Heavy Carriers & Non-Drude Response in Optical Spectra of Correlated Matter

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http://vsf1.phys.rug.nl/~wwwopt/

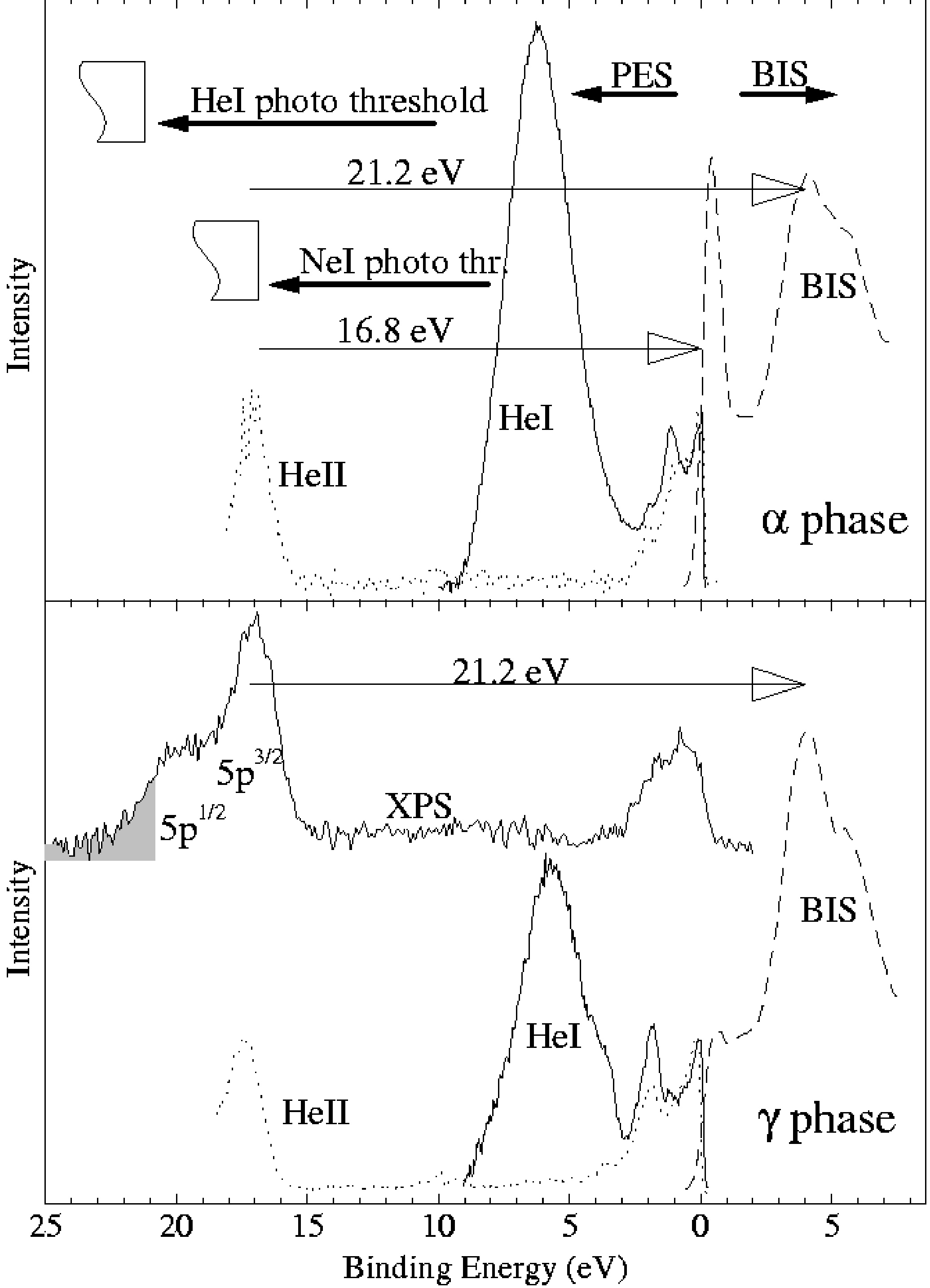
Outline

α-Ce and γ-Ce

Powerlaw of σ(ω) in HTSC

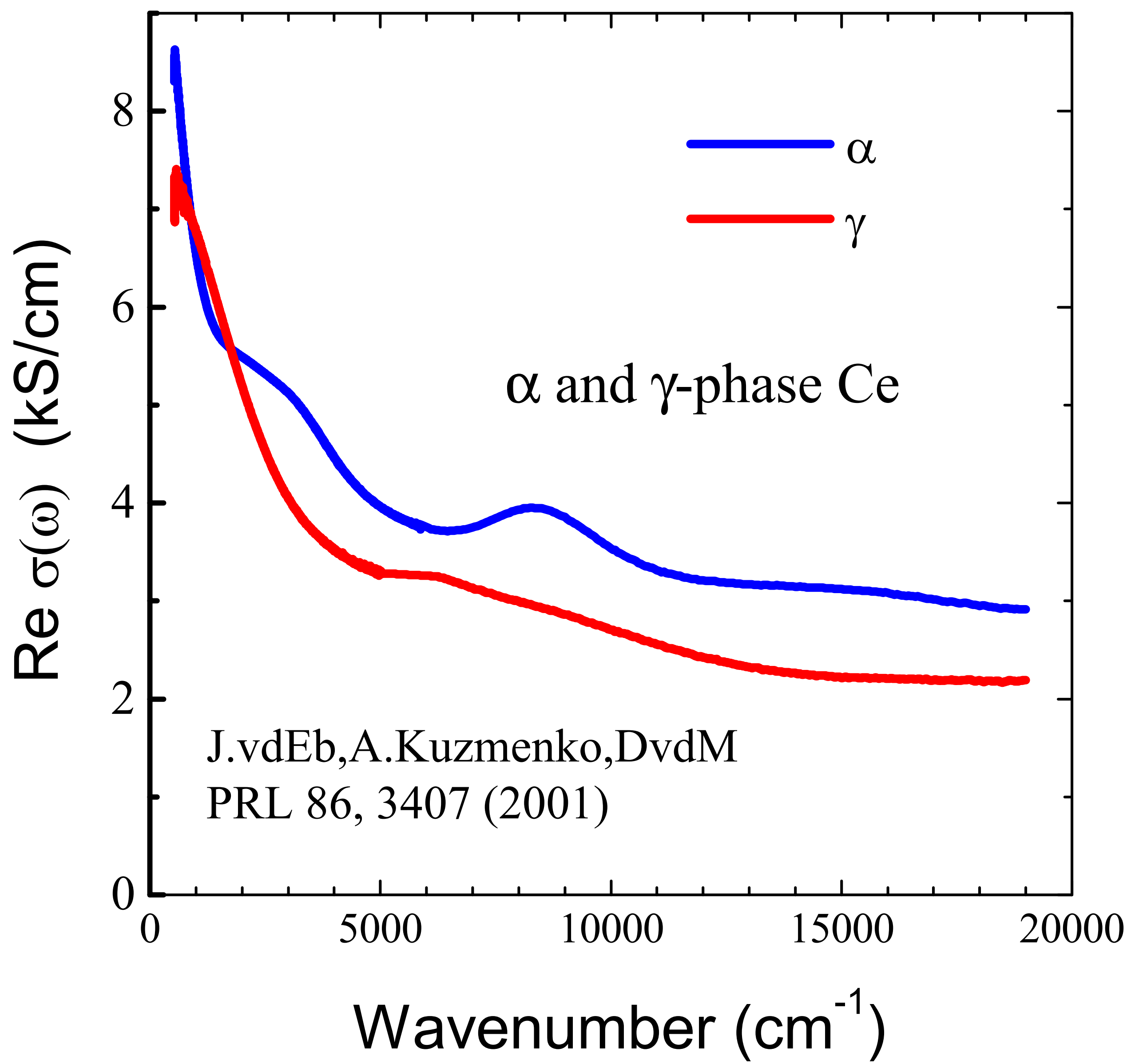
Spectral weight transfer in FeSi

Powerlaw of σ(ω) in MnSi

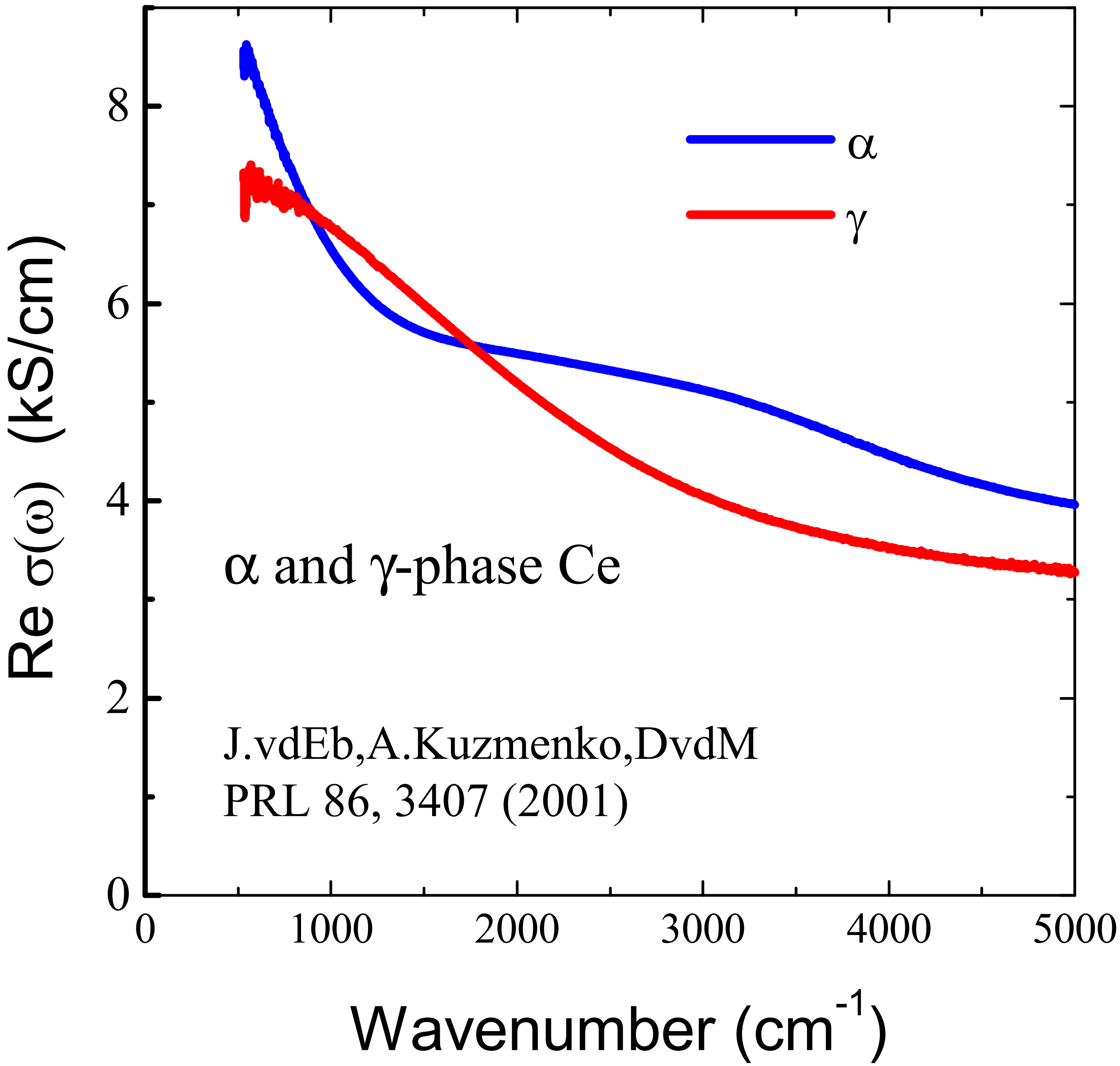


HeI photo threshold
PES
BIS
21.2 eV
NeI photo thr.
16.8 eV
BIS
HeI
HeII
α phase
Intensity
21.2 eV
5p3/2
5p1/2
XPS
BIS
HeI
HeII
γ phase
Intensity
25
20
15
10
5
0
5
Binding Energy (eV)

Optical conductivity



Optical conductivity

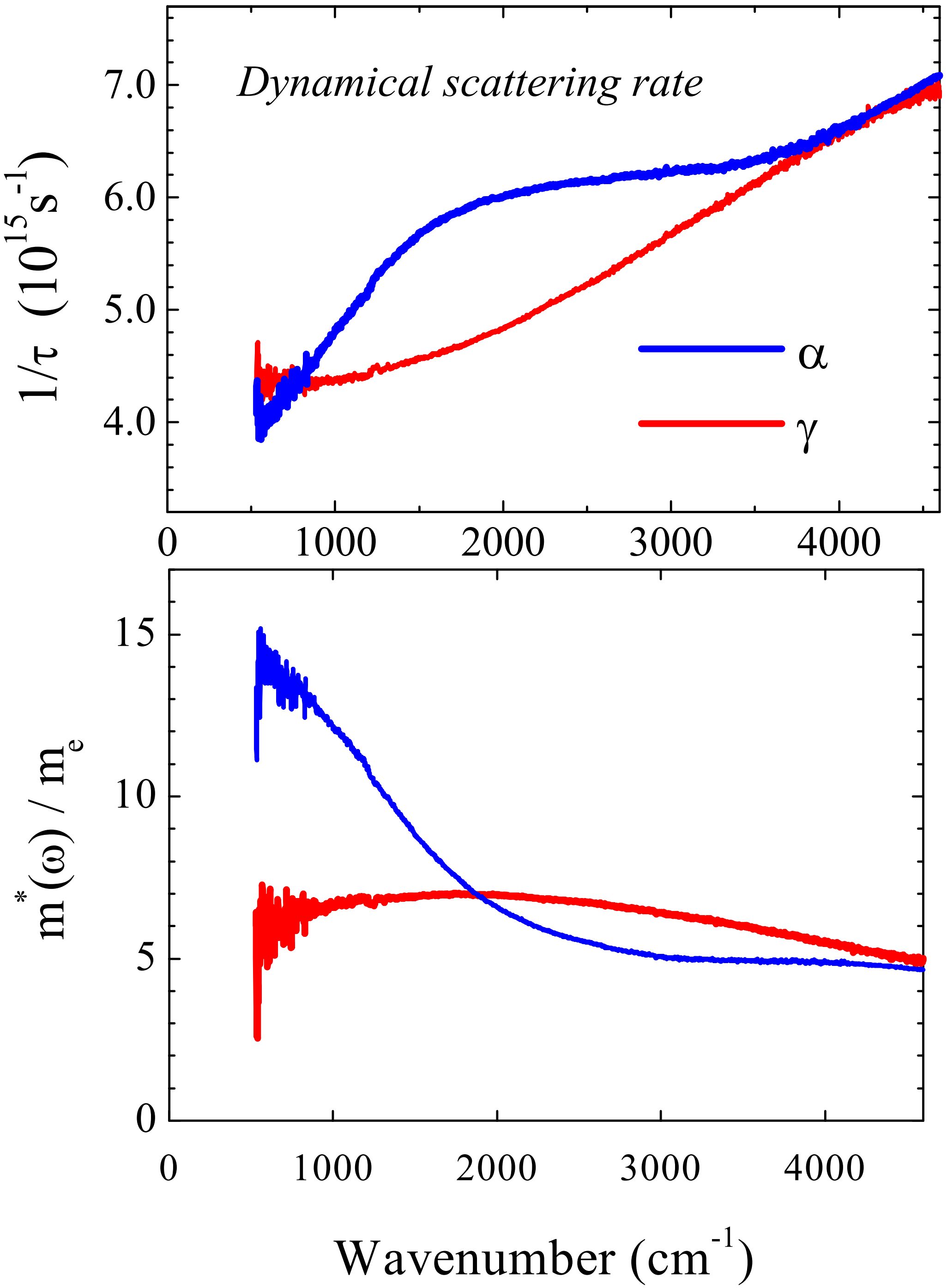


Generalized Drude Conductivity

$$\sigma(\omega) = \frac{\omega_p^2 / 4\pi}{\tau^{-1}(\omega) - i\omega m^*(\omega)/m}$$

J. W. Allen and J. C. Mikkelsen, Phys. Rev. B 15, 2952 (1977) .

$$\tau^{-1}(\omega) = \frac{\omega_p^2}{4\pi}\mathrm{Re}\left(\frac{1}{\sigma(\omega)}\right) \qquad \frac{m^*(\omega)}{m} = \frac{\omega_p^2}{4\pi\omega}\mathrm{Im}\left(\frac{-1}{\sigma(\omega)}\right)$$

