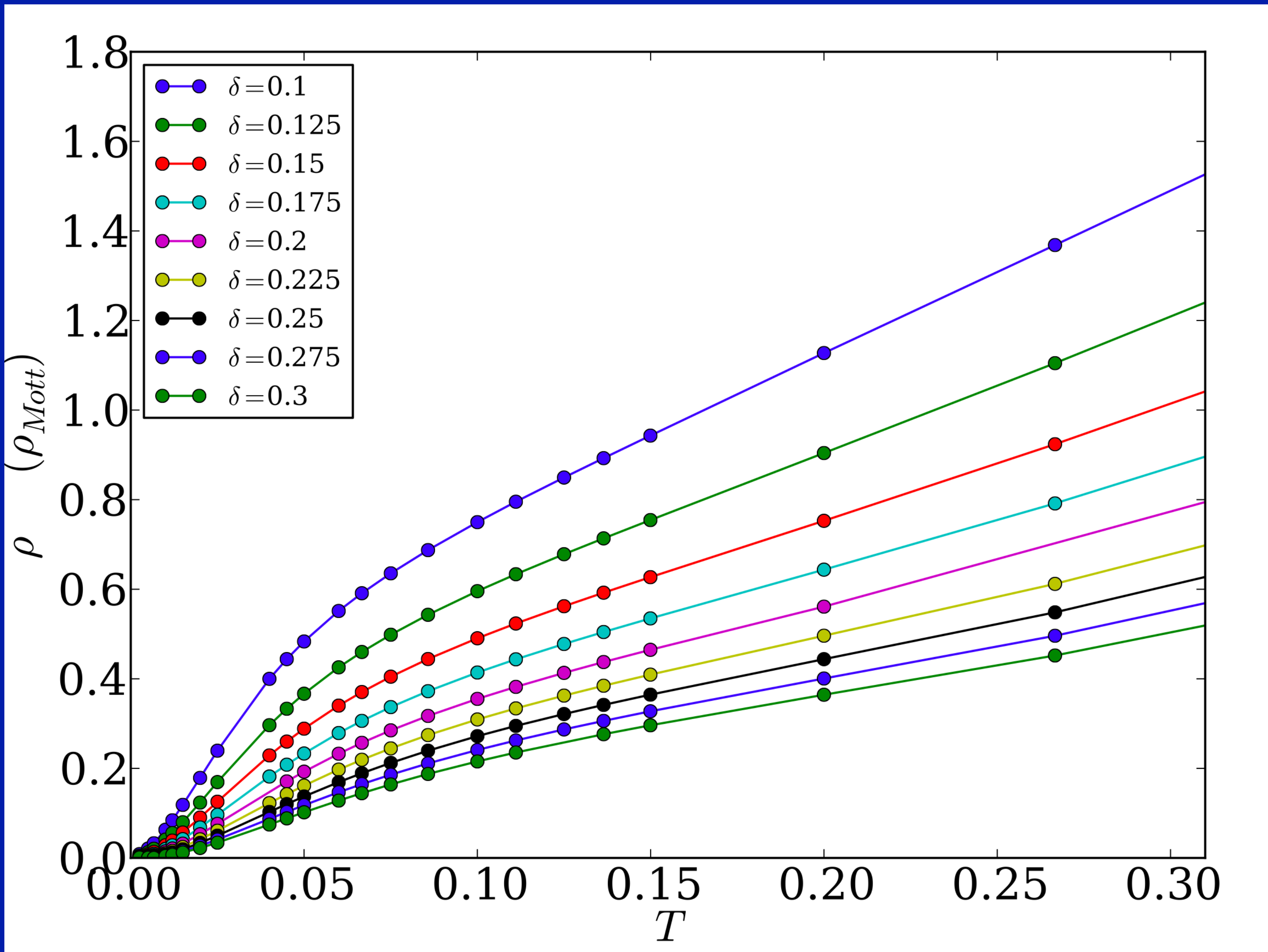
$$\frac{1}{\cosh^2(\frac{\beta\omega-\alpha}{2})} = \frac{1}{\cosh^2(\frac{\alpha}{2})} \left(1 + \beta\omega \tanh(\frac{\alpha}{2}) + \frac{\omega^2\beta^2}{4}\left[3\tanh(\frac{\alpha}{2}) - 1\right]\right).$$

