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Innovations in Strongly Correlated Electronic Systems: School and Workshop

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Optical properties of correlated electron systems: Part II

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Optical properties of correlated electron systems

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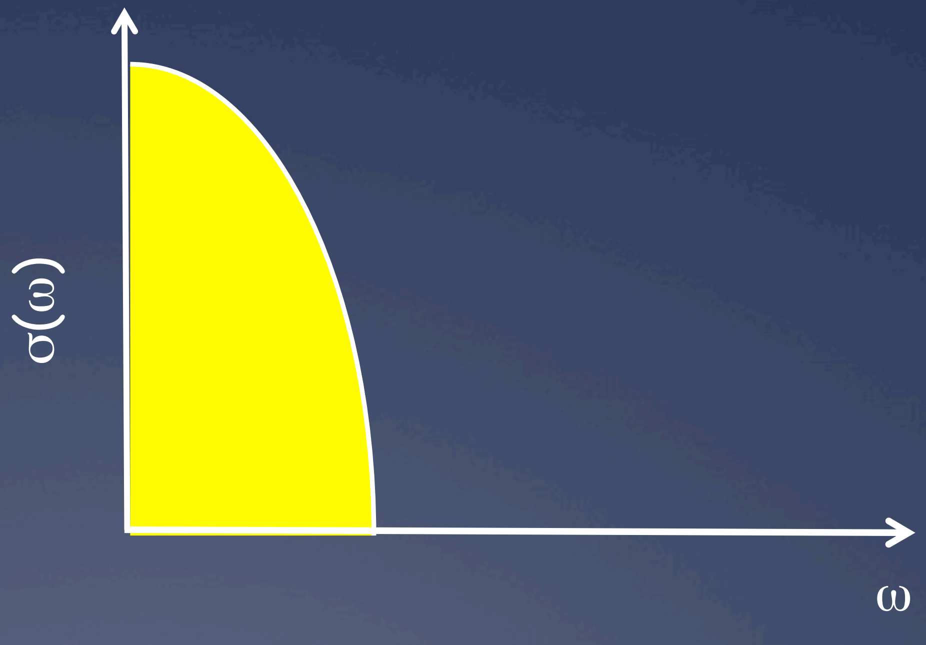
Optical sum rule

$$W = \int_0^\infty d\omega \mathrm{Re}\sigma_{xx}(\omega) = \frac{\pi e^2 n}{m}$$



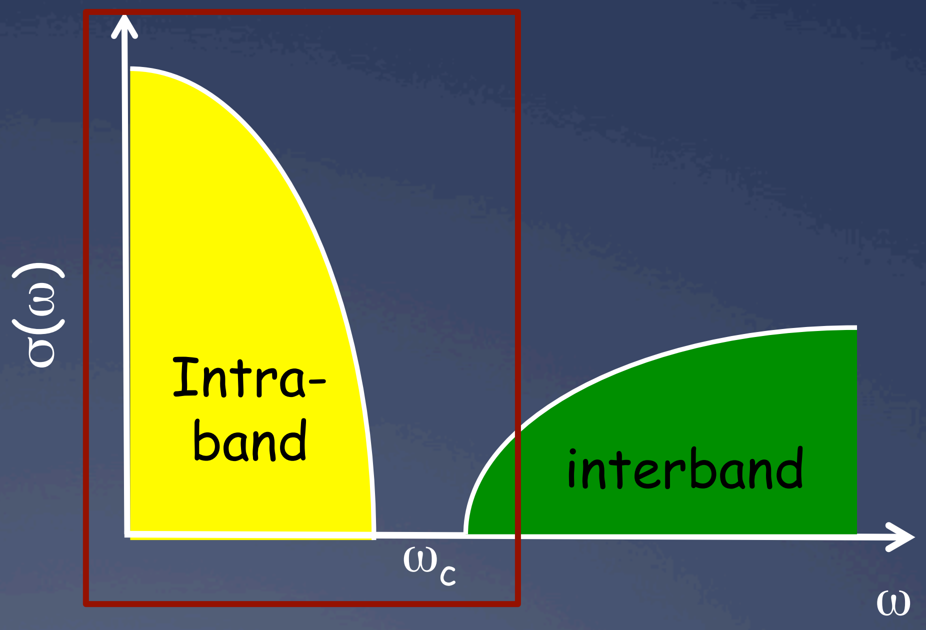
Optical sum rule

$$W = \int_0^{\omega_c} d\omega \mathrm{Re}\sigma_{xx}(\omega) = \frac{\pi e^2}{2N} \sum_{\mathbf{k},\sigma} \frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_x^2} n_{\mathbf{k},\sigma}$$

$$\approx E_{kinetic}$$

- For bands around half-filling W is a measure of the kinetic energy of the carriers, W~K
- W(T) *decreases* as T^2/D in the normal state (Sommerfeld) (D=bandwidth)

$W(T)=W(0)-BT^2$

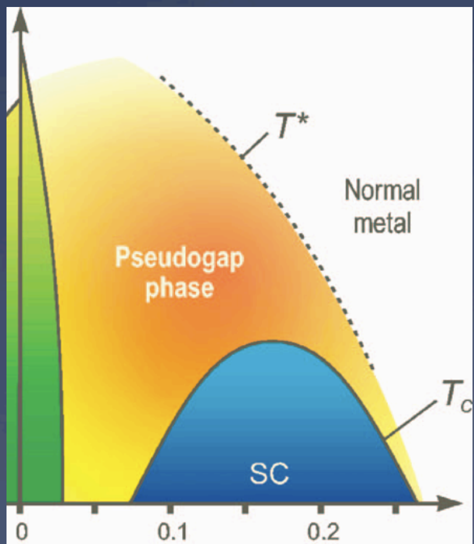


Interacting systems: correlations affect both W and its T dependence

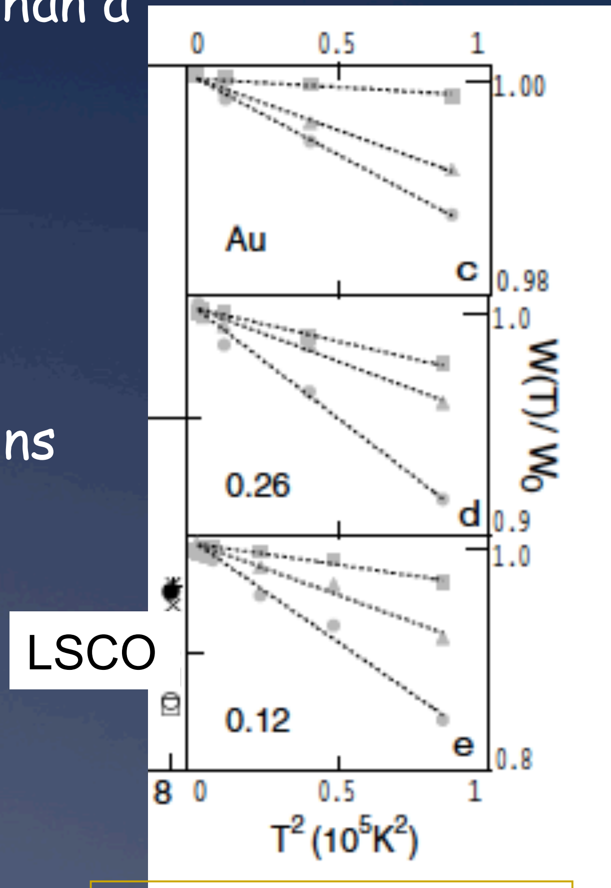
An example: HTSC cuprates

- The absolute value of W is much smaller than a simple LDA estimate W~Dx

- Temperature variations are one order of magnitude *larger* than predicted by Sommerfeld expansion: relative T variations 1-3%



X=hole doping



M.Ortolani et al. PRL 05

A DMFT view

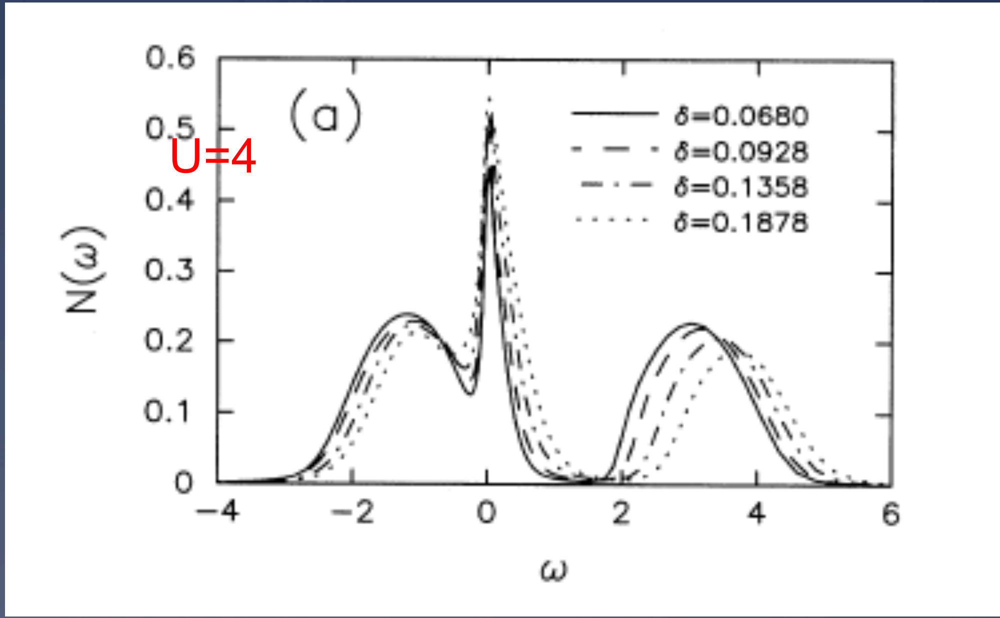
* A model Hamiltonian: the Hubbard model

$$\mathcal{H} = -t\sum_{\langle ij\rangle\sigma} c^{\dagger}_{i\sigma}c_{j\sigma} + U\sum_i n_{i\uparrow}n_{i\downarrow} - \mu\sum_i (n_{i\uparrow} + n_{i\downarrow}),$$