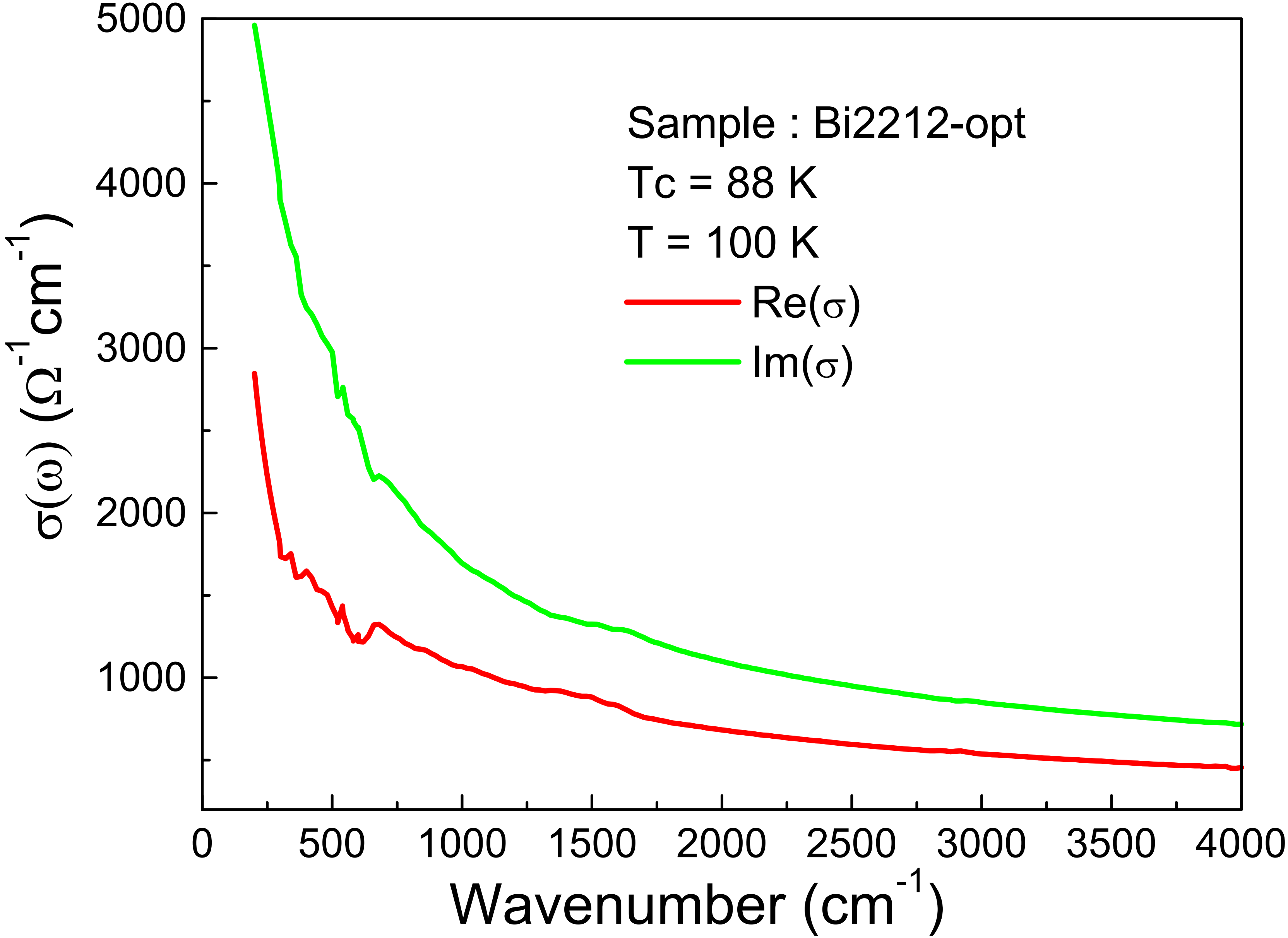


γ and α phase Ce

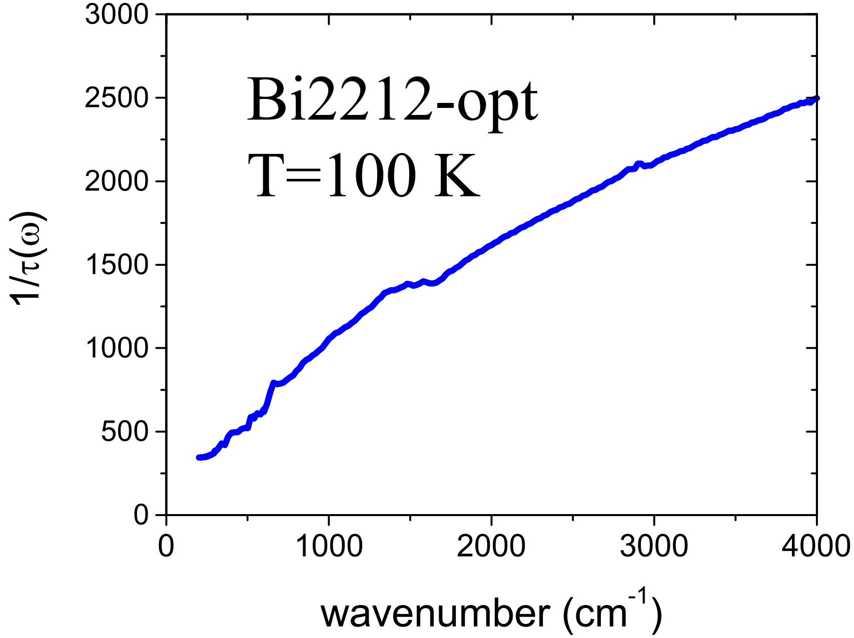
J.vdEb,A.Kuzmenko,DvdM
PRL 86, 3407 (2001)

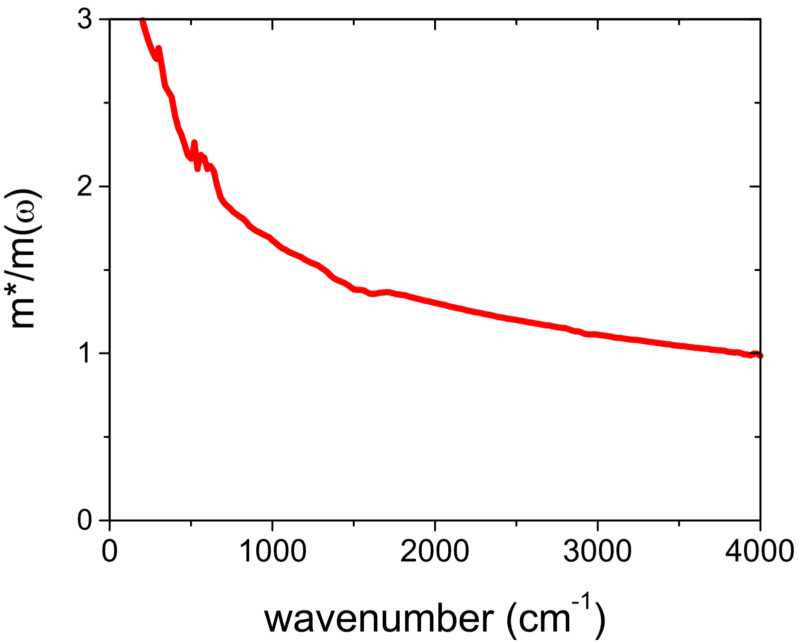
α-phase agrees with Fermi-liquid formula of P.E. Sulewski et al. PRB 38, 5338 (1988)

$$\tau^{-1}(\omega) = \frac{1}{\tau_0} + g\,\frac{\Omega_F\omega^2}{\omega^2 + \Omega_F{}^2}$$



Sample : Bi2212-opt
Tc = 88 K
T = 100 K
Re(σ)
Im(σ)
σ(ω) (Ω⁻¹cm⁻¹)
Wavenumber (cm⁻¹)
0
500
1000
1500
2000
2500
3000
3500
4000
5000
4000
3000
2000
1000





G. Thomas, (1988)
Z. Schlesinger 1990
E. El AzrakN. Bontemps, (1995)
and many other experiments

Marginal Fermi Liquid:

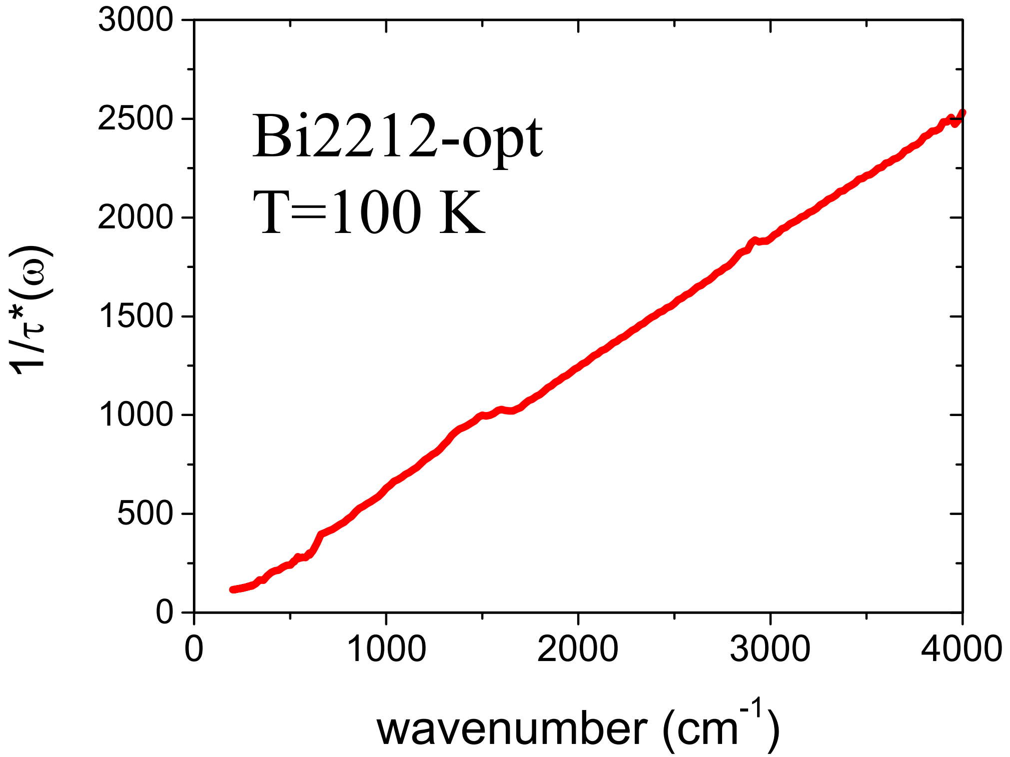
$$\sigma(\omega) = \frac{\omega_p^2 / 4\pi}{\tau^{-1}(\omega) - i\omega m^*(\omega)/m}$$

$$\tau^{-1}(\omega) = \lambda \frac{\pi}{2} \omega$$

$$\frac{m^*(\omega)}{m} - 1 = \lambda \ln \frac{\omega}{\Omega_c}$$

C. M. Varma, P. Littlewood,
S. Schmitt-Rink, E. Abrahams,
A. Ruckenstein,
PRL 63, 1996 (1989)

$$\frac{1}{\tau^*(\omega)} = \frac{m^*(\omega)}{m}\frac{1}{\tau(\omega)} = \omega\,\frac{\mathrm{Re}\,\sigma(\omega)}{\mathrm{Im}\,\sigma(\omega)}$$



$$\Rightarrow \arctan\frac{\mathrm{Re}\,\sigma(\omega)}{\mathrm{Im}\,\sigma(\omega)} = \text{Phase of } \sigma(\omega) = \text{constant}$$