Before we go any further we also define

$$\gamma_n = \int d\epsilon d\omega \omega^n \tilde{\rho}^2(\omega, \epsilon)\Phi(\epsilon) \quad \text{and let} \quad \tau = \tanh(\frac{\alpha}{2}) \quad \text{and} \quad \zeta = \frac{1}{4\cosh^2(\frac{\alpha}{2})}.$$

T-linear resistivity above Ioffe Regel Mott value
(but actually also applies below as one starts coming
Oout of Drude regime)

$$\rho(T) \sim \frac{T}{\gamma_0\zeta}$$



δ=0.1
δ=0.125
δ=0.15
δ=0.175
δ=0.2
δ=0.225
δ=0.25
δ=0.275
δ=0.3
$\rho \quad (\rho_{Mott})$
T
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
0.00
0.05
0.10
0.15
0.20
0.25
0.30

Hi-T expansion, Seebeck:

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

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

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

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

P. M. Chaikin*
Department of Physics, University of California, Los Angeles, California 90024

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

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

The two hi-T (Heike's) limits

1. D < T << U

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

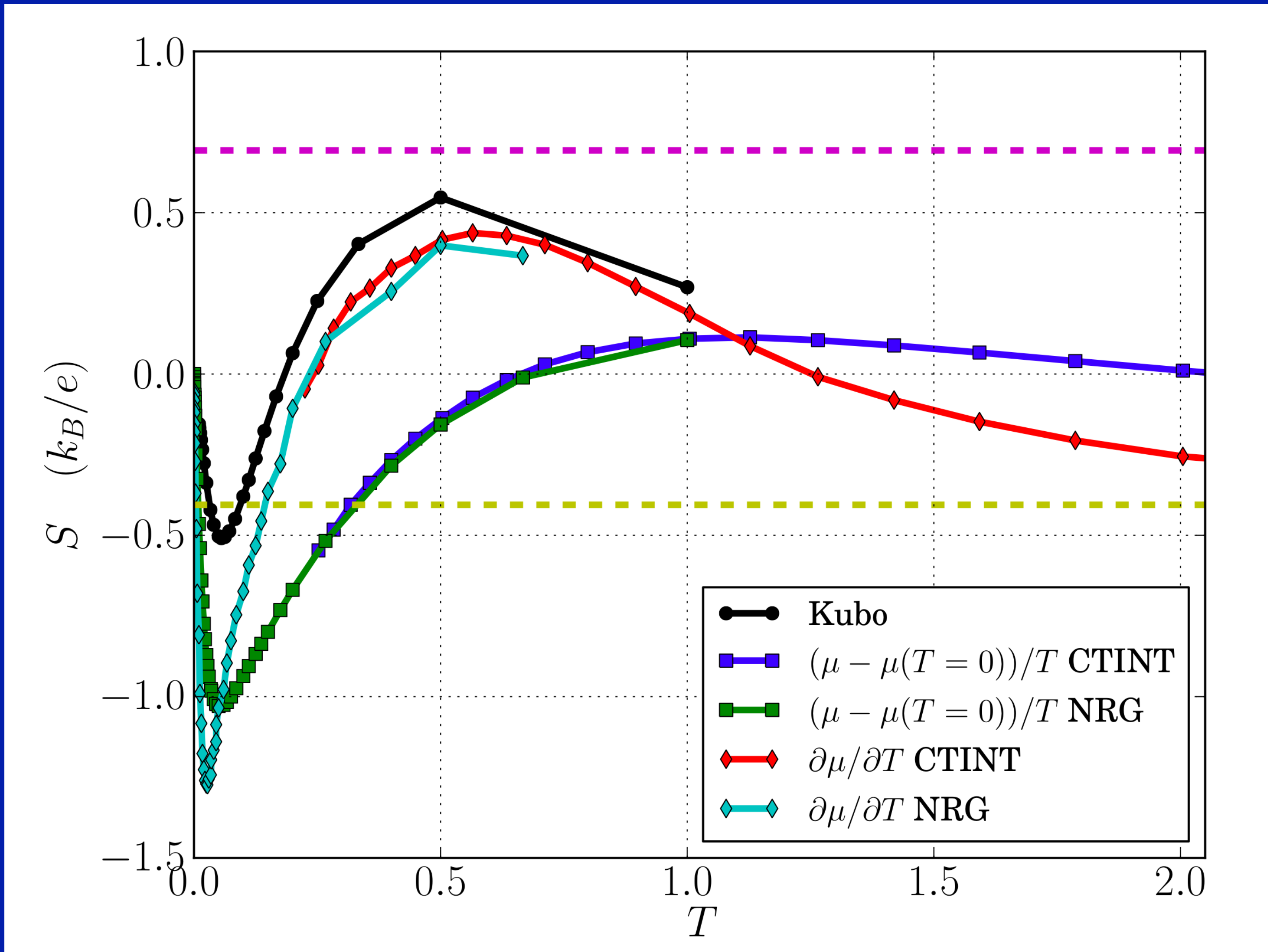
$$s/k = -(1-n)\ln(1-n) - n\ln n/2$$

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

2. T > U

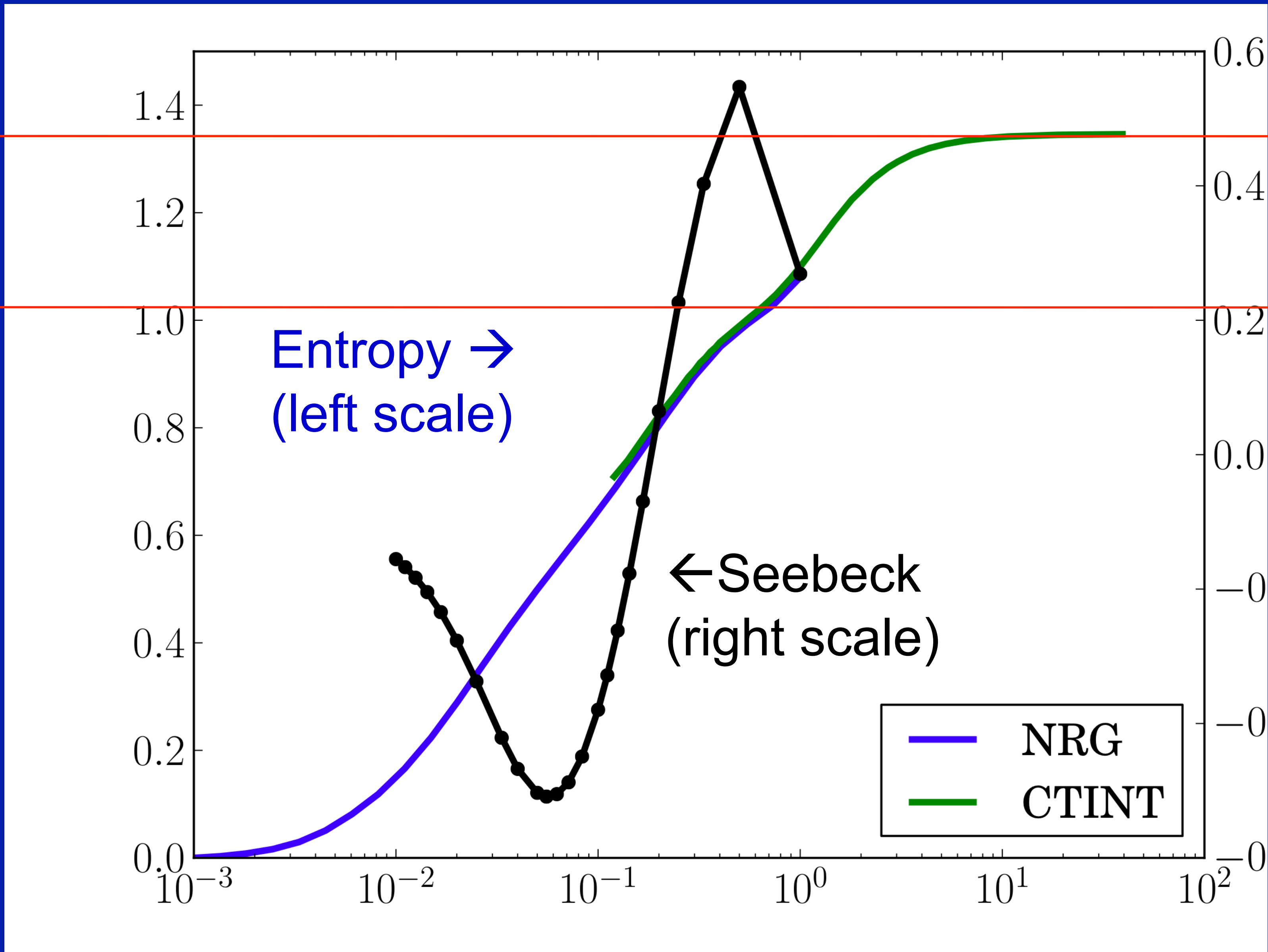
$$s/k = -2\left[\frac{n}{2}\ln\frac{n}{2} + \frac{1-n}{2}\ln\frac{1-n}{2}\right]$$