* The DMFT approach: it retains local quantum dynamics but it neglects space fluctuations

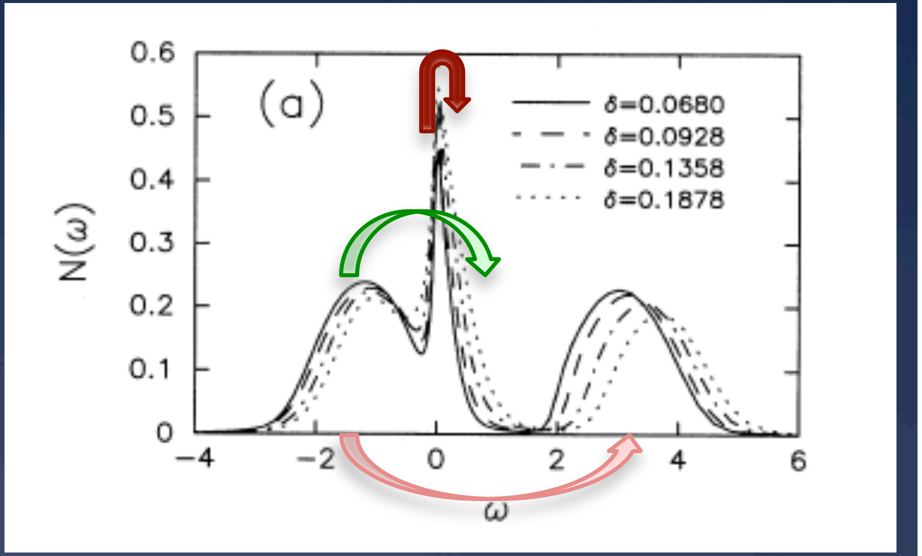
* Main results for the DOS: two incoherent bands around ±U/2, a QP peak with weight Z~doping

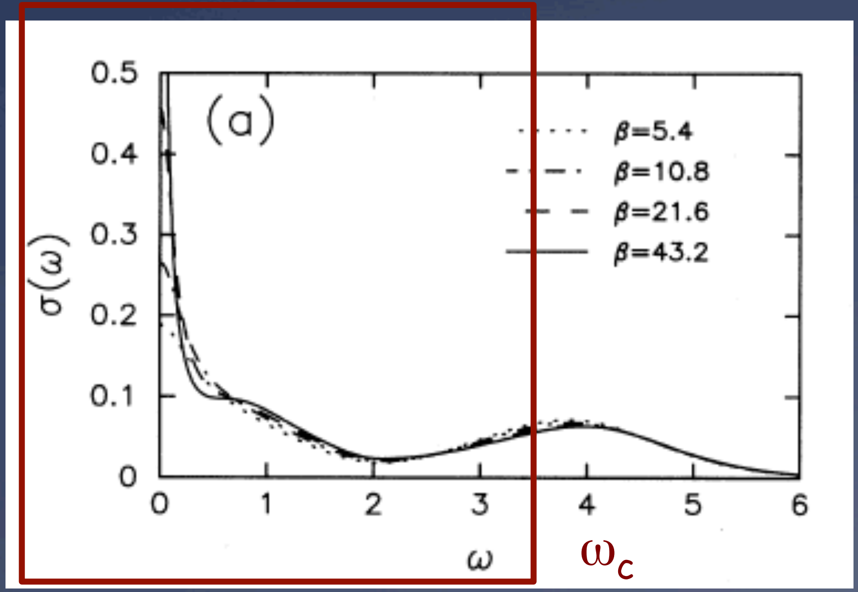
M. Jarrel et al. PRB 51, 11704 (95)



A DMFT viev

* Optical conductivity (bare bubble is exact)
  * coherent part
  * mid-infrared: from QP peak to lower Hubbard band
  * at ω~U transitions between the Hubbard bands

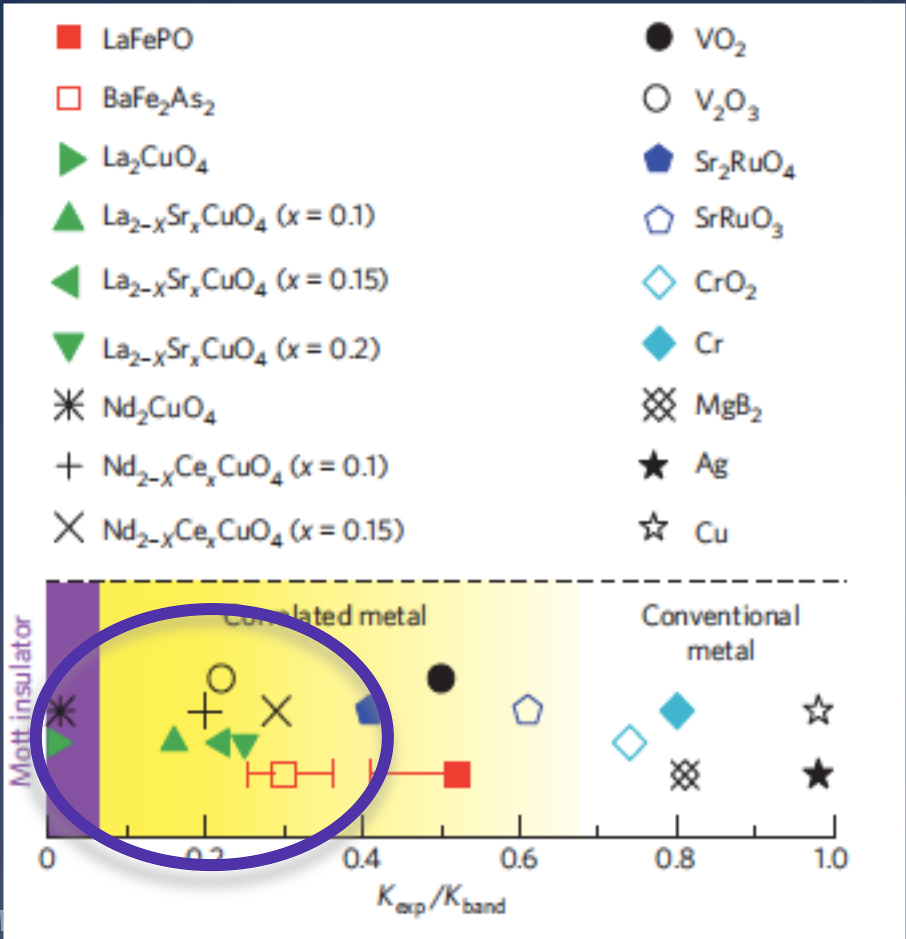


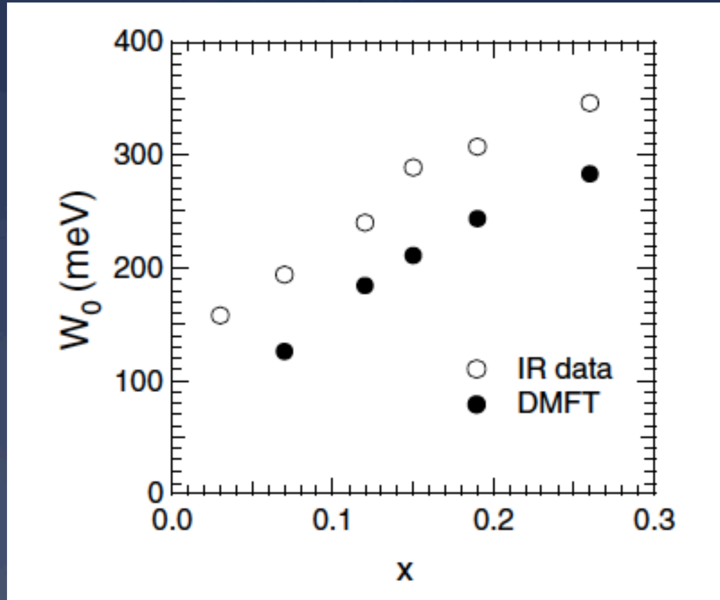


When U>>t a consistent part of the spectral weight is shifted at higher energy than ω_c
The remaining spectral weight is strongly T dependent

A DMFT view

- The 'intraband' spectral weight scales with the strength of the QP peak, i.e. the doping W~Dx





A.Toschi et al. PRL 95, 097002 (05)

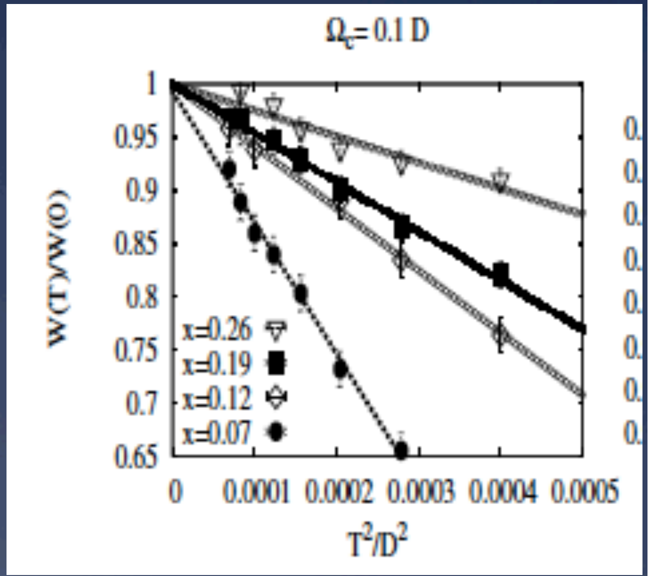
A DMFT view

- The 'intraband' spectral weight scales with the strength of the QP peak, i.e. the doping $W \sim Dx$