Time-scale invariance

$\Downarrow$

$$\sigma(p\omega) = \Lambda\, \sigma(\omega)$$

$\Downarrow$

$$\sigma(\omega) = |C|\, e^{i\phi} (-i\, \omega)^{2\eta-1}$$

Time reversal : $\sigma(\omega) = \sigma^{*}(-\omega)$

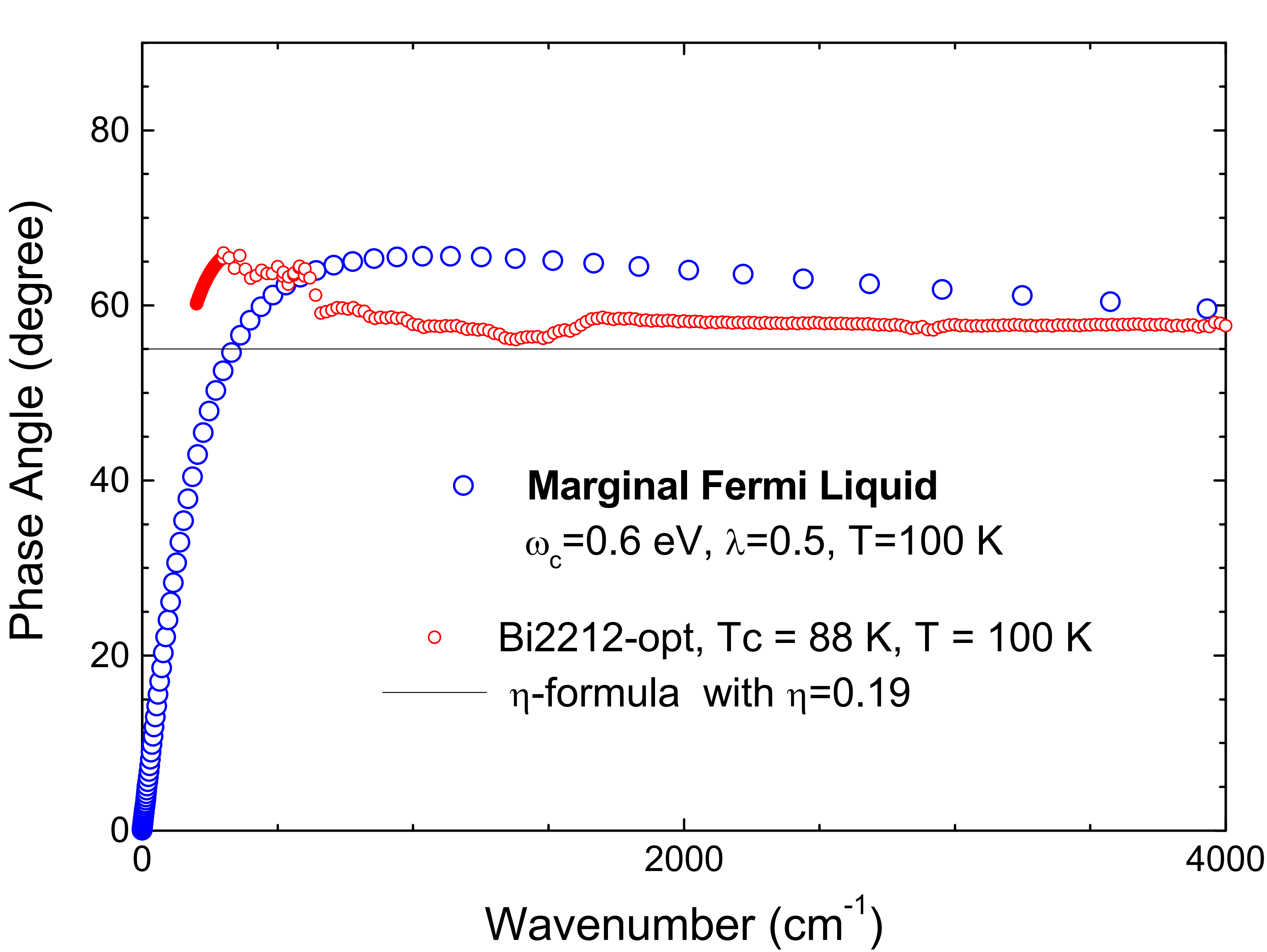
Together: $\phi=0$

$$\sigma(\omega) = |C|\, (-i\, \omega)^{2\eta-1}$$

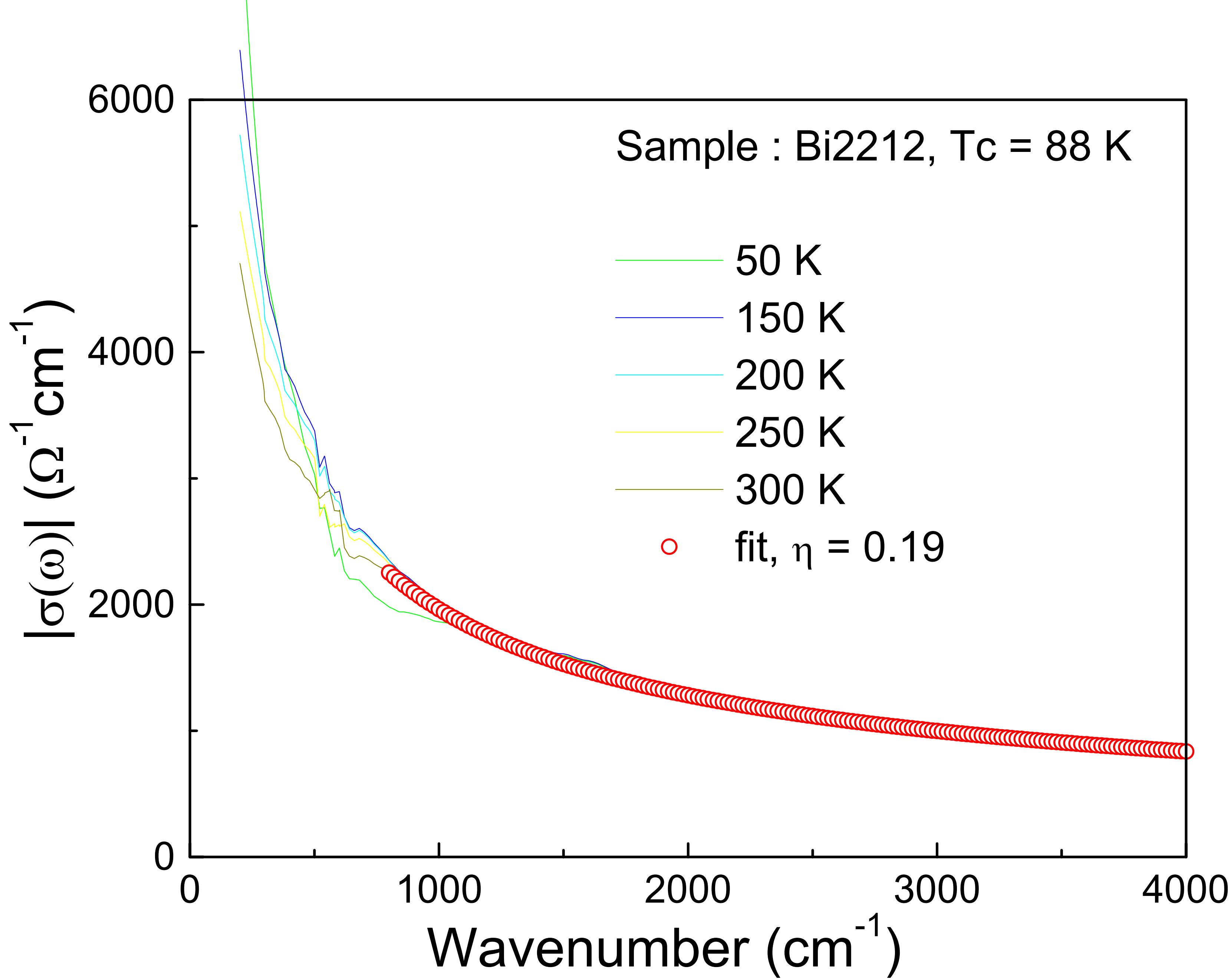
Originally derived in the context of spin-charge separation
by P.W. Anderson, PRB55, 11785 (1997)

$$\phi_\sigma = \arctan\, (\sigma_2 / \sigma_1) = \pi / 2 - \pi\, \eta$$

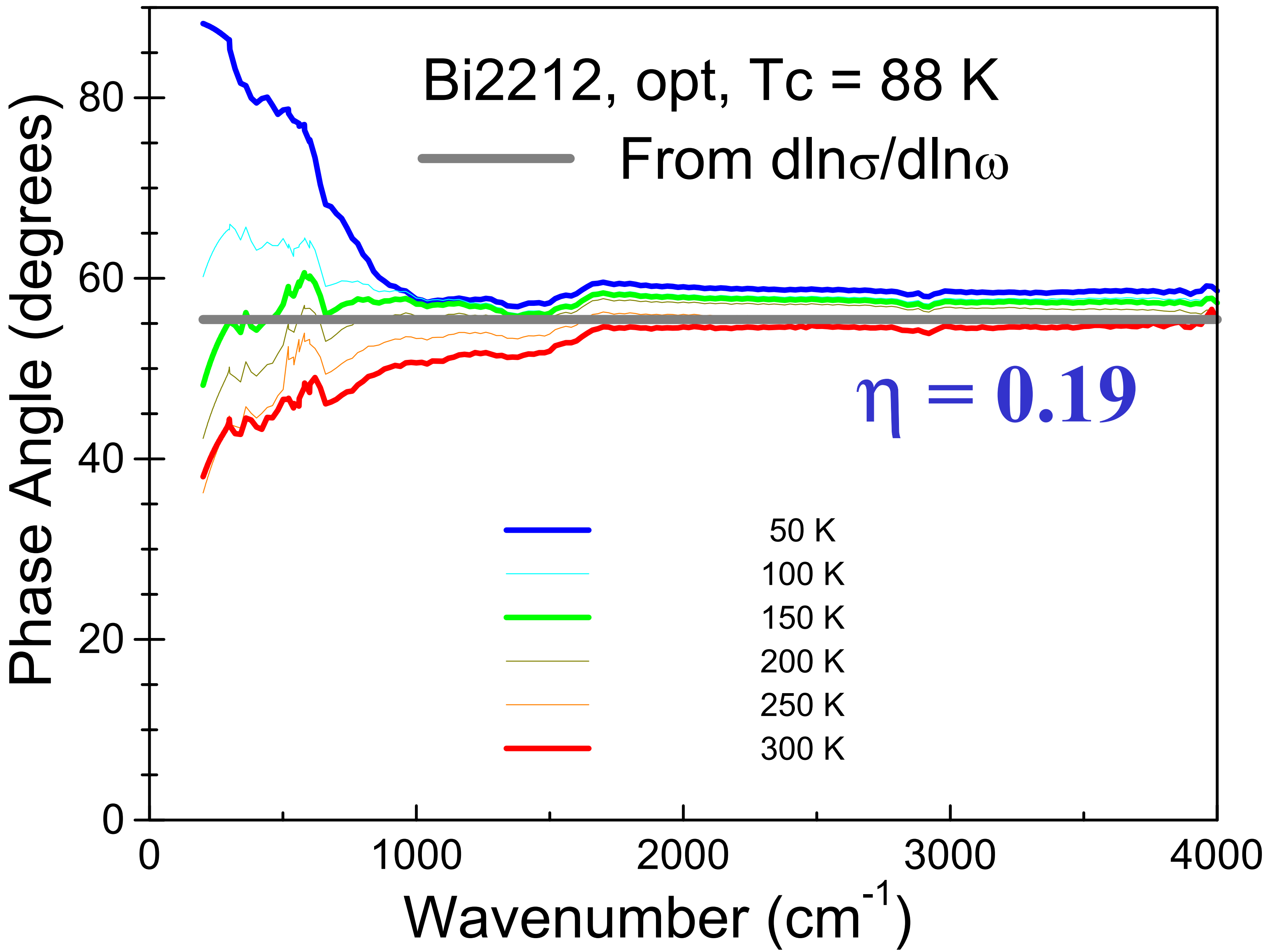
$$d \ln|\sigma| / d \ln \omega = 2\, \eta - 1$$



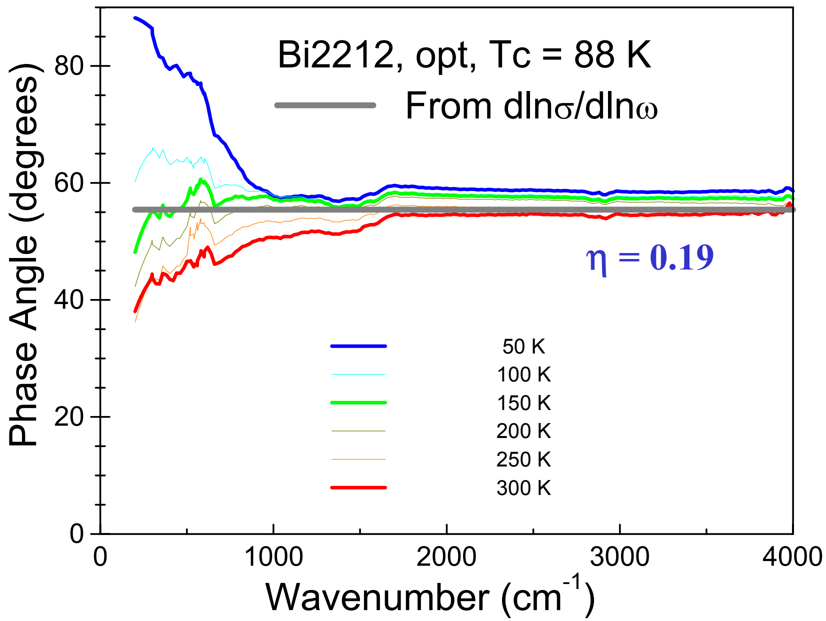
Phase Angle (degree)
0
20
40
60
80
0
2000
4000
Wavenumber (cm-1)
Marginal Fermi Liquid
ωc=0.6 eV, λ=0.5, T=100 K
Bi2212-opt, Tc = 88 K, T = 100 K
η-formula with η=0.19

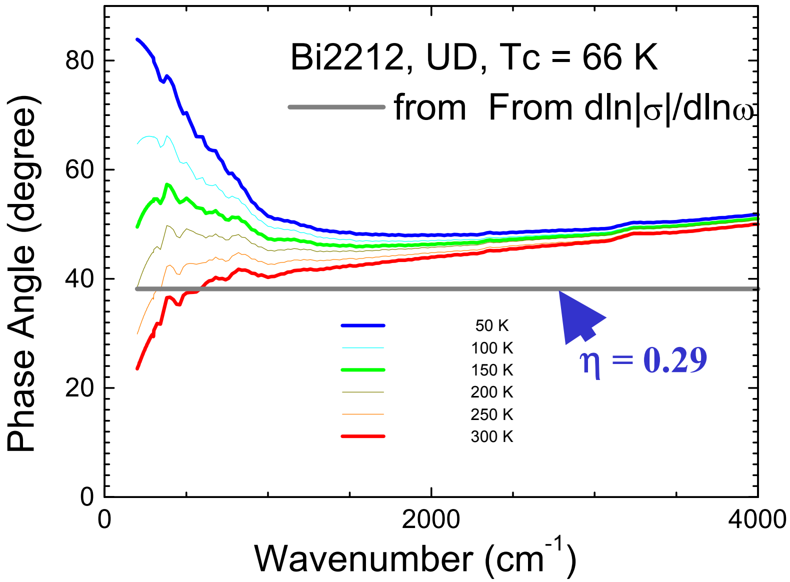


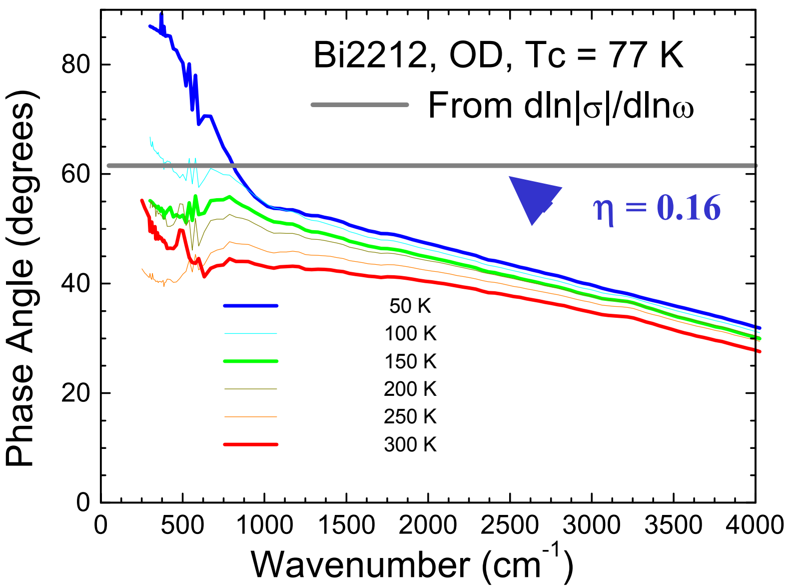
Sample : Bi2212, Tc = 88 K
50 K
150 K
200 K
250 K
300 K
fit, η = 0.19
|σ(ω)| (Ω⁻¹cm⁻¹)
Wavenumber (cm⁻¹)
0
2000
4000
6000
0
1000
2000
3000
4000

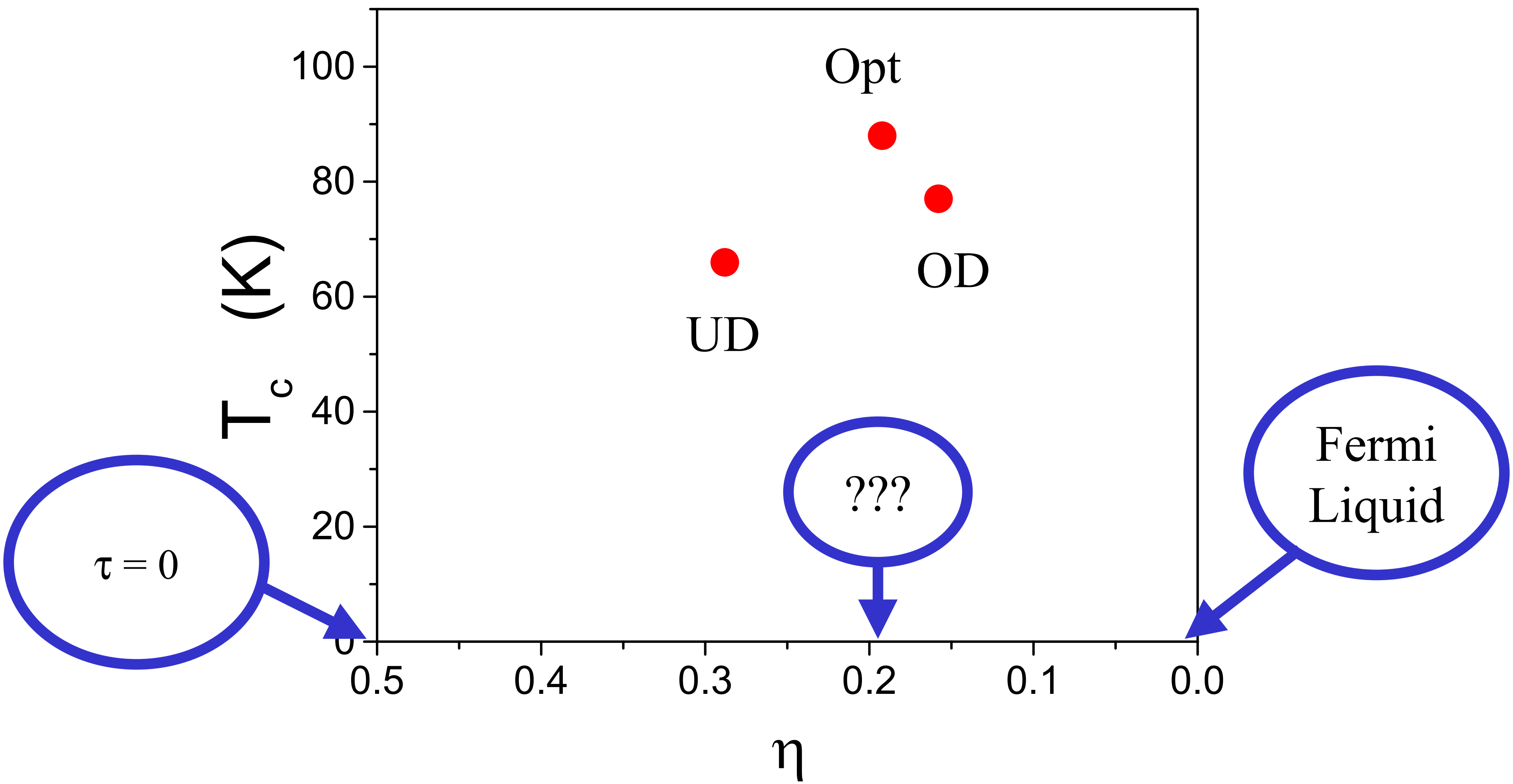


Bi2212, opt, Tc = 88 K
From dlnσ/dlnω
η = 0.19
50 K
100 K
150 K
200 K
250 K
300 K
Phase Angle (degrees)
Wavenumber (cm-1)
0
20
40
60
80
0
1000
2000
3000
4000