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

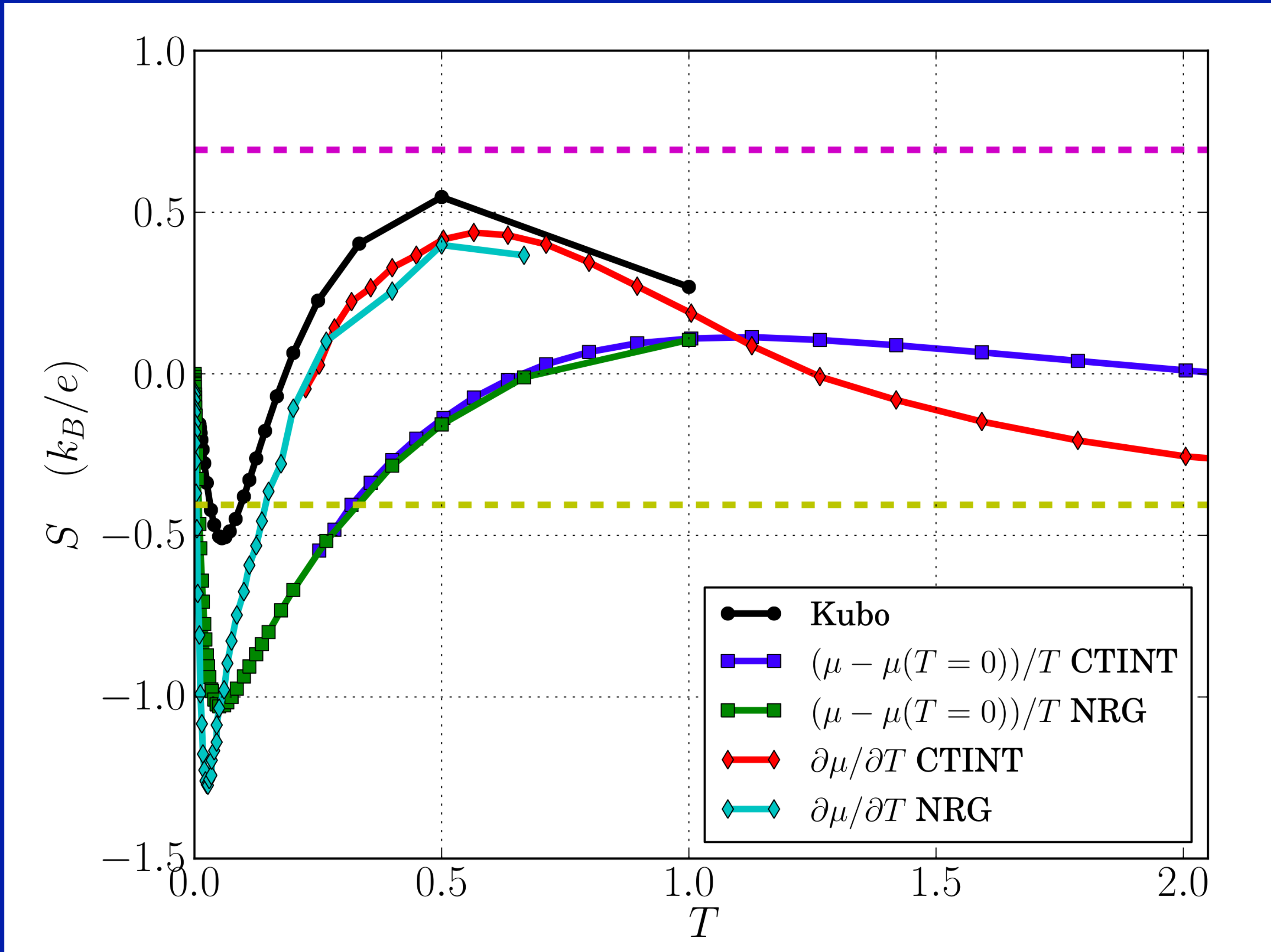


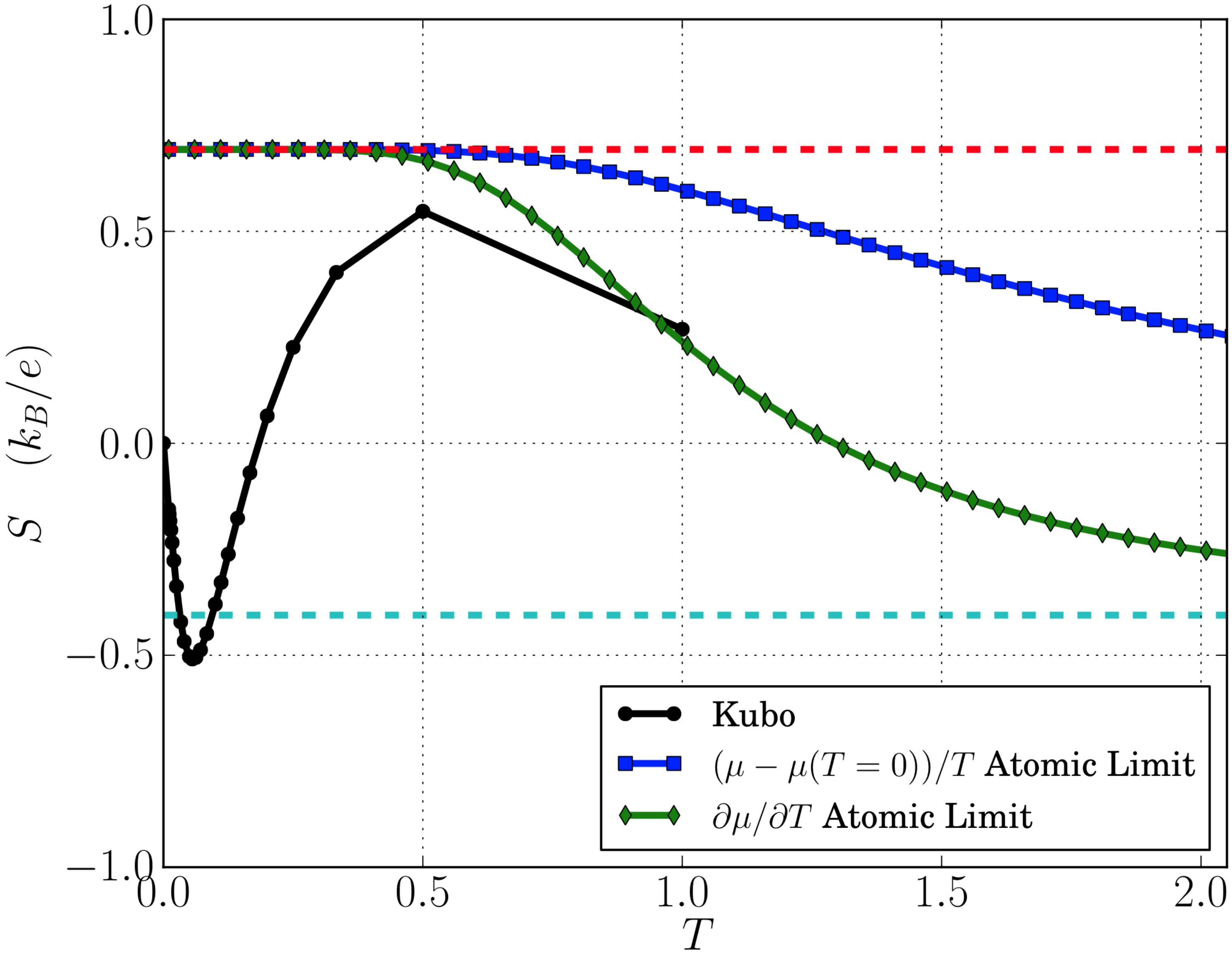
Kubo
$(\mu - \mu(T=0))/T$ CTINT
$(\mu - \mu(T=0))/T$ NRG
$\partial\mu/\partial T$ CTINT
$\partial\mu/\partial T$ NRG
$S \quad (k_B/e)$
T
1.0
0.5
0.0
−0.5
−1.0
−1.5
0.0
0.5
1.0
1.5
2.0