- The coefficient of the T^2 dependence is also renormalized

Proximity to a Mott insulator
$W(0) \sim DZ, \quad Z \sim x$
$W(T)=W(0)-BT^2$
$B \sim 1/(DZ) \quad Z \sim x$

Both effects consequence of the bandwidth reduction due to correlations



A.Toschi et al. PRL 95, 097002 (05)

The Eliashberg model

* Electrons interacting with a bosonic mode (phonon, spin fluct., etc.)

$$\Sigma(i\omega_n) = -TV\sum_{\mathbf{k},m} D(\omega_n-\omega_m)G(i\omega_m,\mathbf{k})$$

$$D(\omega_l) = \int d\Omega \frac{2\Omega B(\Omega)}{(\Omega^2+\omega_l^2)}$$

* Bare bubble is enough. The Kubo formula can be recast as (Allen approximation)

$$\sigma(\omega) = \frac{\Omega_P^2}{4\pi}\int d\varepsilon \frac{f(\varepsilon)-f(\varepsilon+\omega)}{-i\omega}\frac{1}{\omega-\Sigma(\omega+\varepsilon)+\Sigma^*(\varepsilon)}$$

* At low energy the self-energy can be approximated as

$$\Sigma = -\lambda\omega - i\Gamma_0$$

$$\lambda = 2VN\int \frac{d\Omega}{\Omega}B(\Omega)$$

* and one recovers a Drude-like formula

$$\sigma(\omega) = \frac{ne^2}{m^*}\frac{\Gamma_0^*}{\omega^2+(\Gamma_0^*)^2}$$

$$m^* = m(1+\lambda) \quad \Gamma_0^* = \frac{2\Gamma_0}{1+\lambda}$$

The 'extended' Drude

* From the Allen formula one can also derive

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

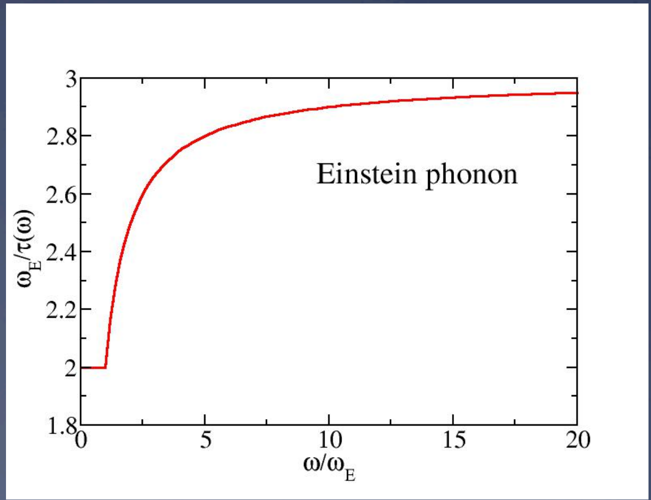
where

$$\frac{1}{\tau(\omega)} = -\mathrm{Im}\Sigma_{opt}(\omega) \simeq \frac{2}{\omega}\int_0^\omega d\Omega(\omega-\Omega)B(\Omega)$$

$$m^*(\omega) = 1 + \mathrm{Re}\Sigma_{opt}/\omega$$

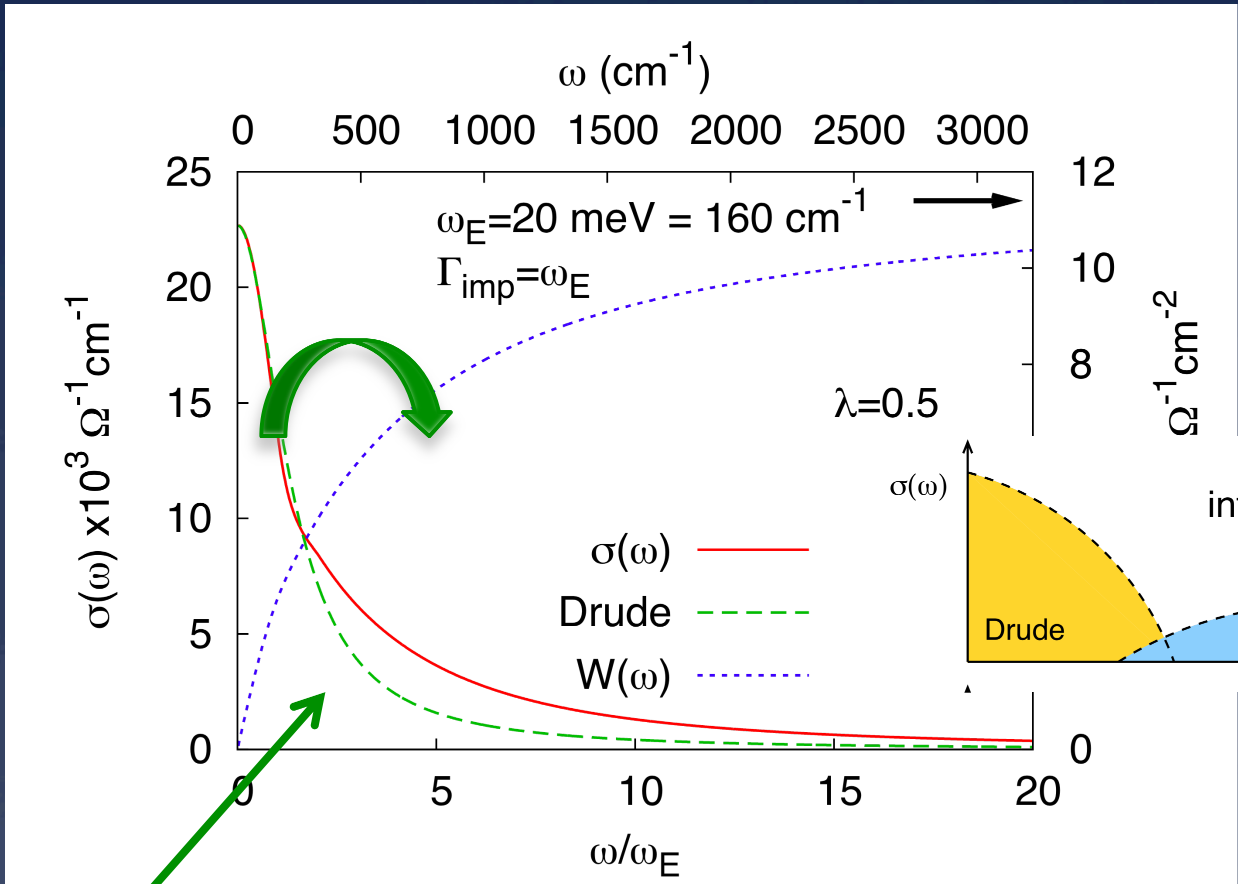
* Einstein phonon $B(\Omega) = \frac{\omega_E}{2}\delta(\Omega - \omega_E)$

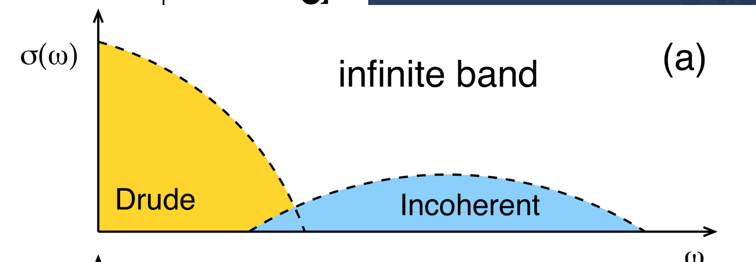
$1/\tau$ saturates at $\omega \gg \omega_E$



See e.g. Shulga, Dolgov and Maksimov Physica C 178, 266 (1991)
Norman and Chubukov PRB 73, 14501 (2006)

The optical sum rule

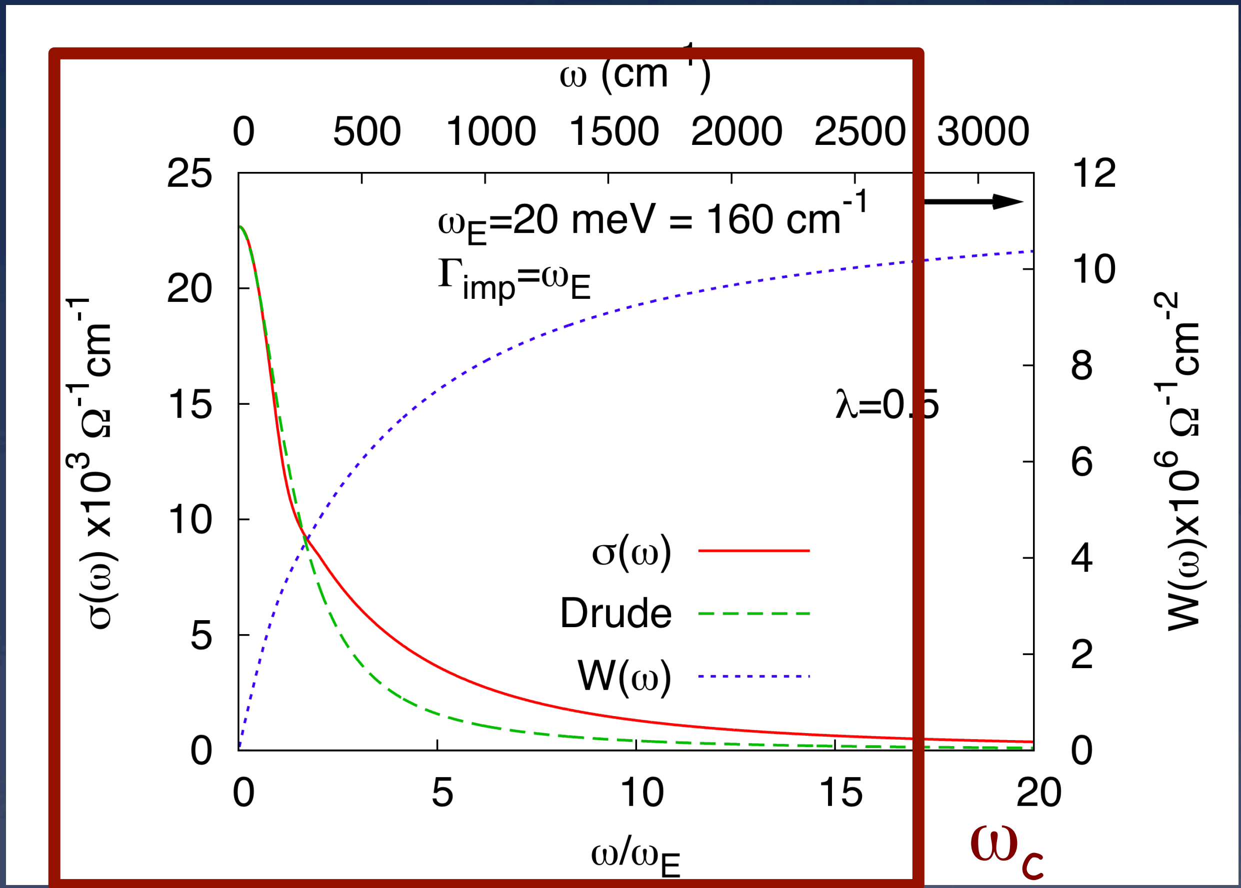




The sum rule is more or less satisfied

$$W_{Drude} \sim \frac{n}{m^*} = \frac{n}{m(1+\lambda)}$$ + Incoherent= $$W \sim \frac{n}{m}$$