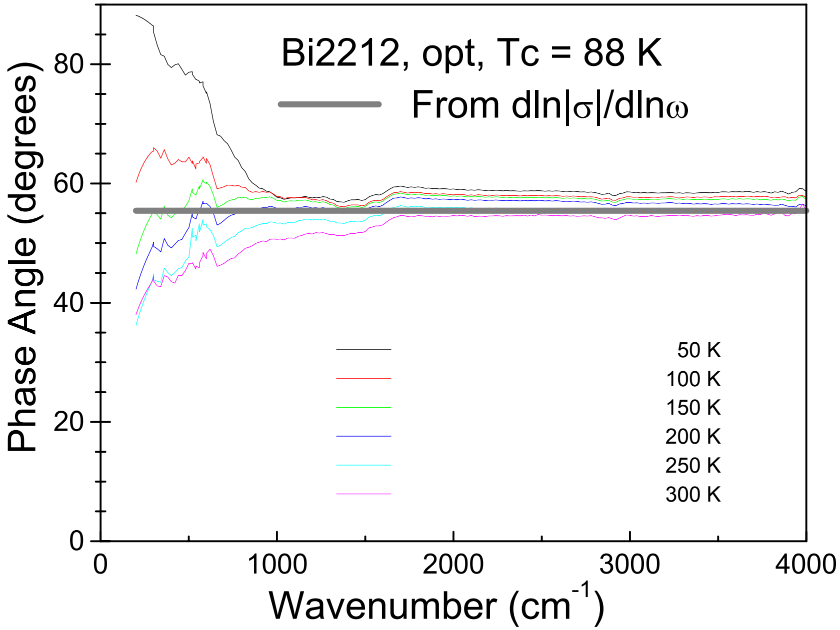

Bi2212, opt, Tc = 88 K
From dlnσ/dlnω
η = 0.19
50 K
100 K
150 K
200 K
250 K
300 K
Phase Angle (degrees)
Wavenumber (cm-1)

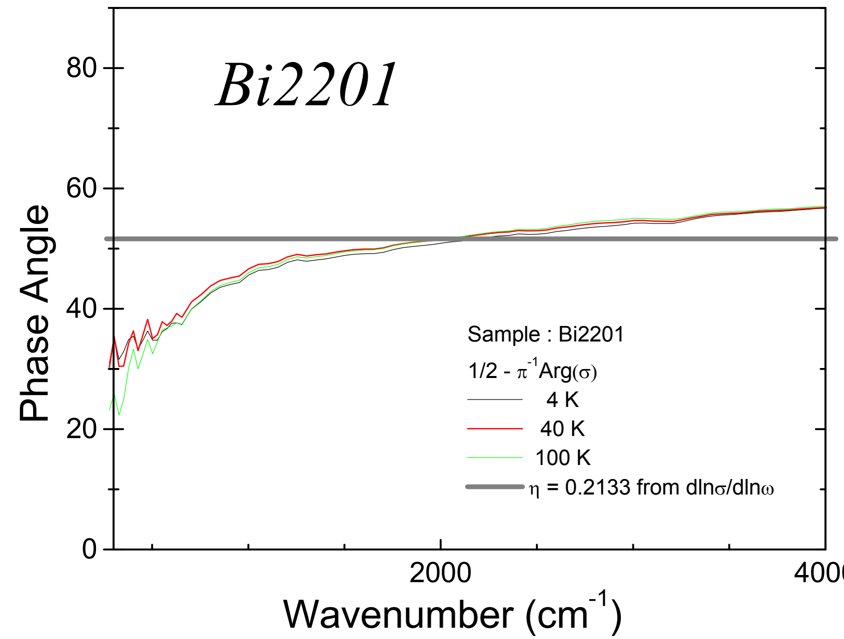

Bi2212, UD, Tc = 66 K
from From dln|σ|/dlnω
η = 0.29
50 K
100 K
150 K
200 K
250 K
300 K
Phase Angle (degree)
Wavenumber (cm-1)

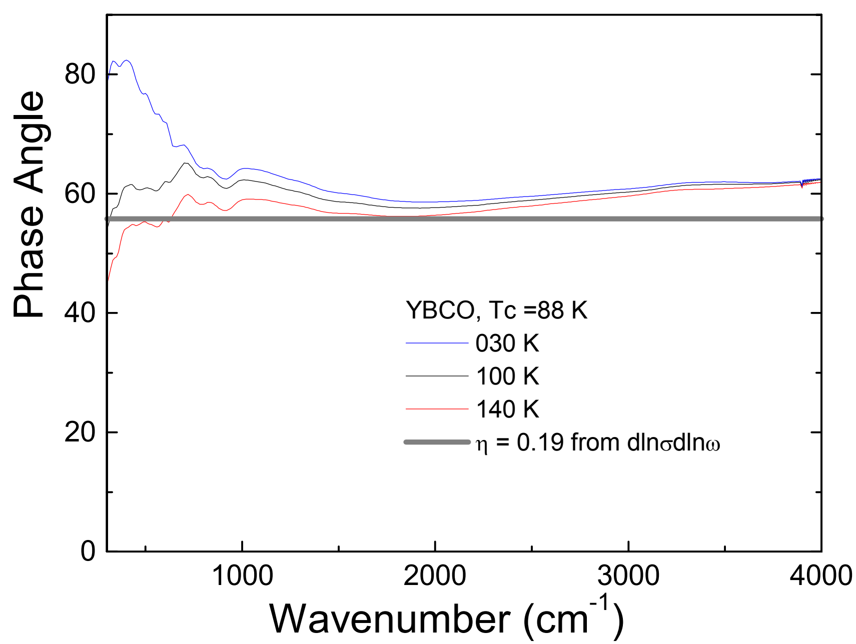

Bi2212, OD, Tc = 77 K
From dln|σ|/dlnω
η = 0.16
50 K
100 K
150 K
200 K
250 K
300 K
Phase Angle (degrees)
Wavenumber (cm-1)



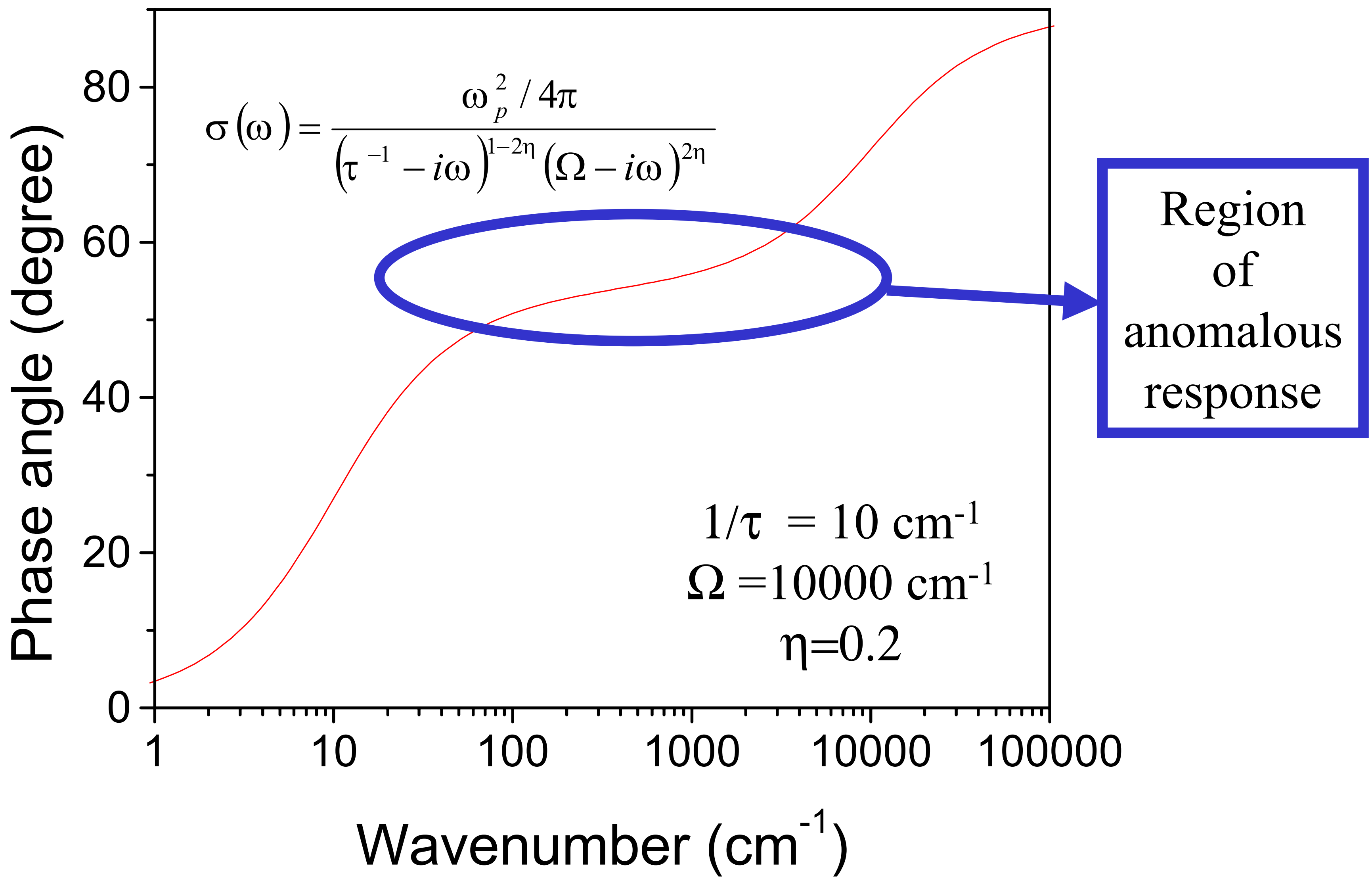
Opt
OD
UD
T_c (K)
100
80
60
40
20
0
0.5
0.4
0.3
0.2
0.1
0.0
η
τ = 0
???
Fermi Liquid


Bi2212, opt, Tc = 88 K
From dln|σ|/dlnω
50 K
100 K
150 K
200 K
250 K
300 K
Phase Angle (degrees)
Wavenumber (cm-1)
0
20
40
60
80
0
1000
2000
3000
4000

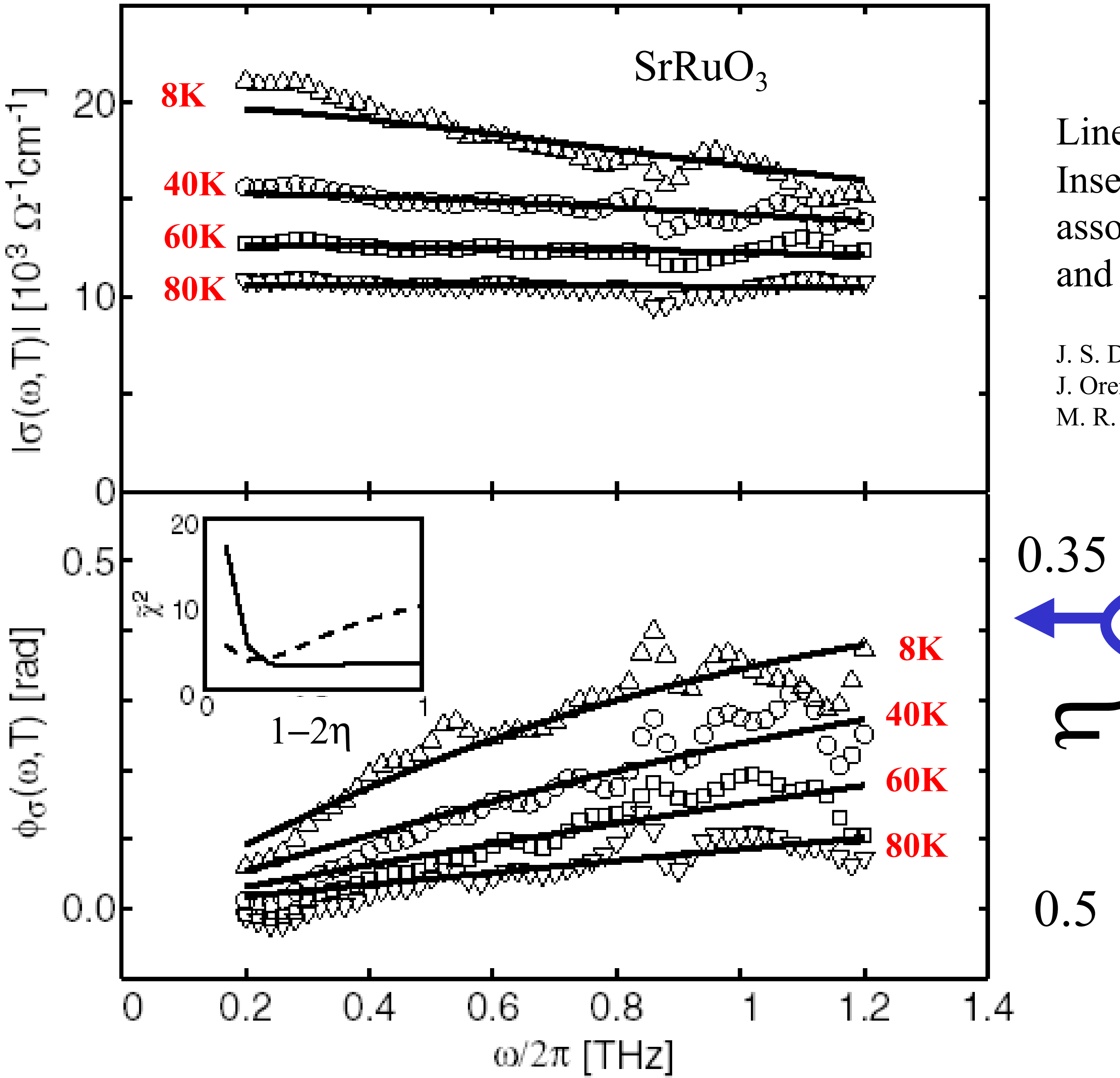

Bi2201
Sample : Bi2201
1/2 - π-1Arg(σ)
4 K
40 K
100 K
η = 0.2133 from dlnσ/dlnω
Phase Angle
Wavenumber (cm-1)
0
20
40
60
80
2000
4000


YBCO, Tc =88 K
030 K
100 K
140 K
η = 0.19 from dlnσdlnω
Phase Angle
Wavenumber (cm-1)
0
20
40
60
80
1000
2000
3000
4000

Y123



$\sigma(\omega) = \frac{\omega_p^2 / 4\pi}{(\tau^{-1} - i\omega)^{1-2\eta} (\Omega - i\omega)^{2\eta}}$
Region of anomalous response
$1/\tau = 10\ cm^{-1}$
$\Omega = 10000\ cm^{-1}$
$\eta = 0.2$
Phase angle (degree)
0
20
40
60
80
1
10
100
1000
10000
100000
Wavenumber (cm^{-1})



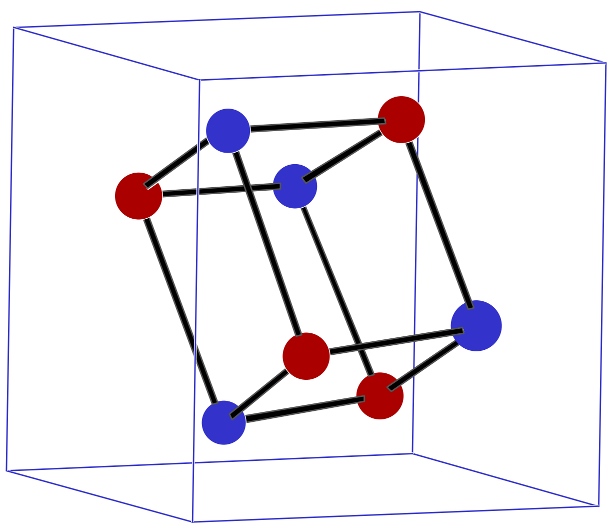
Lines: fits with **η= 0.3**
Inset: reduced χ^2 error associated with phase (solid) and amplitude (dashed)

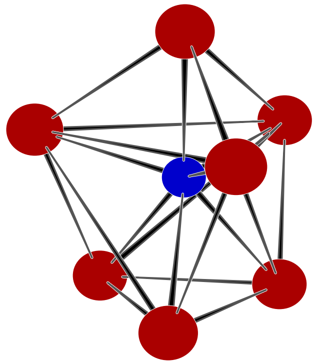
J. S. Dodge, C. P. Weber, J. Corson, J. Orenstein, Z. Schlesinger, J.W. Reiner, M. R. Beasley, PRL 85, 4932 (2000)