Seebeck and Entropy



Heike's vs. Kelvin formulas → cf. Sriram Shastry's lecture





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

Main messages: Seebeck

- Seebeck sensitive probe of the different regimes: FL, Drude, hi-T regimes
- Particle-hole asymmetry crucial: not only of `fermiology' also of scattering rate
- Fermi liquid theory insufficient even for lowest T behaviour !
- Simple generalizations of hi-T formula work quite nicely, better than Heike's → possibly useful for material design ?

Comparison to exp. On LaSrTiO3

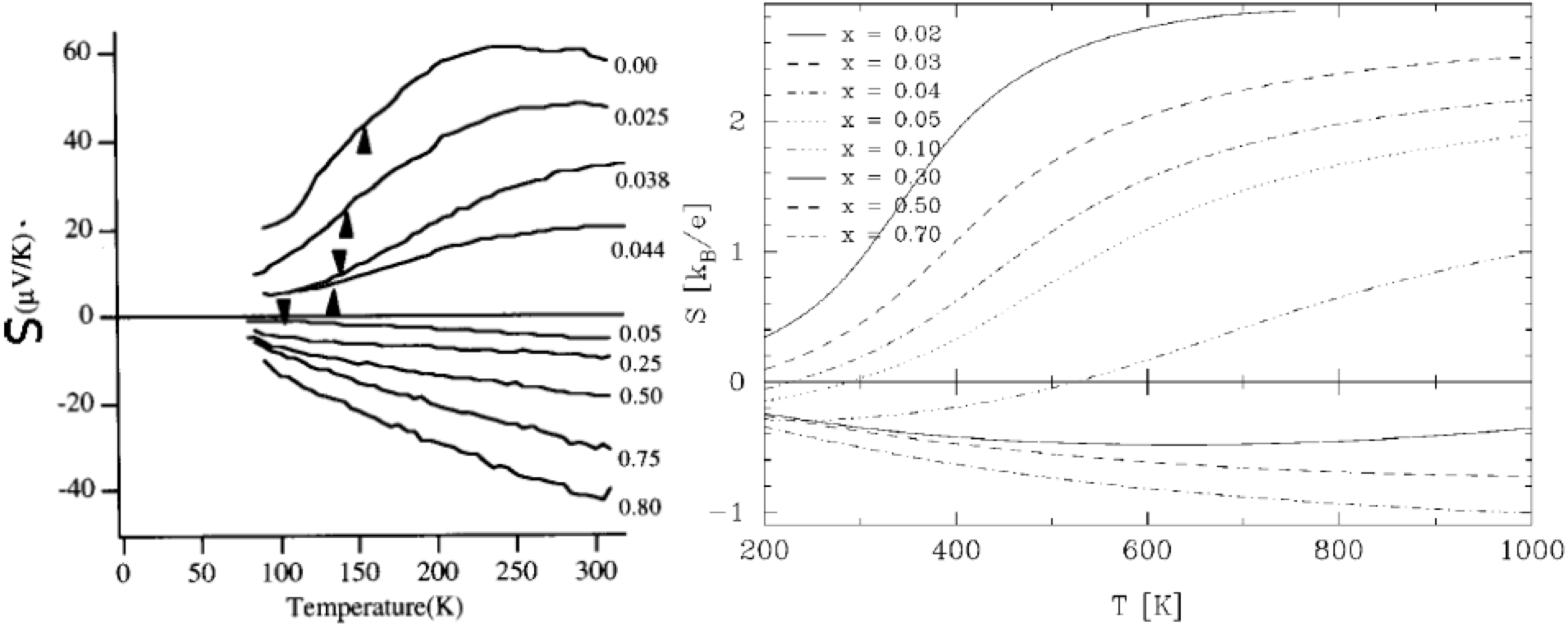


Fig. 4 Experimental (left panel) and theoretical computations of the thermoelectric power (S) of the $La_{1-x}Sr_xTiO_3$ from Refs. [16] and [9].