The optical sum rule



$$\Delta W(\omega_c) = \Delta W_K + \Delta f(\omega_c)$$

Norman et al.
PRB 76, 220509 (2007)

The sum rule is more or less satisfied
unless the boson energy or the coupling are very large,
so the cut-off introduces an additional T dependence
… as it could be in the case of cuprates

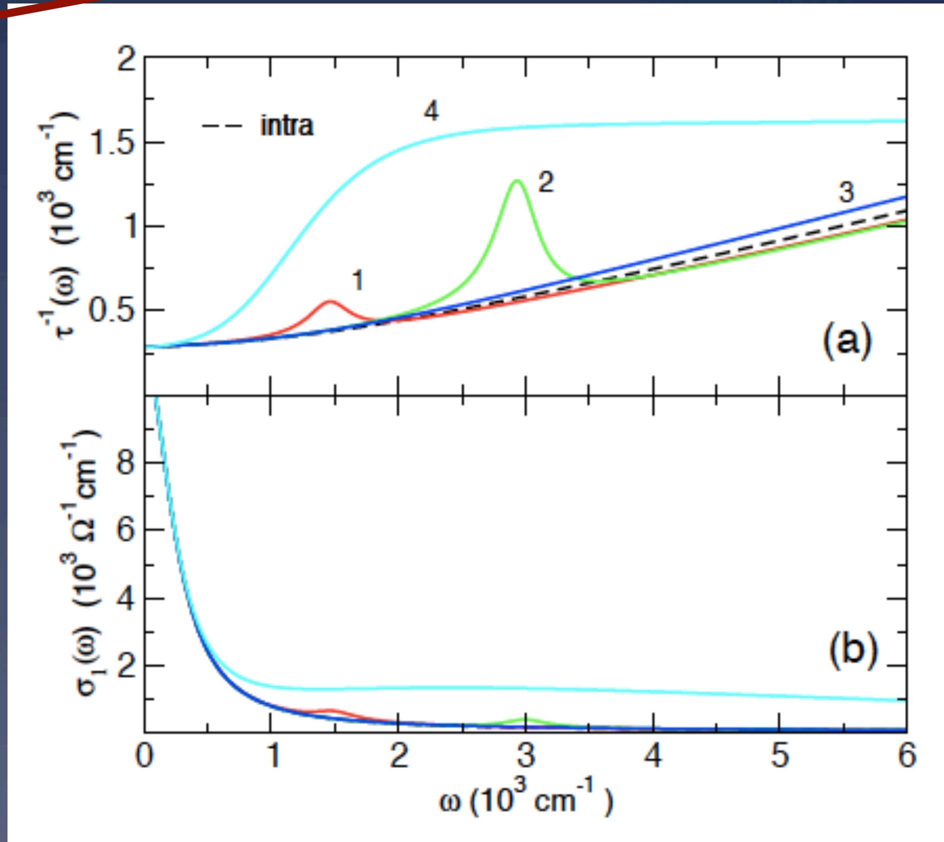
The extended Drude in practice

* The Eliashberg theory is the foundation of the 'extended' Drude analysis of the experiments

$$\frac{1}{\tau(\omega)} = -\mathrm{Im}\Sigma_{opt}(\omega) \simeq \frac{2}{\omega}\int_0^{\omega} d\Omega(\omega-\Omega)B(\Omega)$$

$$\tau^{-1}(\omega) = \frac{\omega_P^2}{4\pi}\frac{\sigma_1(\omega)}{\sigma_1^2(\omega)+\sigma_2^2(\omega)},$$

$$\frac{m^*(\omega)}{m} = \frac{\omega_P^2}{4\pi\omega}\frac{\sigma_2(\omega)}{\sigma_1^2(\omega)+\sigma_2^2(\omega)},$$



* However, one should be very careful to avoid spurious contributions that could be erroneously attributed to the 'anomalous' intraband part

L. Benfatto et al. PRB 83, 224514 (11)

Take-home messages

* Hubbard-like models with strong interactions (U~2-3 eV) lead to a sizeable bandwidth renormalization, that reflects in an overall reduction of the sum rule with respect to DFT estimates and to an increase of the T^2 dependence

$$\frac{W_{\exp}}{W_{DFT}} \rightarrow \frac{m_{DFT}}{m_{\exp}}$$

"High-energy" correlation effect

* Eliashberg-like calculations can account for (temperature-dependent) spectral-weight redistribution in the far-infrared/infrared due to interaction with collective modes at energies ~0.1-0.5 3V

"Low-energy" correlation effect

$$\frac{1}{\tau(\omega)} = -\mathrm{Im}\Sigma_{opt}(\omega) \simeq \frac{2}{\omega}\int_0^{\omega} d\Omega(\omega-\Omega)B(\Omega)$$

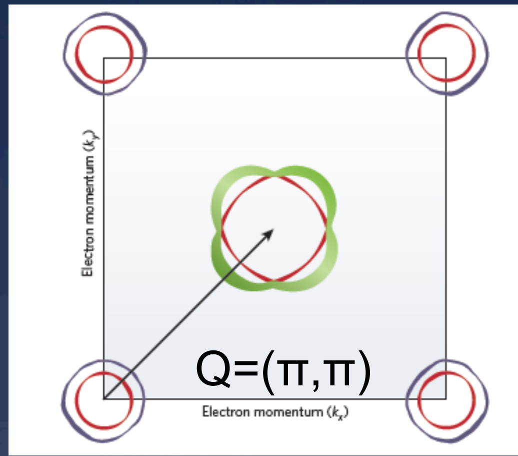
However, such a distinction is not always straightforward
The cut-off used in the experimental estimate can induce additional temperature effects

Pnictides

What is new in pnictides?

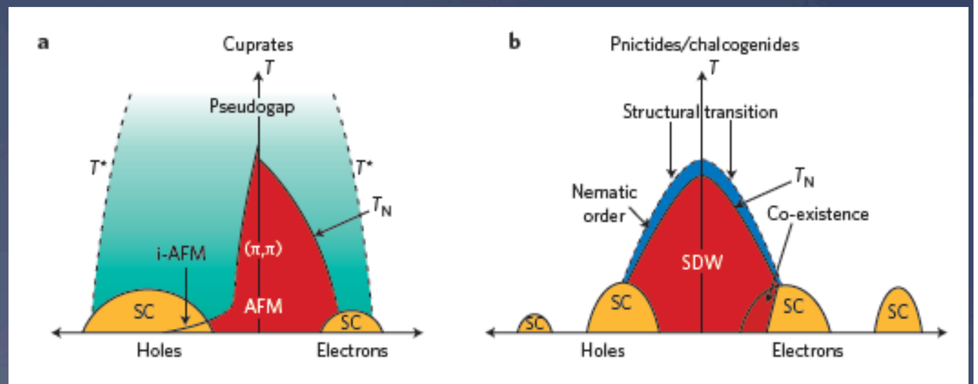
* Multiband systems

(already seen in MgB2, graphene, etc.)



* Proximity/coexistence with an AF phase, possible relevance of spin-fluctuations mediated interactions

(already seen in heavy fermions, cuprates, etc.)

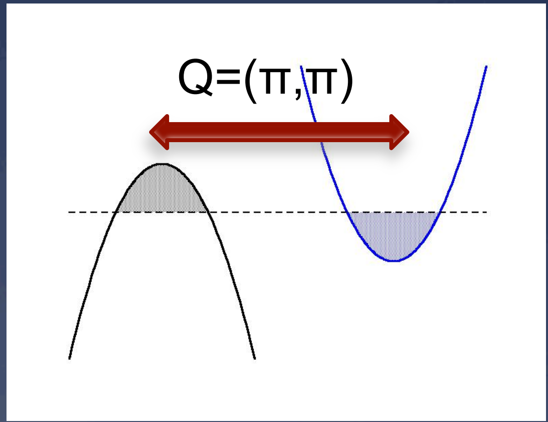


What is new in pnictides?