Transition metal mono-sillicides

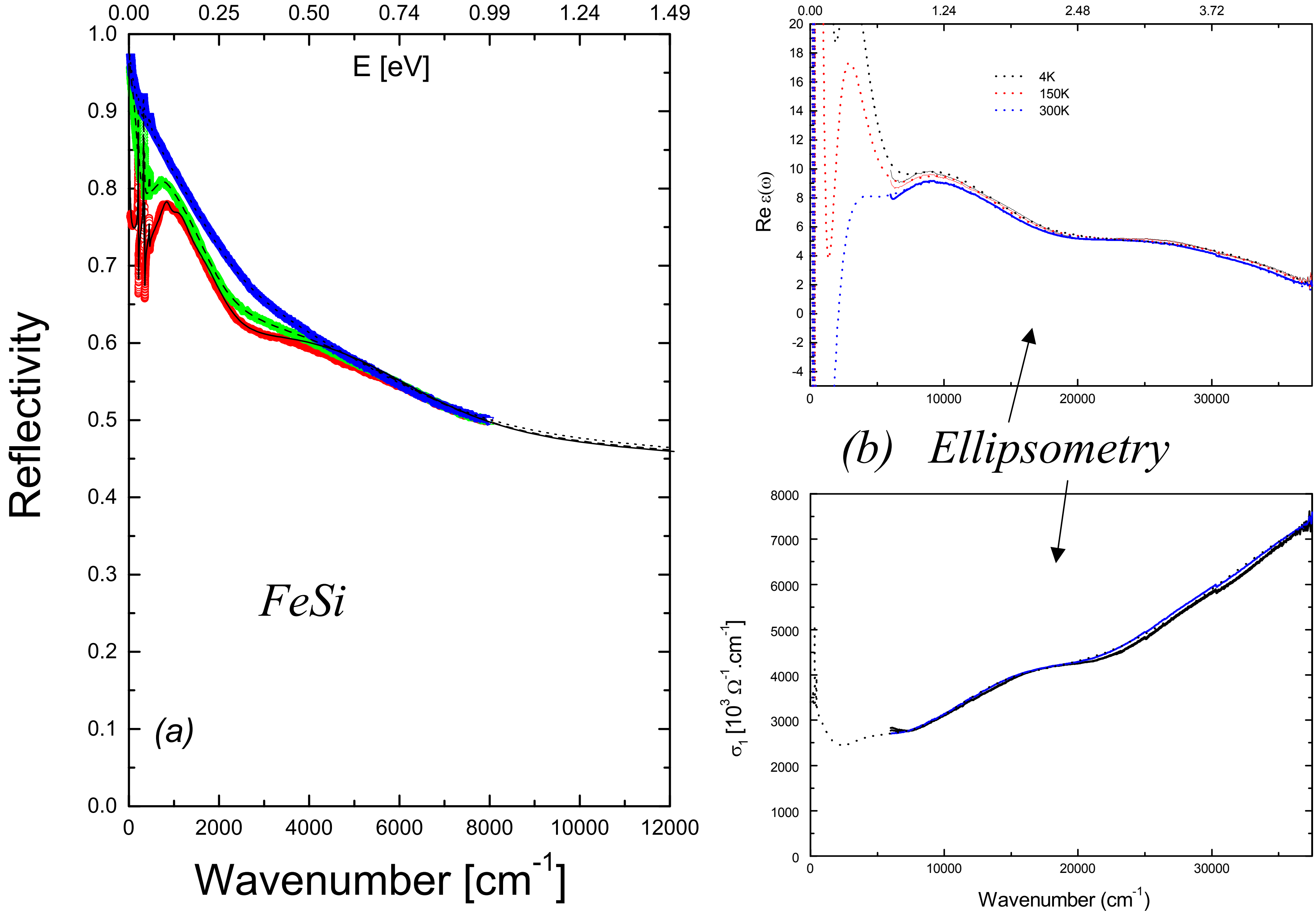
Sc ?	Ti	V	Cr	Mn	Fe	Co	Ni + NiAs
Y CrB	Zr FeB	Nb ?	Mo ?	Tc ?	Ru	Rh	Pd
La ?	Hf ?	Ta ?	W ?	Re	Os	Ir	Pt

- *for TM = Cr Mn Fe Co Ni Ru Rh Re Os:*
- Simple Cubic Space Group (T^4-$P2_13$)
- 4 equivalent TM and Si atoms



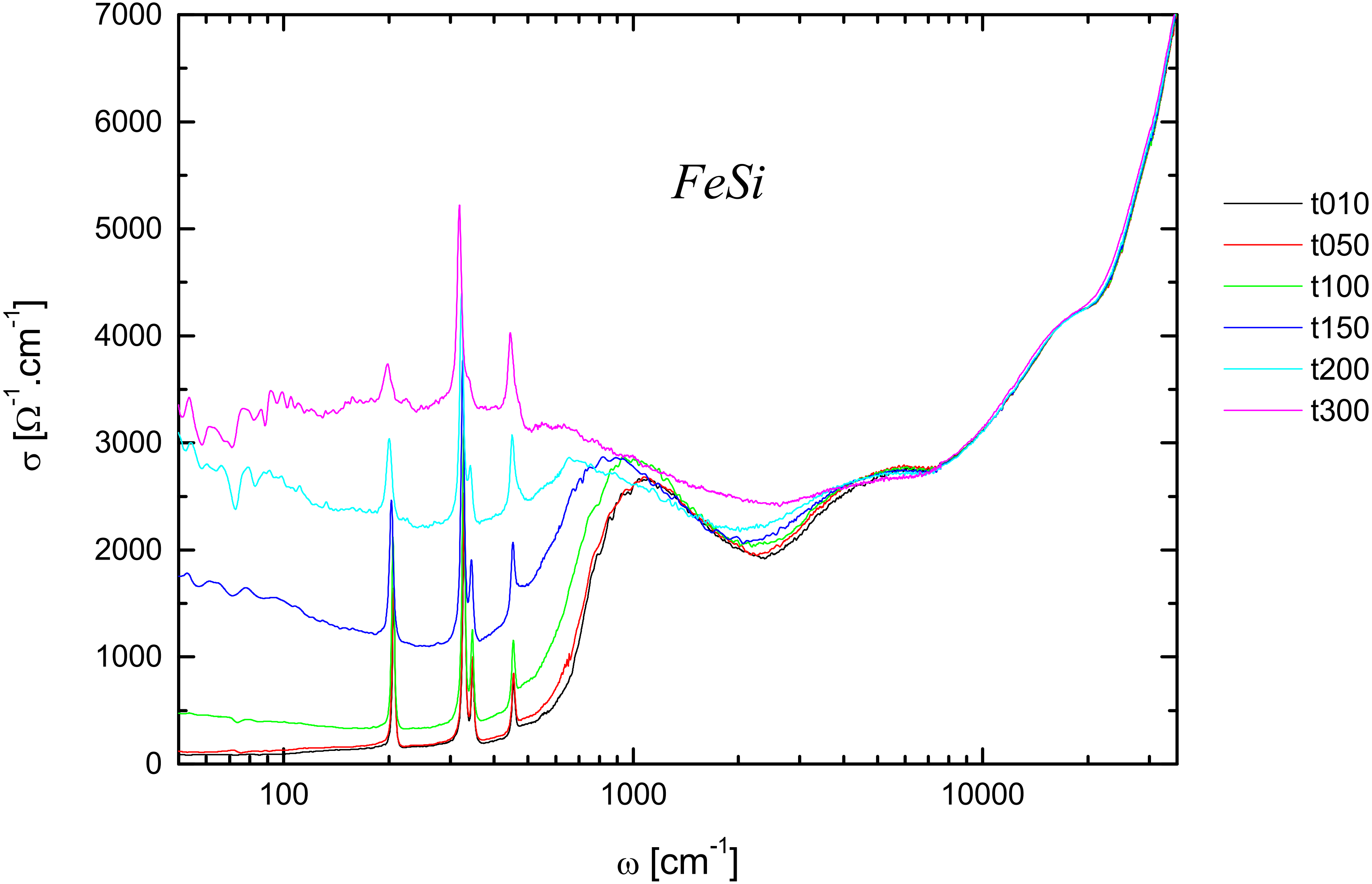


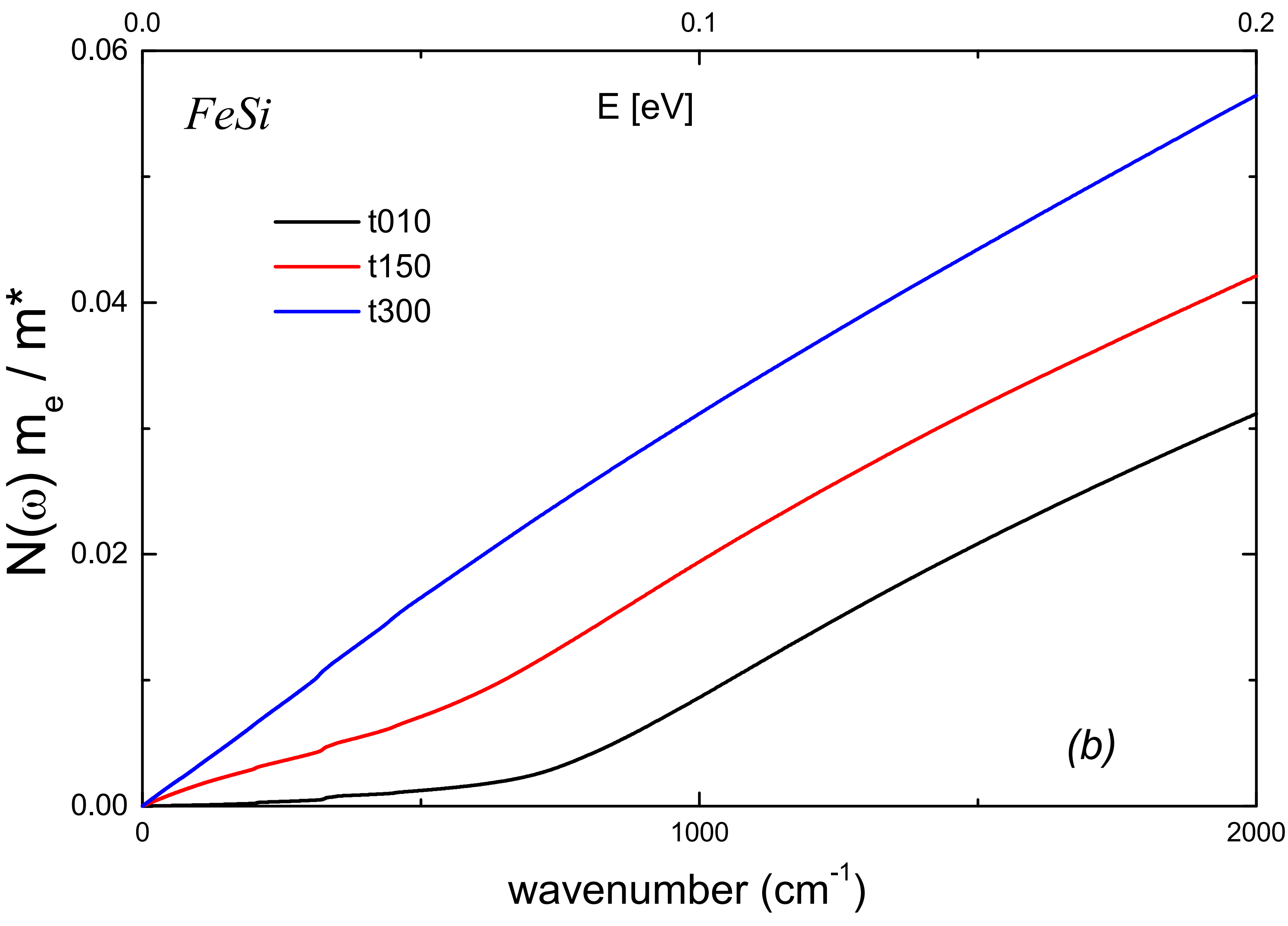
- Site symmetry: C_3
- 7-fold coordination of Tm-atoms

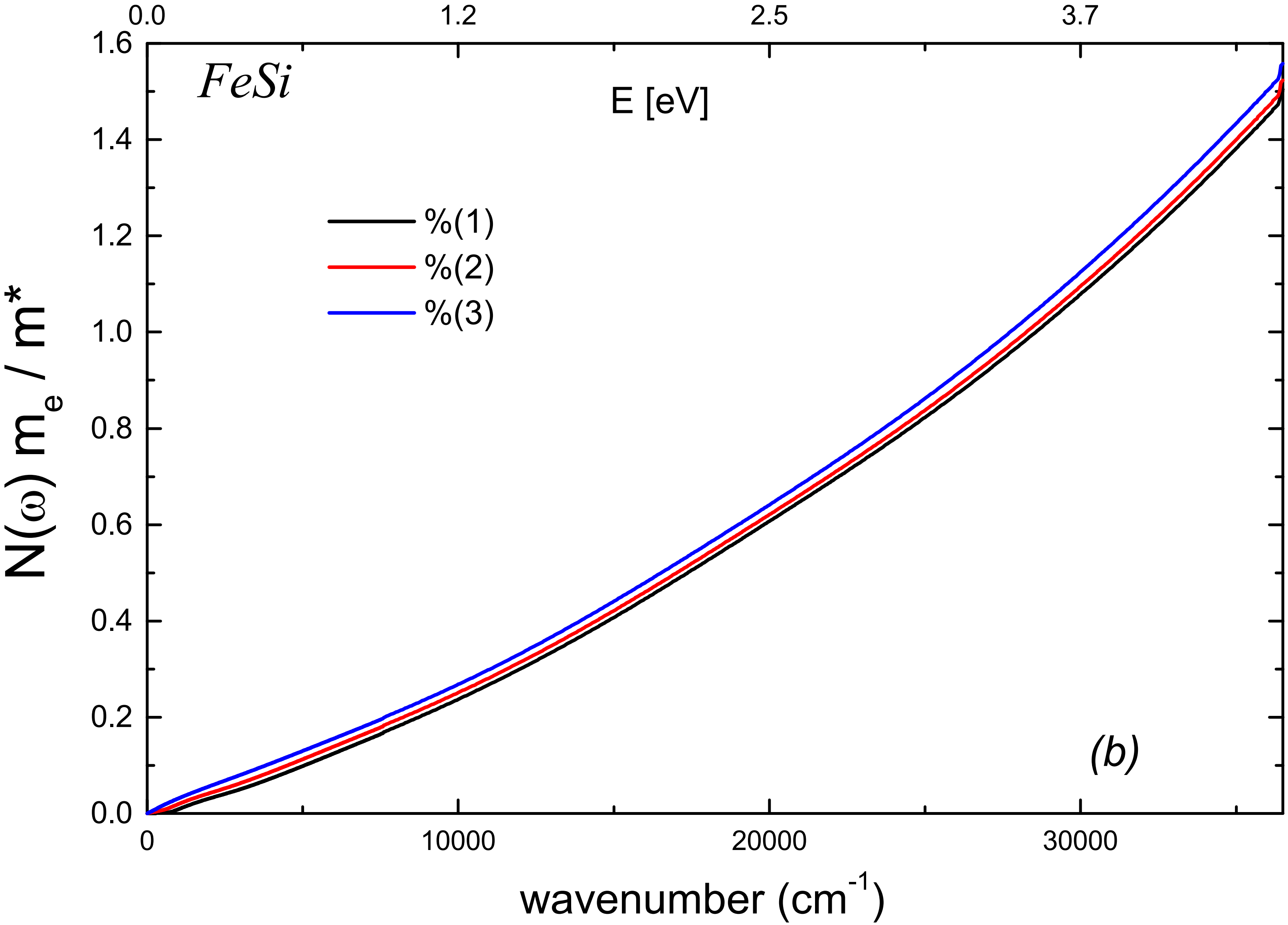


E [eV]
0.00
0.25
0.50
0.74
0.99
1.24
1.49
Reflectivity
Wavenumber [cm^{-1}]
FeSi
(a)
(b) Ellipsometry
Re ε(ω)
4K
150K
300K
2.48
3.72
σ$_1$ [10^3 Ω^{-1}.cm^{-1}]
Wavenumber (cm^{-1})