- Multiband systems

(already seen in MgB2, graphene, etc.) → **Coexistence of hole and electron bands**

- Proximity/coexistence with an AF phase, possible relevance of spin-fluctuations mediated interactions



(already seen in heavy fermions, cuprates, etc.) → Interband interaction

Optical sum rule in pnictides

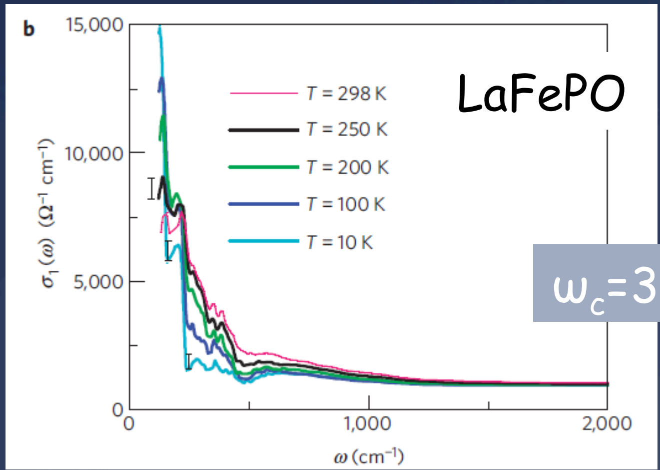
$$W = \int_0^{\omega_c} d\omega \mathrm{Re}\sigma_{xx}(\omega) = \frac{\pi e^2}{2N} \sum_{\mathbf{k},\sigma} \frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_x^2} n_{\mathbf{k},\sigma} \approx \frac{\pi e^2 n}{m_b}$$

- Almost empty bands, W~n/m
- One would expect negligible T dependence, but correlations can affect m_b with resepct to DFT predictions and then the absolute value

$$\frac{W_{\exp}}{W_{DFT}} \rightarrow \frac{m_{DFT}}{m_{\exp}}$$

Optical sum rule in pnictides

* Experimental spectral weight smaller than DFT prediction

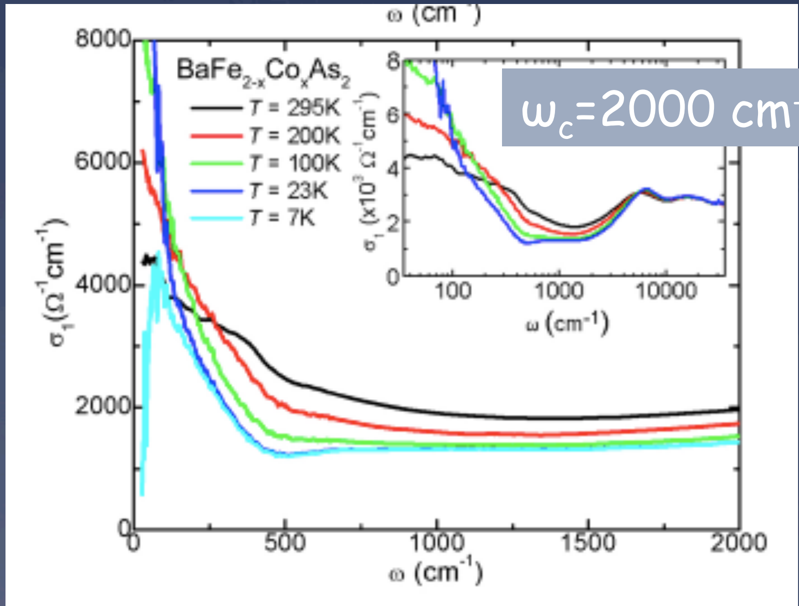


$$W_{exp} = \int_0^{\omega_c} d\omega \mathrm{Re}\sigma(\omega)$$

$$\frac{W_{exp}}{W_{DFT}} \approx 0.5$$

ω_c=3000 cm^{-1}

M. M. Qazilbash et al. Nat. Phys. 5, 647 (2009)



ω_c=2000 cm^{-1}

"Standard" correlated metal

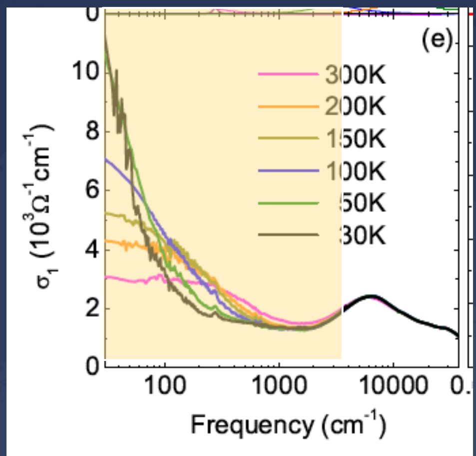
$$\frac{W_{exp}}{W_{DFT}} \approx 0.3$$

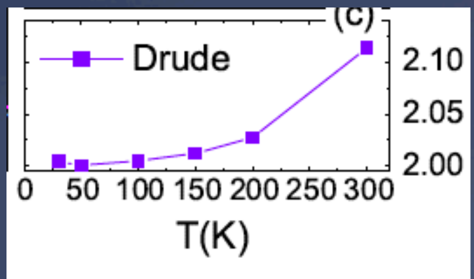
A.A.Schafgans et al. PRL 108, 147002

The T dependence and spectral-weight redistribution

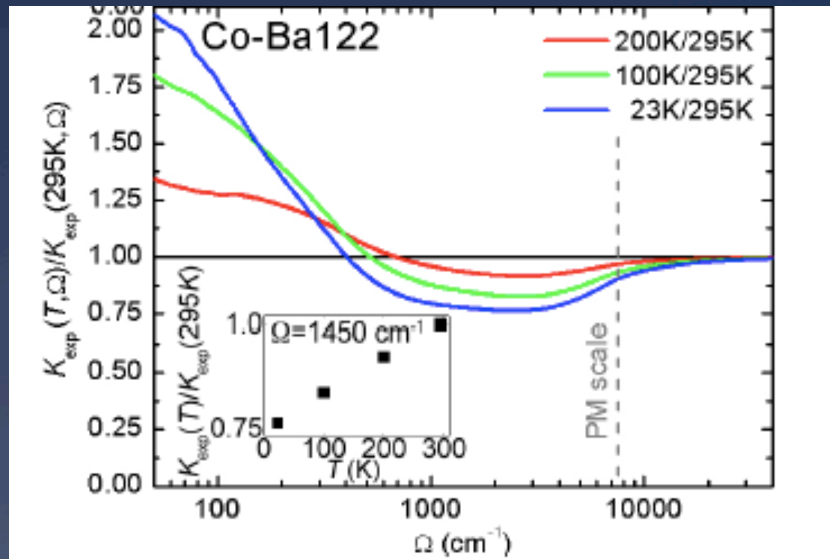
- The low-energy optical sum rule *increases* as T increases, in contrast to any 'standard' correlated material

$EuFe_2(As_{0.82}P_{0.18})_2$





D. Wu et al. PRB 83, 100503 (11)



A.A.Schafgans et al. PRL 108, 147002

The spectral weight moves to a very large energy range (but smaller than ~U)

'Anomalous' sum rule in pnictides

- Absolute value of the sum rule: bandwidth reduction due to correlations $m_b \sim 2m_{DFT}$ (see also ARPES, de Hass van Alphen, specific heat, etc)

$$W_{exp} \approx \frac{\pi e^2 n}{m_b} \qquad W_{DFT} = \frac{\pi e^2 n_{DFT}}{m_{DFT}}$$

- Temperature dependence of the 'intraband' part: anomalous increase with temperature

- One more problem: how do we interpret the sum rule in a compensated multiband metal???

If $n=n_{DFT}$ then mass renormalization $m_b/m_{DFT} \sim 2$

What happens if n is different form its DFT estimate n_{DFT} ???

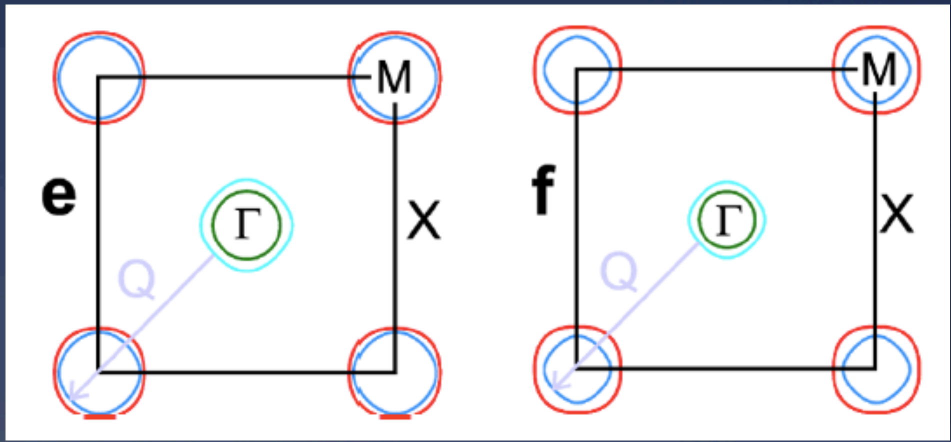
Fermi-surface shrinking in dHvA

- De Haas Van Alphen experiments measure the Fermi surface areas for various bands

- Shrinking of all the FS with respect to DFT:
  - *Upward* shift of the electron band
  - *Downward* shift of the hole bands

A. Coldea et al., Phys. Rev. Lett. **101**, 216402 (2008)

unshifted LDA shifted LDA



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A. Coldea et al., Phys. Rev. Lett. **101**, 216402 (2008)

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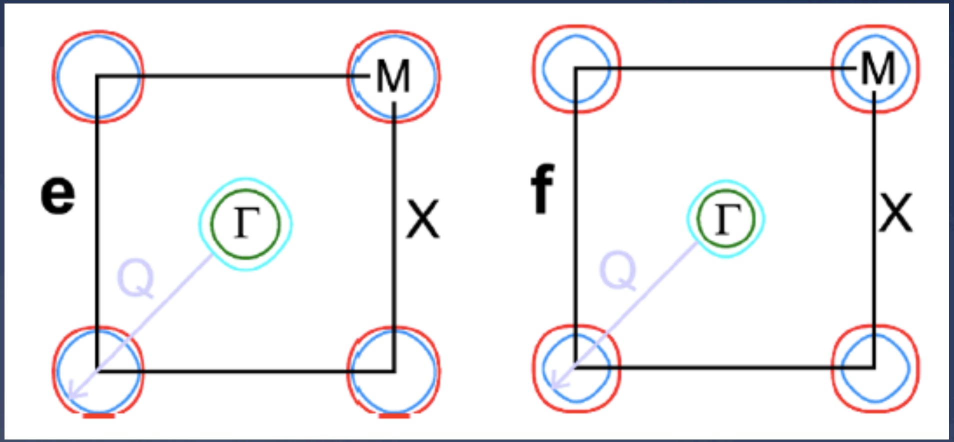
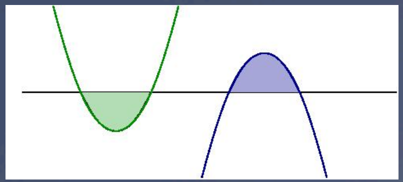


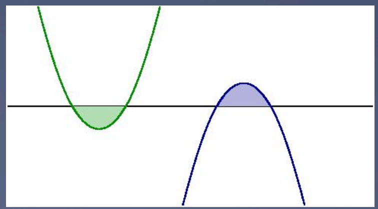
- Shrinking of all the FS with respect to DFT

- Total number of particles is still conserved

$n=n_e+(2-n_h)$

n_e-n_h is conserved, not n_e+n_h

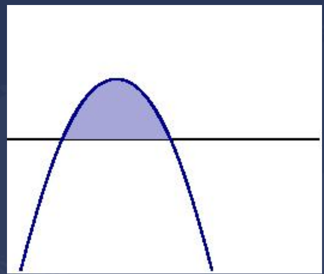


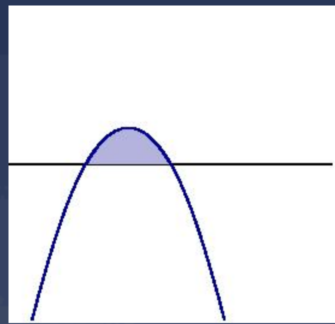


Fermi-surface shrinking and sum rule

* FS shrinking from a rigid-band shift of LDA bands: change in the number of carriers in *each* band

n_{DFT}





$n < n_{DFT}$

* In this case the sum-rule reduction cannot be attributed only to correlations (see I. Mazin arXiv:0910.4117)

$$W_{DFT} = \frac{\pi e^2 n_{DFT}}{m_{DFT}}$$

$$W_{exp} \approx \frac{\pi e^2 n}{m_b}$$

Fermi-surface shrinking and sum rule

- FS shrinking from a rigid-band shift of LDA bands: change in the number of carriers in *each* band

n_{DFT}

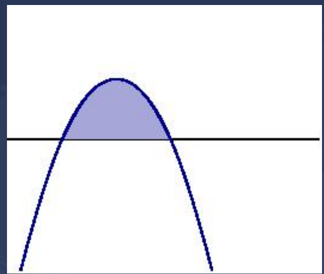
$n < n_{DFT}$

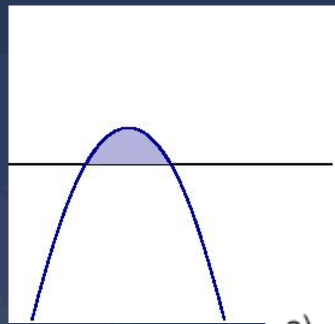
However, experiments (dHvA and ARPES) show a **systematic** shrinking of the Fermi surface, that can hardly be understood as a systematic inaccuracy of DFT

Fermi-surface shrinking and sum rule

- FS shrinking from a rigid-band shift of LDA bands: change in the number of carriers in *each* band

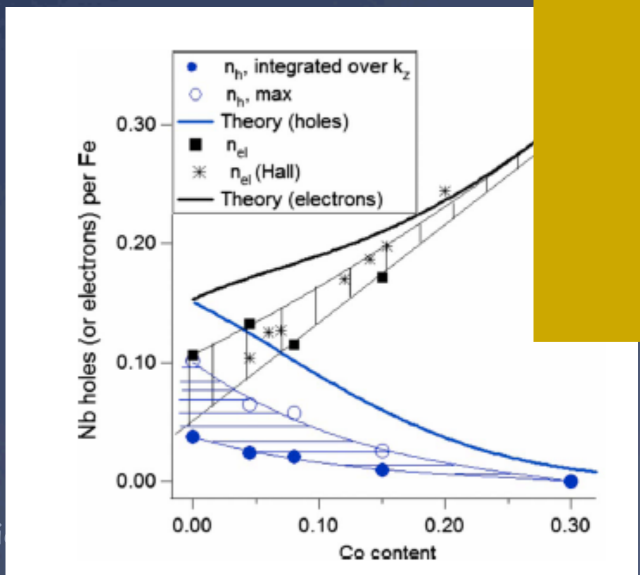
n_{DFT}



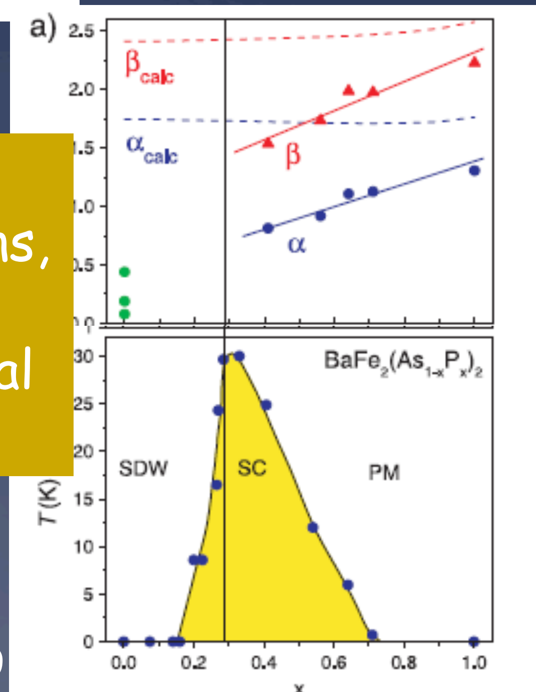


$n < n_{DFT}$

V. Brouet et al. PRB 2009



The FS shrinking follows from interband interactions, and it has non trivial consequences on the optical sum rule



H. Shishido et al. PRL 2010

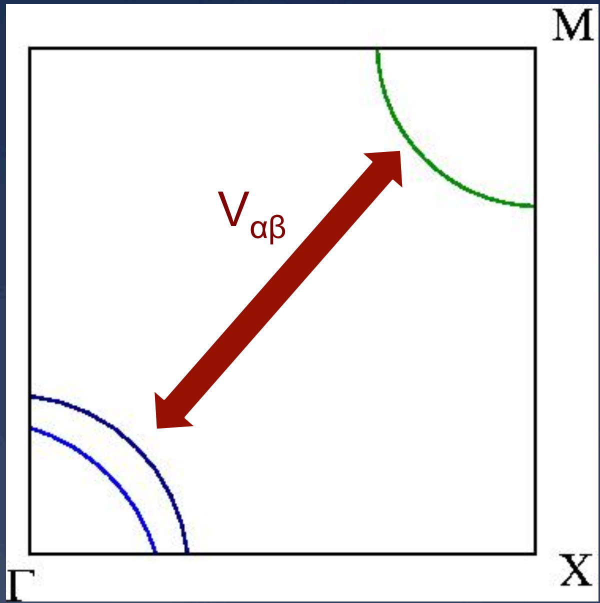
Eliashberg approach

- hole+electron (parabolic) bands coupled to a bosonic mode with characteristic energy ω_0 and coupling λ=VN

$$\Sigma_\alpha(i\omega_n) = -T\sum_{m,\beta} V_{\alpha,\beta} D(\omega_n-\omega_m) G_\beta(i\omega_m)$$

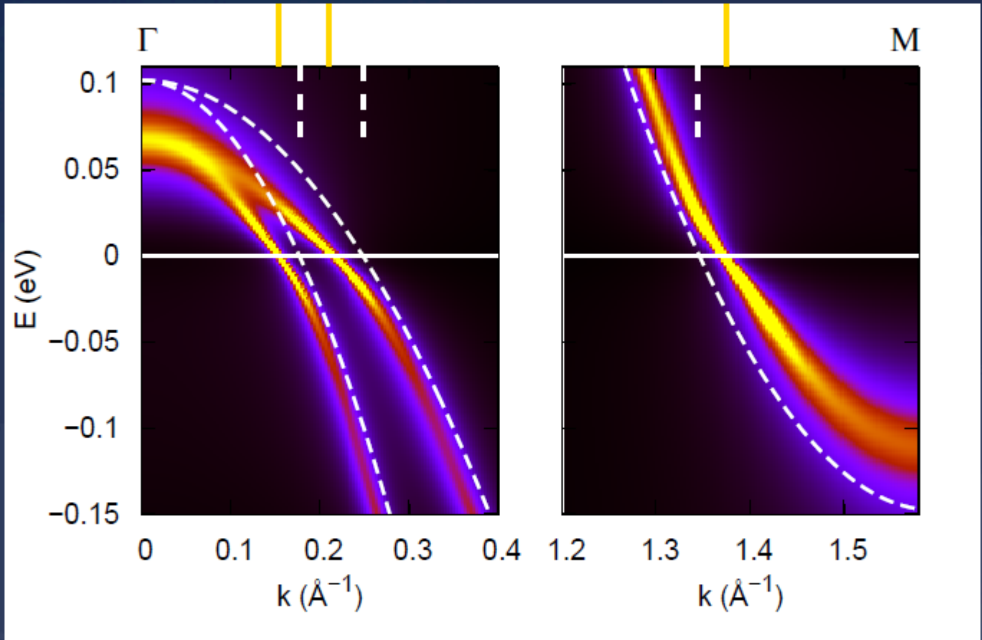
$$B(\Omega) = \frac{1}{\pi}\frac{\Omega\omega_0}{\omega_0^2+\Omega^2}$$

$$D(\omega_l) = \int d\Omega \frac{2\Omega B(\Omega)}{(\Omega^2+\omega_l^2)}$$



- We assume only interband interactions (spin fluctuations)
- Strong particle-hole asymmetry (almost empty bands) crucial to explain the Fermi-surface shrinking