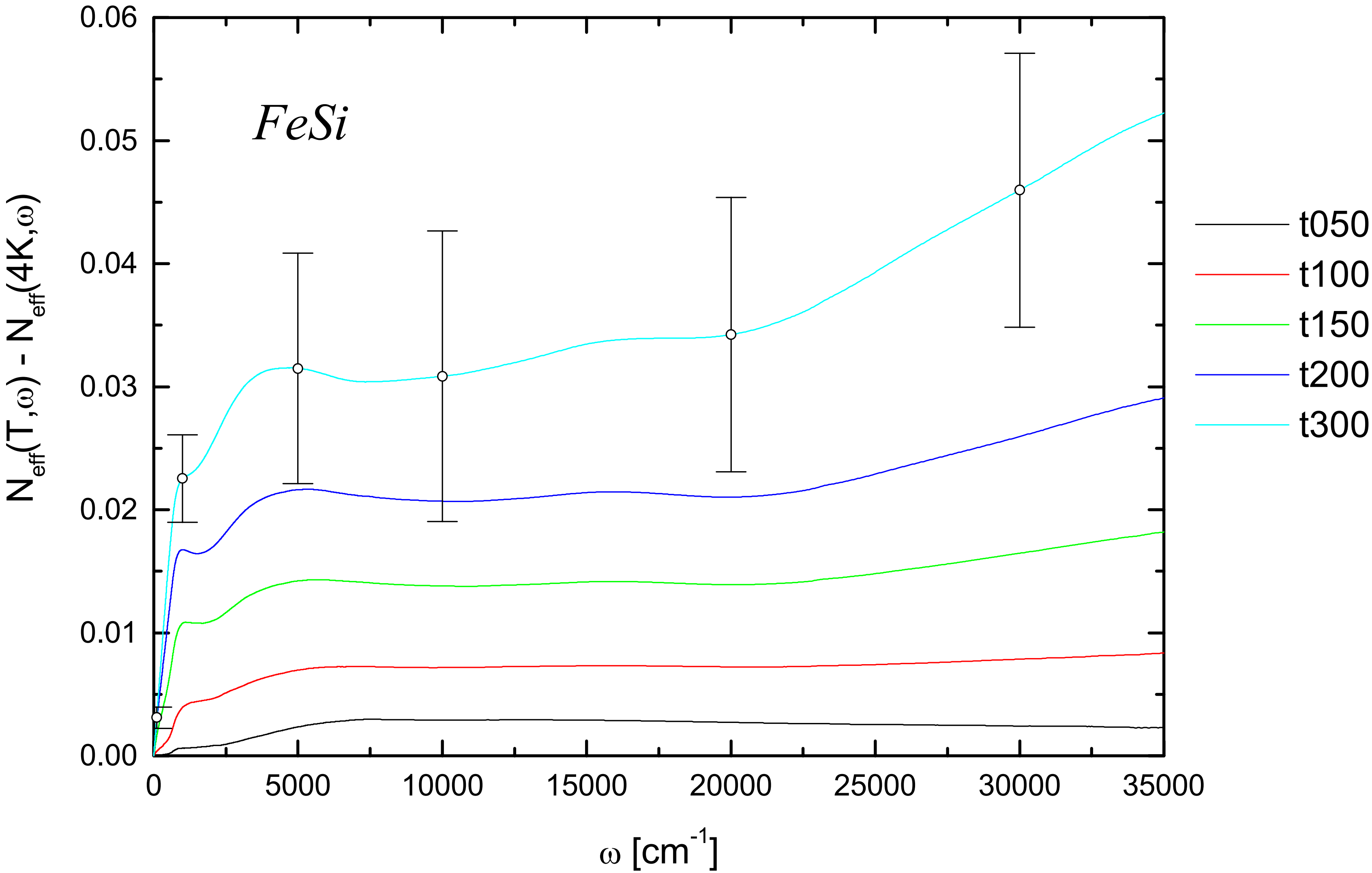
FeSi (Andrea's data + vis. ellipsometer)




0.0
0.1
0.2
E [eV]
FeSi
t010
t150
t300
(b)
0.06
0.04
0.02
0.00
$N(\omega)\, m_e / m^*$
0
1000
2000
wavenumber (cm^{-1})



FeSi
E [eV]
0.0
1.2
2.5
3.7
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
%(1)
%(2)
%(3)
(b)
N(ω) m_e / m*
0
10000
20000
30000
wavenumber (cm^-1)



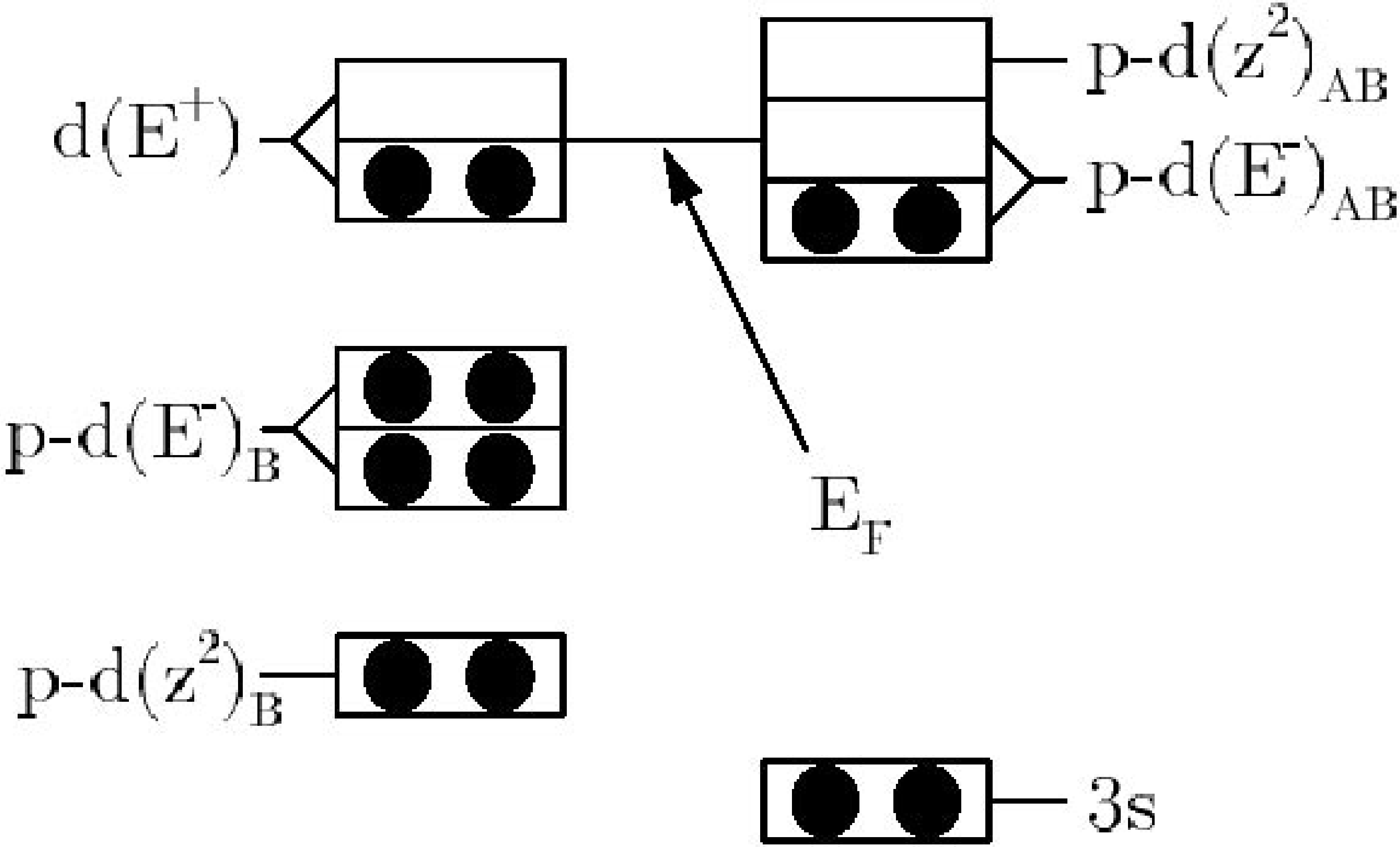
FeSi
0.06
0.05
0.04
0.03
0.02
0.01
0.00
0
5000
10000
15000
20000
25000
30000
35000
$N_{eff}(T,\omega) - N_{eff}(4K,\omega)$
ω [cm^{-1}]
t050
t100
t150
t200
t300

•*Z. Schlesinger, Fisk, Zhang, Maple, DiTusa, Aeppli, PRL'93:*
*"**Sum-rule not recovered up to 1 eV**"*
(Reflectivity&Kramers-Kronig)

•*Degiorgi, Hunt,Ott,Dressel,Feenstra, Gruener, Fisk, Canfield, EPL'94:*
"Sum-rule recovered at 0.4 eV"
(Reflectivity&Kramers-Kronig)

•*Damascelli, PhD-thesis'98:*
"Sum-rule not recovered up to 1.25 eV"
(Reflectivity&Kramers-Kronig)

•*F.P. Mena, this work:*
"Sum-rule not recovered up to 4.5 eV"
(VIS-Ellipsometry & IR-Reflectivity&Kramers-Kronig)



d(E+)
p-d(z2)AB
p-d(E-)AB
p-d(E-)B
EF
p-d(z2)B
3s

Formation of S = 0 state out of itinerant band electrons and localized 3d - spins

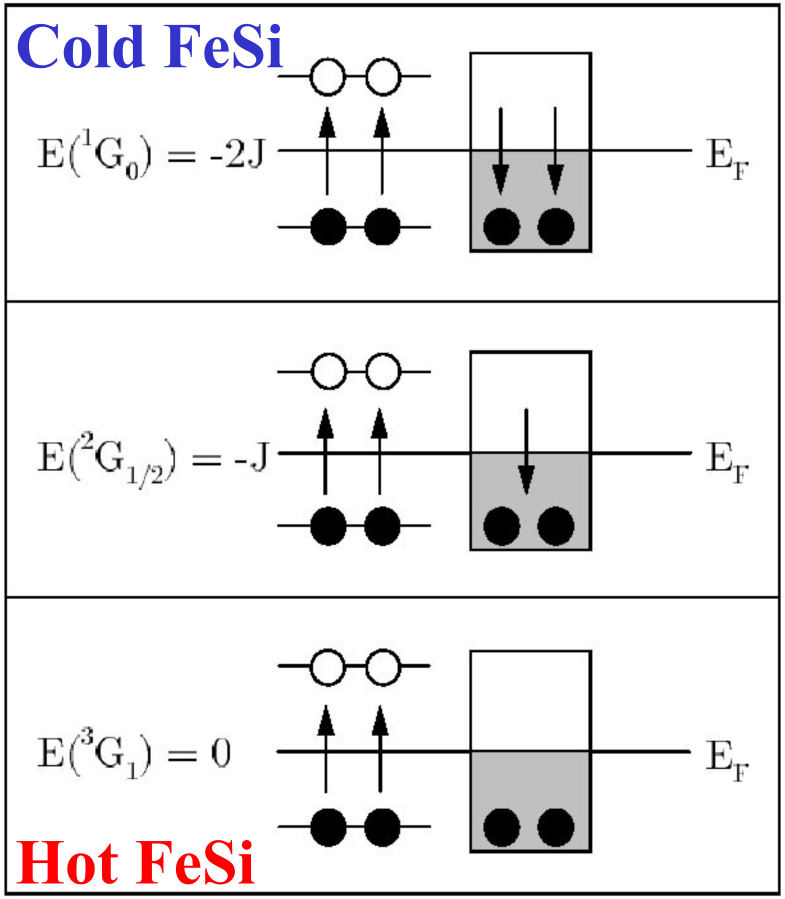
$\Rightarrow$ Increased presence of itinerant band electrons in local 3d states

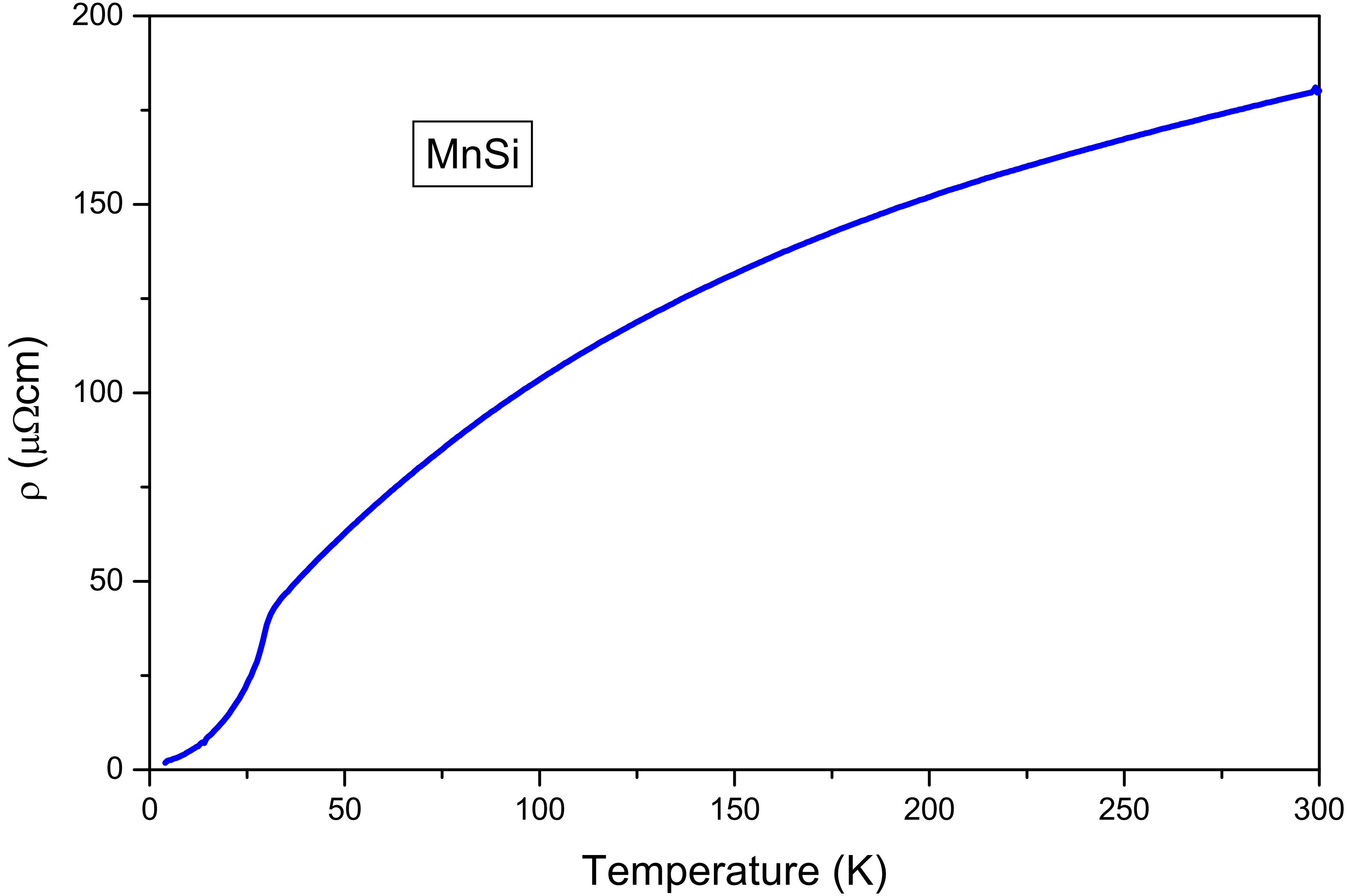
$\Rightarrow$ Localizati on of charge carriers and opening of a charge gap