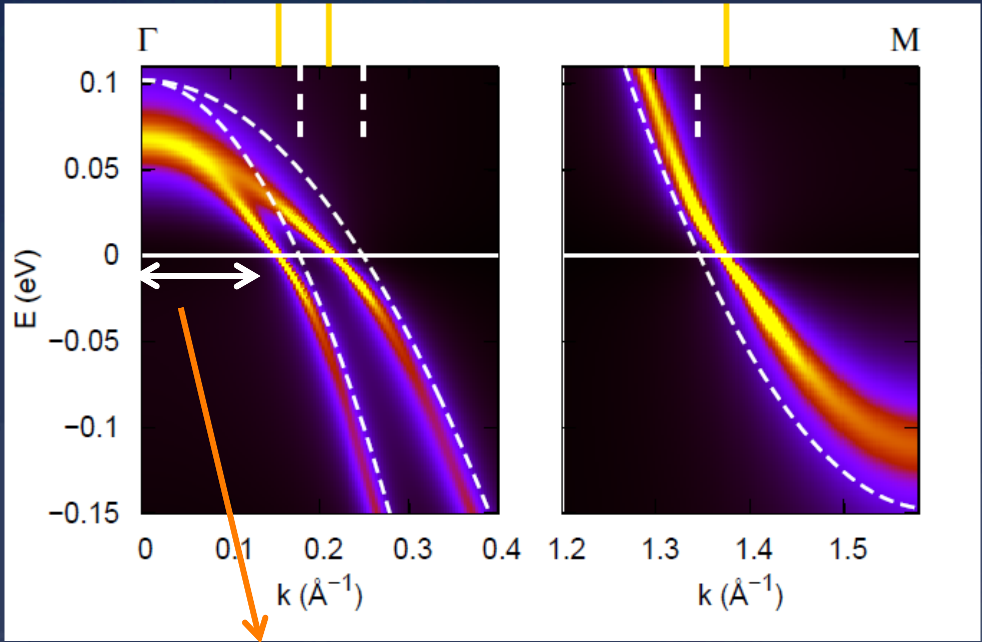
Spectral functions

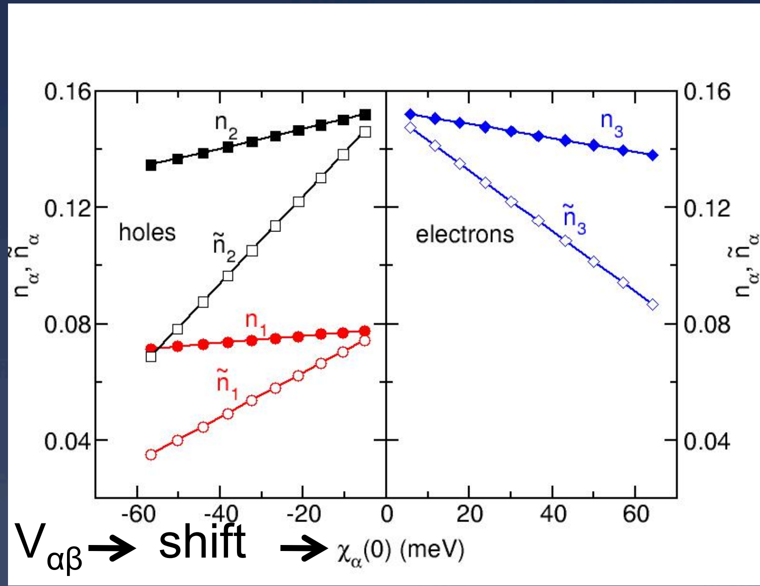


L.Ortenzi, E.Cappelluti, L.B., L.Pietronero, Phys Rev Lett 103, 046404 (2009

* Band shift due to interactions
* When interactions have predominant interband character the Fermi-surfaces get reduced
* At the same time, one observes a redistribution of the spectral weight towards the unoccupied part of the band

Fermi-surface shrinking and number of particles





$$\tilde{n} = \frac{a^2 k_F^2}{2\pi}$$

≠

$$n = \int d\omega f(\omega) N(\omega)$$

Number of carriers associated to the Fermi area

Strongly affected by the interactions -> shift -> shrinking

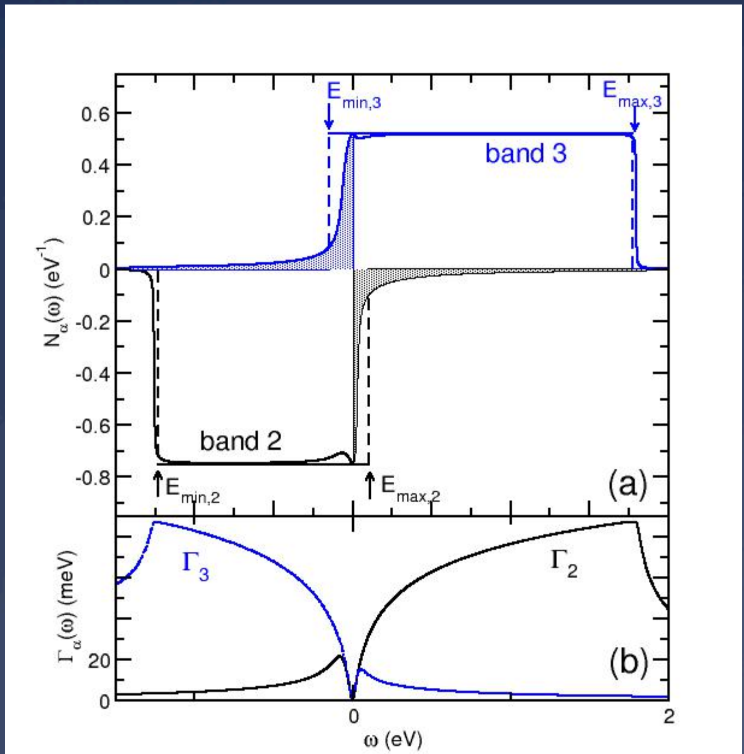
Total carrier concentration (accounts for occupation at all energies)

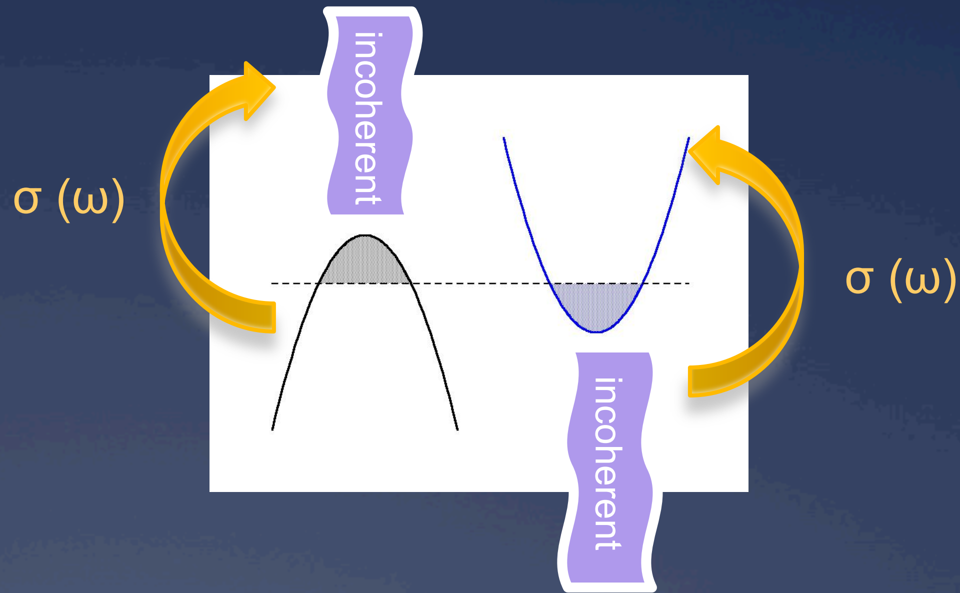
Almost unaffected by interactions

This has profound consequences on the optical conductivity

Transfer of spectral weight

* The interband interaction leads to a finite DOS in the otherwise unoccupied part of the spectrum





Incoherent optical transitions up to very large energy scales (of the order of the bandwidth)