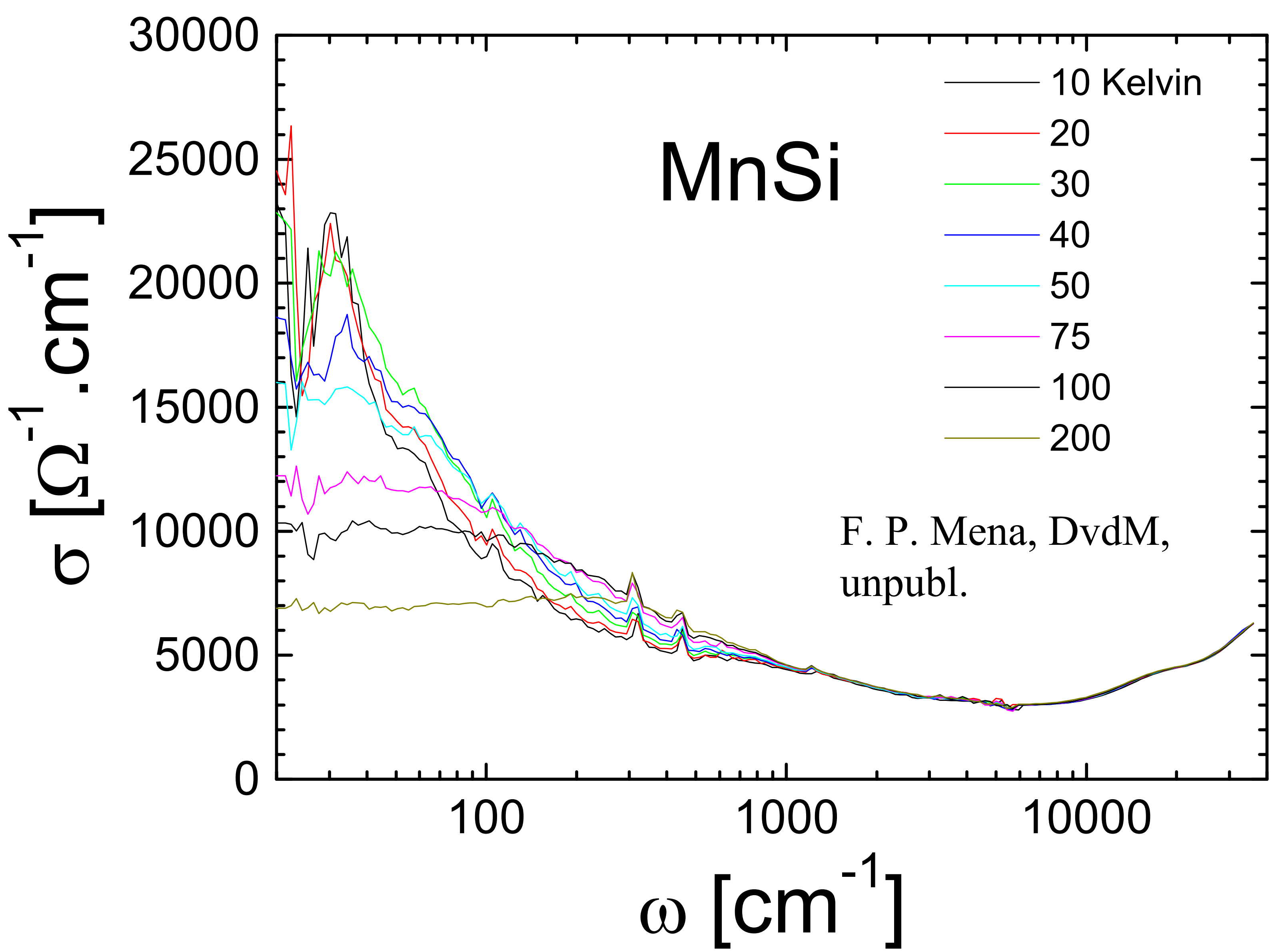
$\Rightarrow$ *In*crease of charge kinetic energy

$\Rightarrow$ D*e*crease of Drude spectral weight

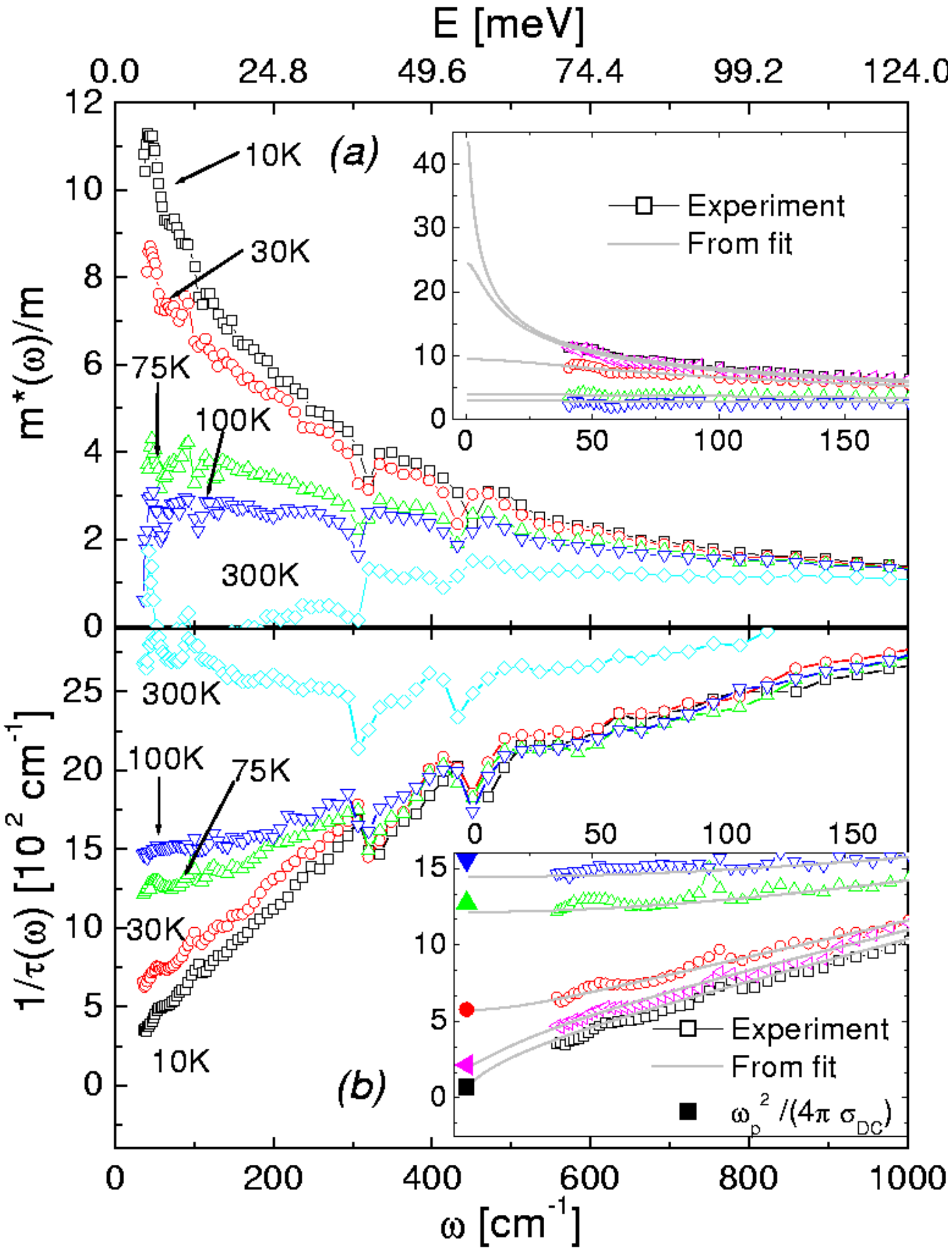
$\Rightarrow$ *In*creased 3d - 4f spectral weight



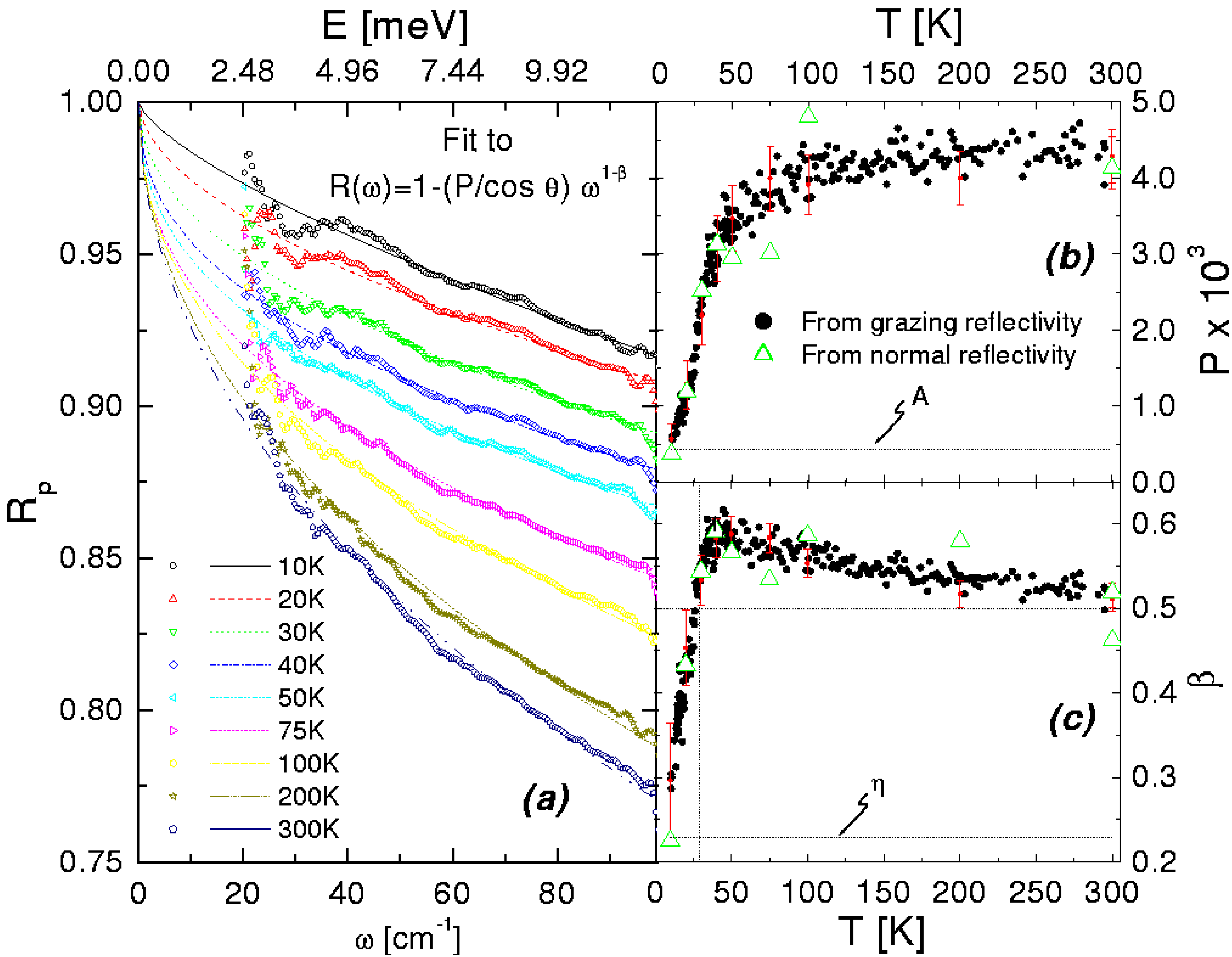

MnSi
ρ (μΩcm)
Temperature (K)
0
50
100
150
200
0
50
100
150
200
250
300



30000
25000
20000
15000
10000
5000
0
σ [Ω-1.cm-1]
100
1000
10000
ω [cm-1]
MnSi
10 Kelvin
20
30
40
50
75
100
200
F. P. Mena, DvdM,
unpubl.



E [meV]
0.0
24.8
49.6
74.4
99.2
124.0
m*(ω)/m
10K
30K
75K
100K
300K
(a)
Experiment
From fit
1/τ(ω) [10² cm⁻¹]
300K
100K
75K
30K
10K
(b)
Experiment
From fit
ωp²/(4π σDC)
ω [cm⁻¹]


E [meV]
0.00
2.48
4.96
7.44
9.92
T [K]
Fit to
R(ω)=1-(P/cos θ) ω^1-β
R_p
10K
20K
30K
40K
50K
75K
100K
200K
300K
(a)
ω [cm^-1]
(b)
From grazing reflectivity
From normal reflectivity
A
P x 10^3
(c)
η
β
T [K]

Constraints on σ(ω)

$$\sigma(\omega) = |C| / (-i\,\omega)^{1-2\eta}$$

Thermodynamics	$\mathrm{Re}\,\sigma(\omega) > 0$	
Causality	$\mathrm{Im}\,\sigma(\omega) = \frac{1}{\pi}\int_{-\infty}^{\infty} \frac{\mathrm{Re}\,\sigma(x)}{\omega - x}dx$	
Time reversal symmetry	$\sigma(-\omega) = \sigma^*(\omega)$	
f-sumrule	$\lim_{\omega\to\infty} \sigma(\omega) = i\frac{ne^2}{m\omega}$	✗
DC-limit	$\lim_{\omega\to 0} \sigma(\omega) = \frac{1}{\rho_{DC}(T)}$	✗

$$\sigma(\omega,T)=\frac{\omega_p^2/4\pi}{(\Gamma-i\omega)^{1-2\eta}(\Omega-i\omega)^{2\eta}}$$

Thermodynamics $\qquad \mathrm{Re}\,\sigma(\omega)>0$

Causality $\qquad \mathrm{Im}\,\sigma(\omega)=\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{\mathrm{Re}\,\sigma(x)}{\omega-x}dx$

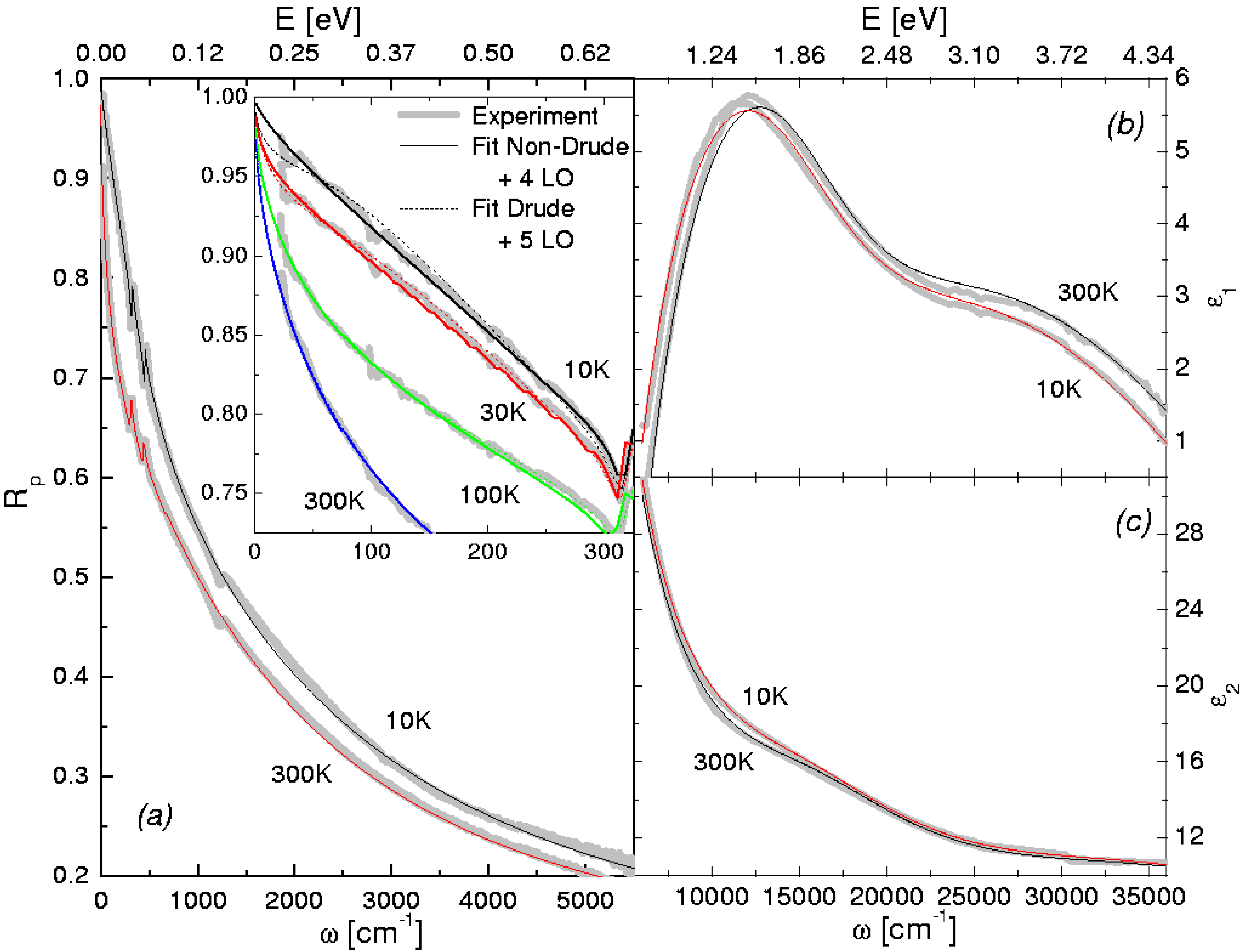
Time reversal symmetry $\qquad \sigma(-\omega)=\sigma^{*}(\omega)$

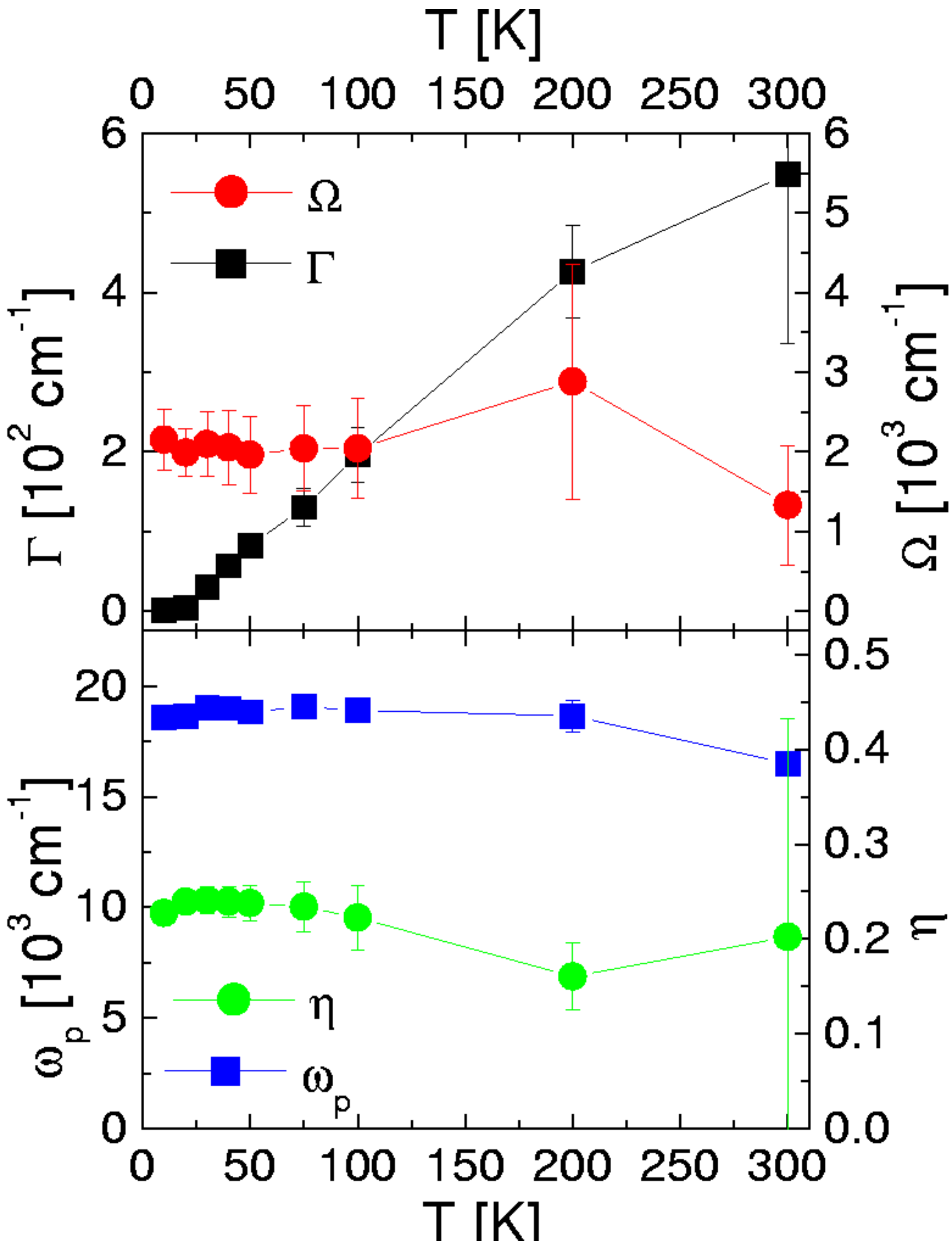
f-sumrule $\qquad \lim_{\omega\to\infty}\sigma(\omega)=i\frac{ne^2}{m\omega}$

DC-limit $\qquad \lim_{\omega\to 0}\sigma(\omega)=\frac{1}{\rho_{DC}(T)}$

DvdM, PRB60, R768 (1999)

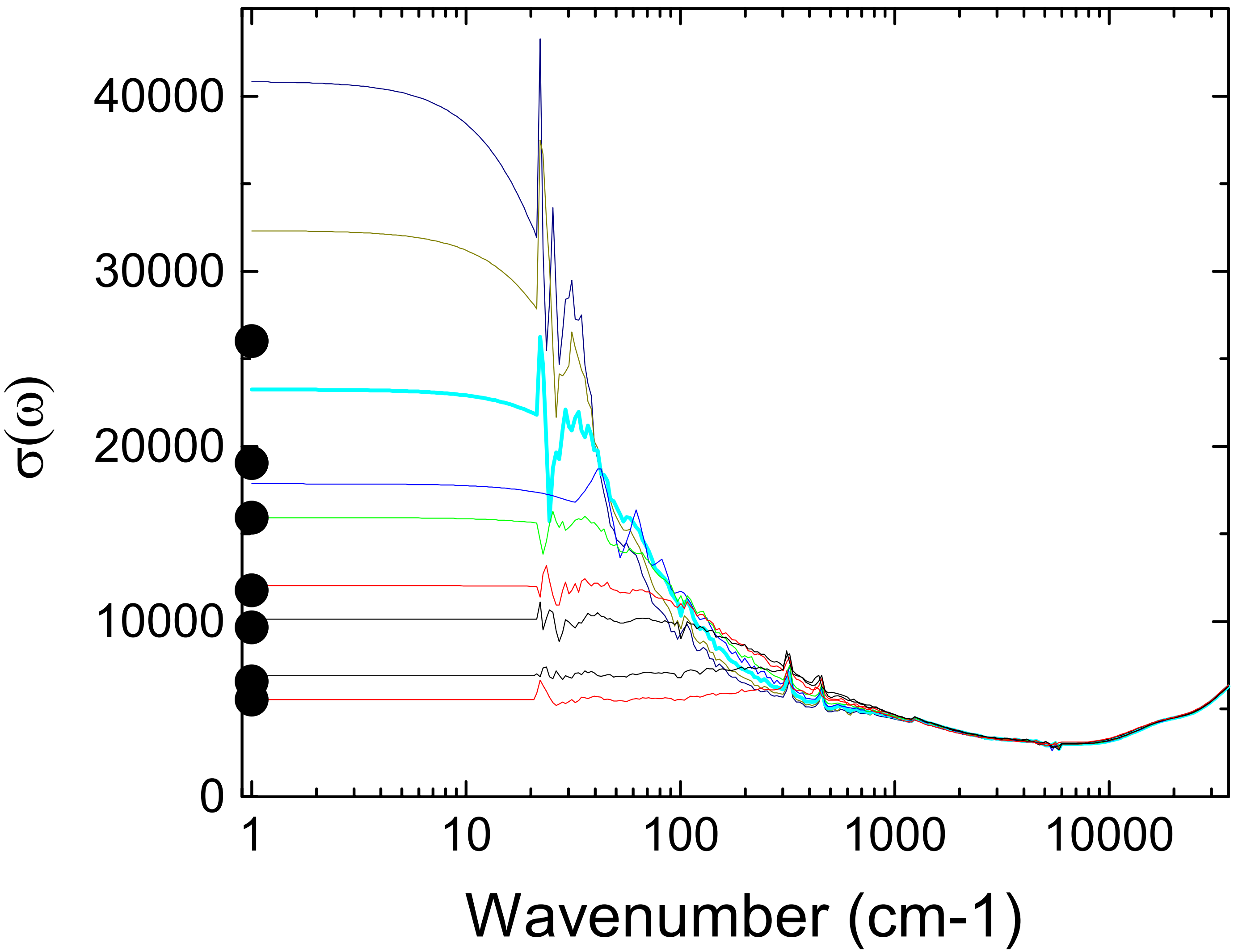
η=0.25: Ioffe&Millis, PRB58 (1998)


E [eV]
0.00
0.12
0.25
0.37
0.50
0.62
Experiment
Fit Non-Drude
+ 4 LO
Fit Drude
+ 5 LO
10K
30K
100K
300K
10K
300K
(a)
R_p
ω [cm^{-1}]
E [eV]
1.24
1.86
2.48
3.10
3.72
4.34
(b)
300K
10K
ε_1
(c)
10K
300K
ε_2
ω [cm^{-1}]

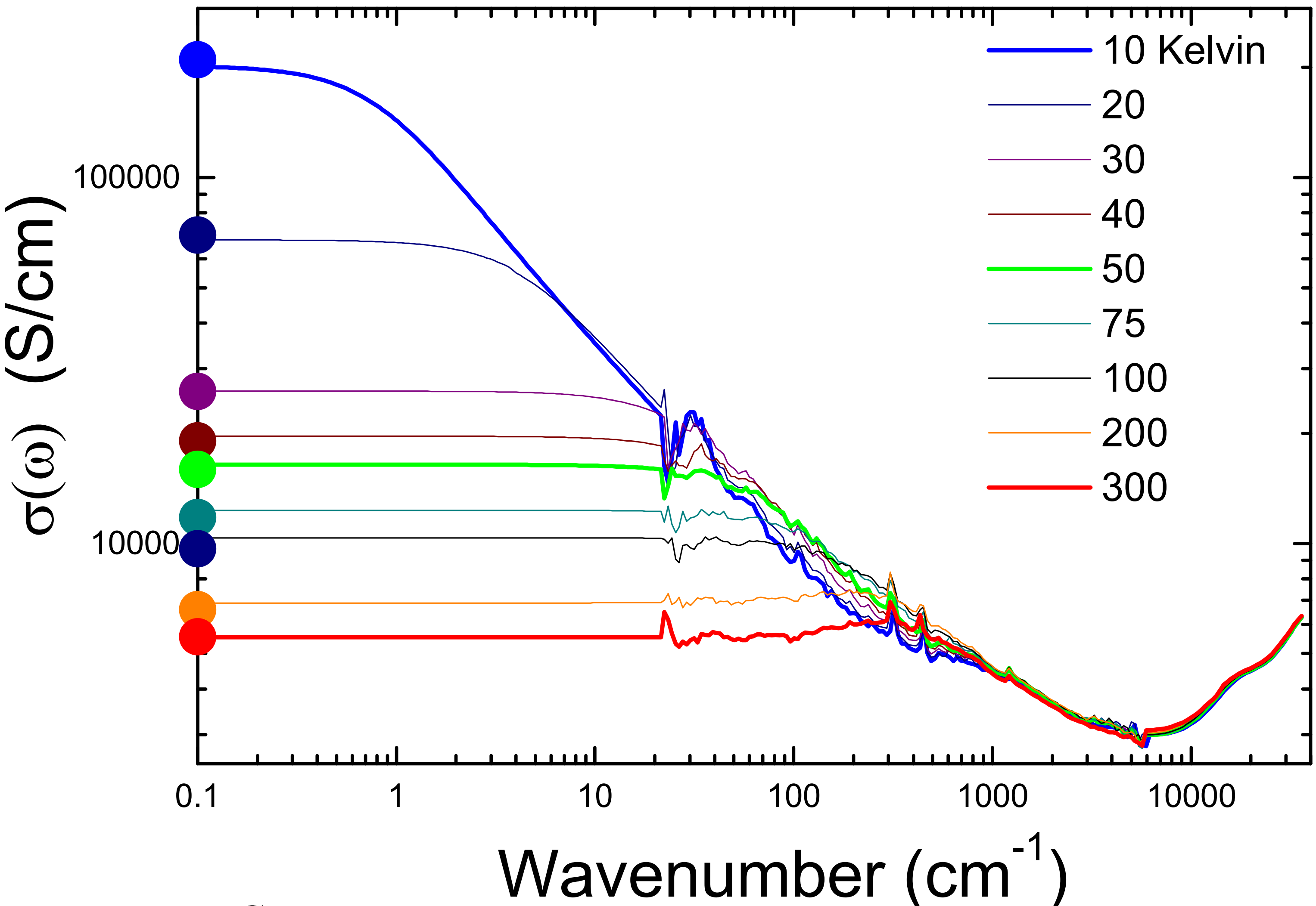


T [K]
0
50
100
150
200
250
300
6
4
2
0
Γ [10² cm⁻¹]
Ω [10³ cm⁻¹]
5
3
1
Ω
Γ
20
15
10
5
0
ω_p [10³ cm⁻¹]
0.5
0.4
0.3
0.2
0.1
0.0
η
η
ω_p
T [K]

Low frequency extrapolation with Drude fit

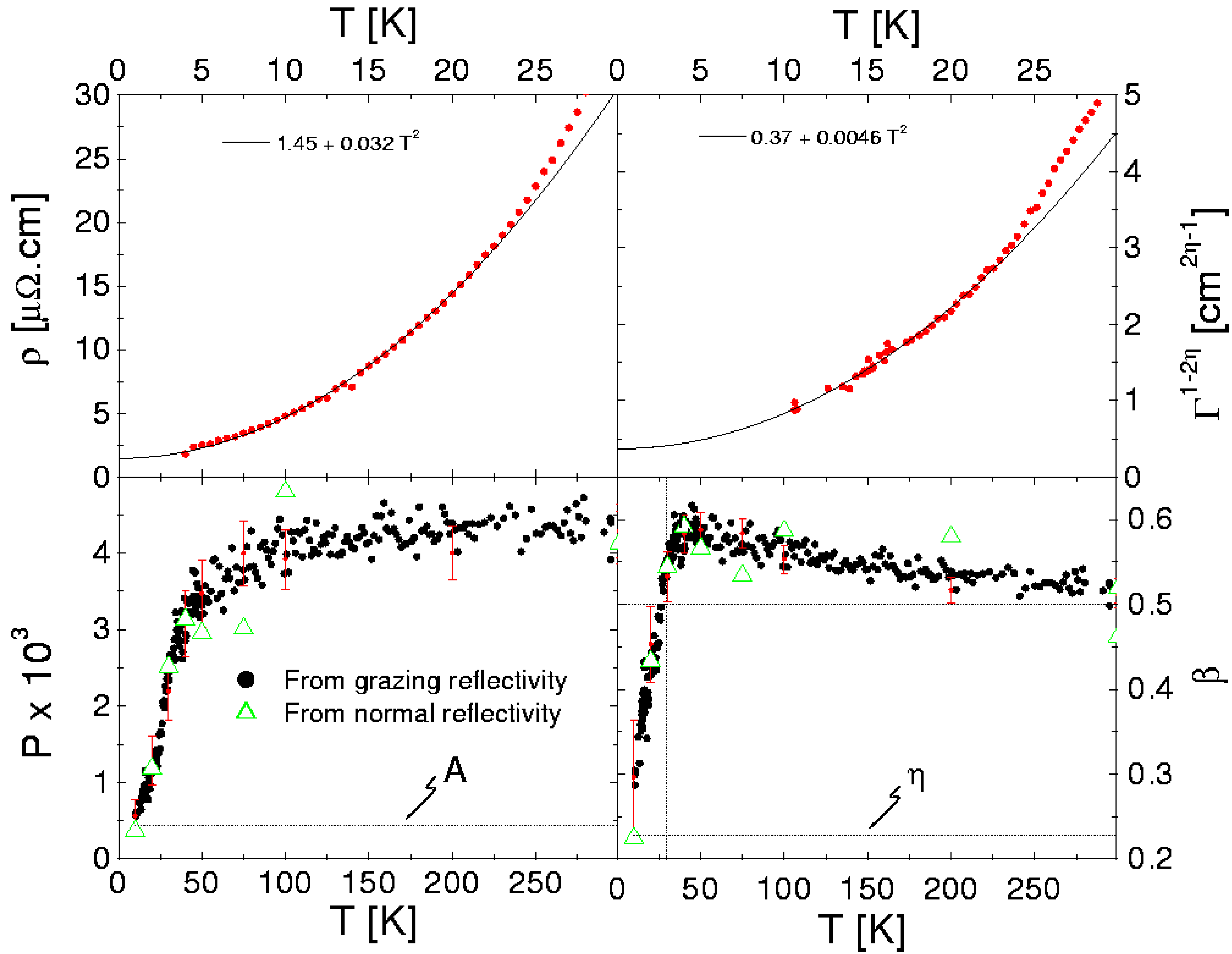


Low frequency extrapolation using fit to η-formula, η=0.25



$$\sigma(\omega) = \frac{C}{(\Gamma - i\omega)^{1-2\eta}}$$

F. P. Mena, DvdM, unpublished



T [K]
T [K]
ρ [μΩ.cm]
1.45 + 0.032 T^2
0.37 + 0.0046 T^2
Γ^{1-2η} [cm^{2η-1}]
P x 10^3
From grazing reflectivity
From normal reflectivity
A
η
β
T [K]
T [K]

Summary

α-Ce is a Fermi-liquid with mass 15
γ-Ce is a bad metal

HTSC at optimal doping

1) **Powerlaw: $\sigma(\omega)$ is proportional to $(i\omega)^{-2.4}$**
2) **Phase of $\sigma(\omega)$ is $0.3\,\pi$ independent of frequency**
3) $\eta = 0.2 \pm 0.01$

FeSi: Gap-spectral weight transferred beyond 4.5 eV

MnSi at low temperatures (helical magnetic phase):

$\rho(T)$ is proportional to T^2
$\sigma(\omega)$ is proportional to $(i\omega)^{-0.5}$