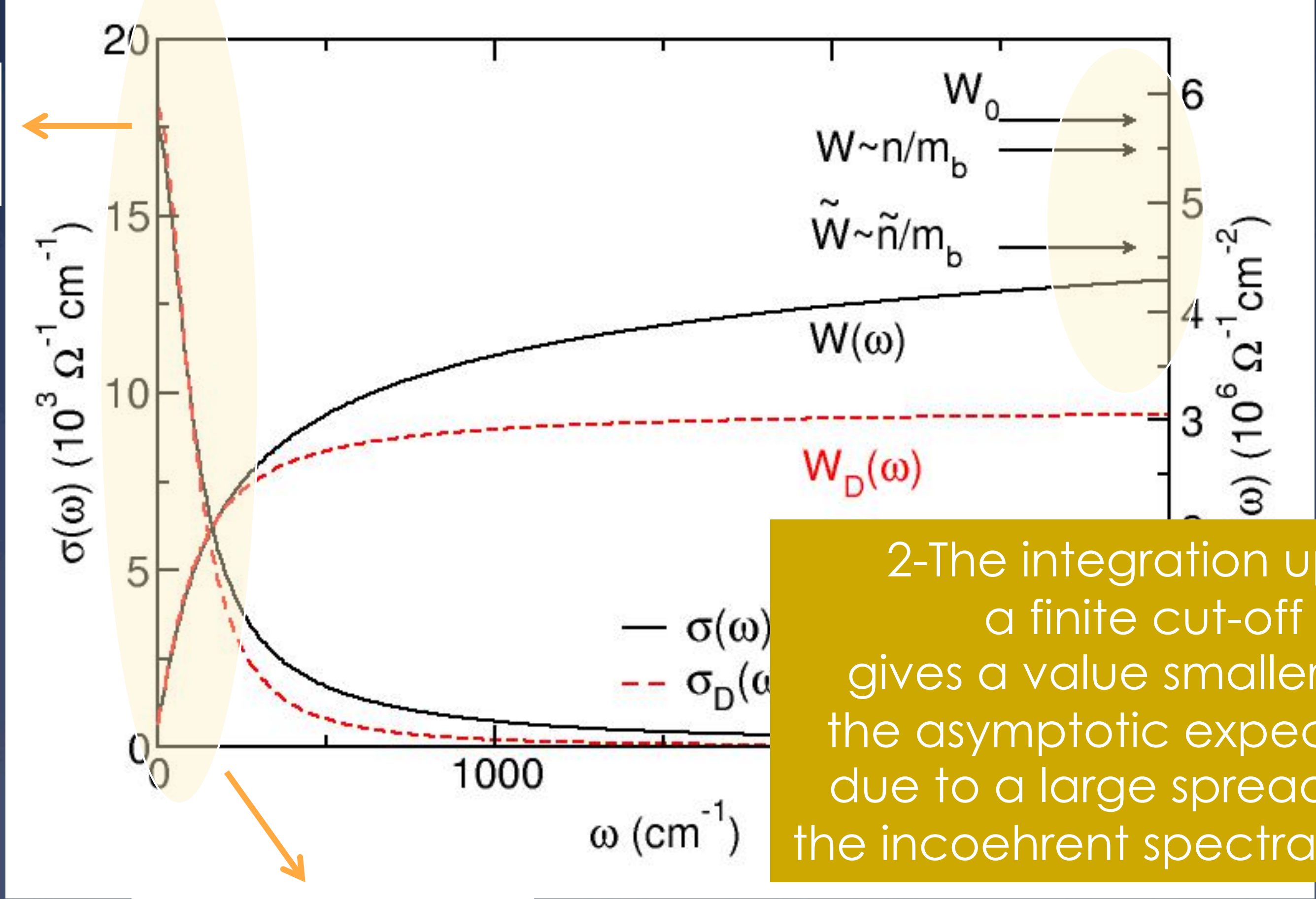
$$N_\alpha(\omega) = \int d\varepsilon A_\alpha(\varepsilon,\omega)$$

$$A_\alpha(\varepsilon,\omega) = \frac{1}{\pi}\frac{\Gamma_\alpha(\omega)}{[\omega-\varepsilon-\chi_\alpha(\omega)]^2+[\Gamma_\alpha(\omega)]^2}.$$

Optical conductivity

1-The dc value measures the Fermi-surface area

$$\sigma_{dc} = \frac{\tilde{n} e^2 \tau_{tr}}{m^*}$$

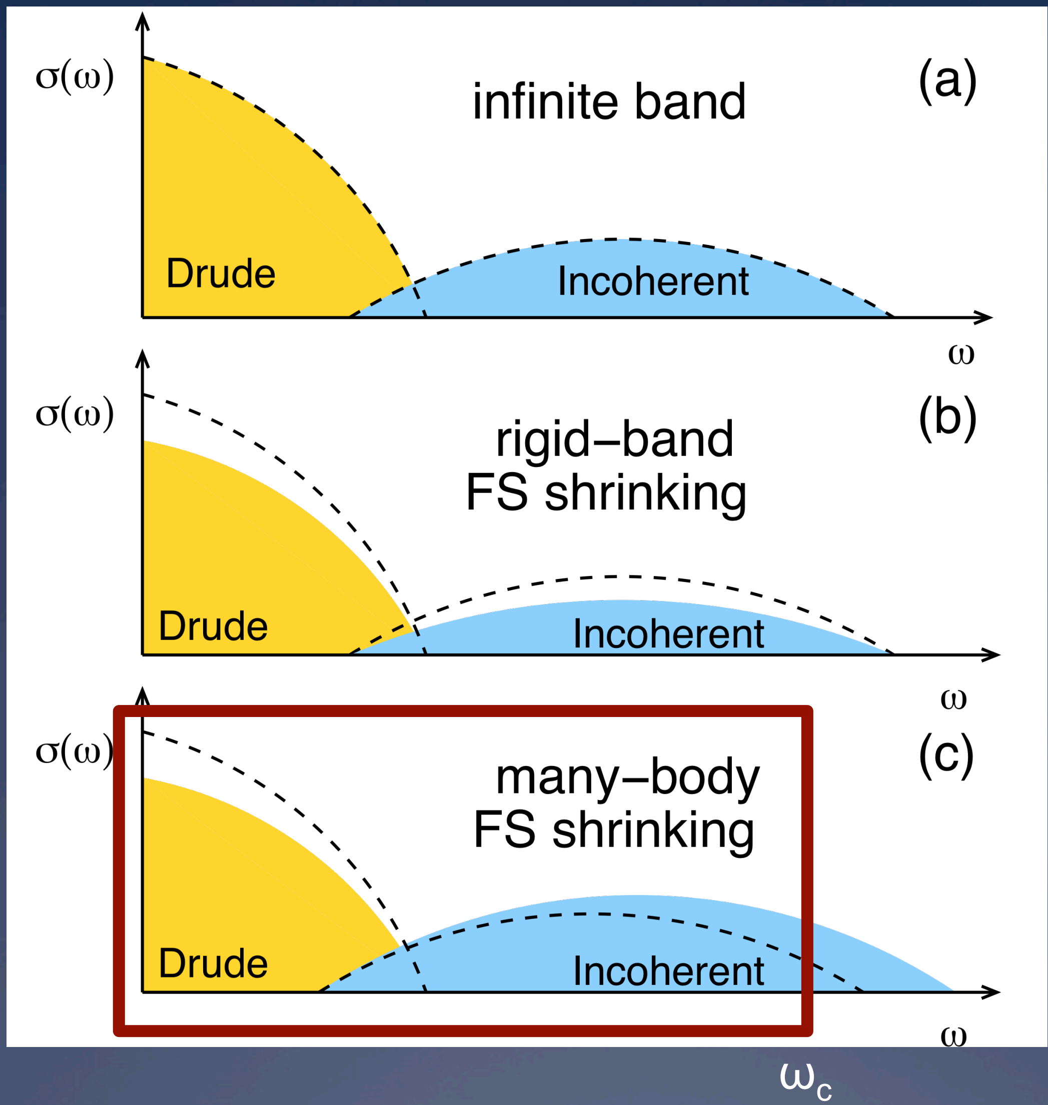


2-The integration up to
a finite cut-off
gives a value smaller than
the asymptotic expectation
due to a large spreading of
the incoehrent spectral weight

$$W_{Drude} \approx \frac{\pi e^2 \tilde{n}}{m^*}$$

$m^* = (1+\lambda)\, m_b$
renormalization due to spin fluctuations

Rigid-band shift vs interactions



Total area n/m_b

Total area n'/m_b

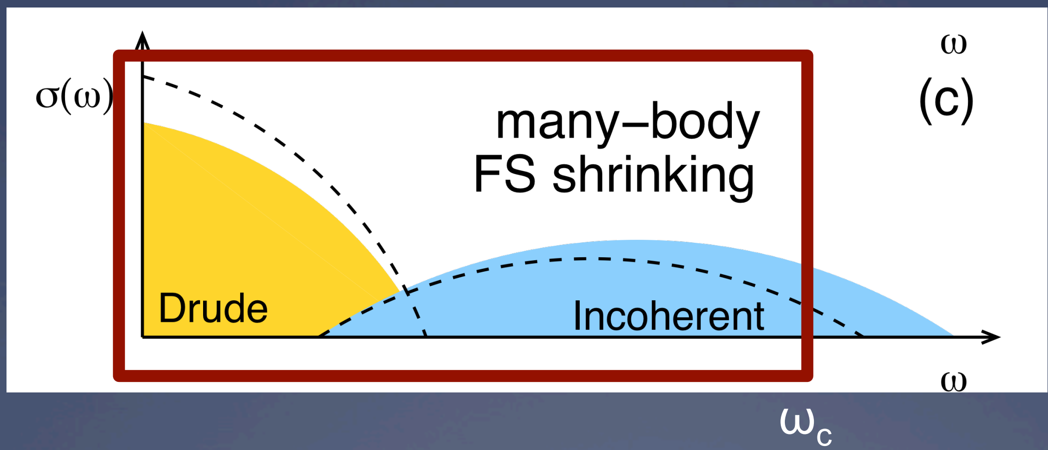
Total area: in principle n/m_b

in practice it depends on the cutoff

Rigid-band shift vs interactions

N.B. The incoherent spectral weight is transferred up to energies much larger than the typical threshold ω_c for interband transitions

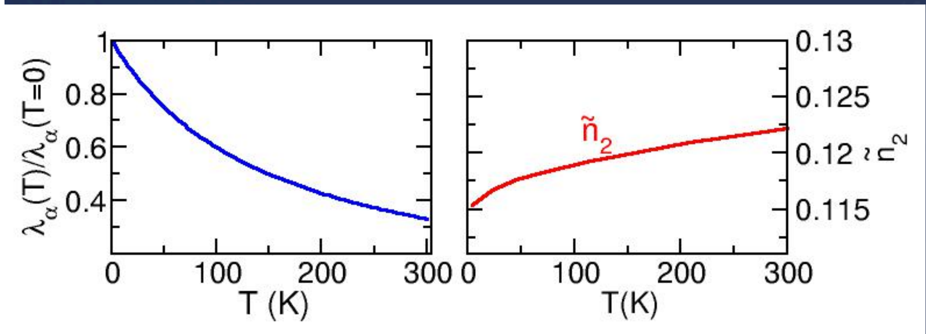
As a consequence, the experimental spectral weight probes the T dependence of the interactions, which control both the Fermi-surface shrinking and the transfer of spectral weight to the incoherent processes



Total area:
in principle
n/m_b
in practice
it depends
on the cutoff

Temperature dependence

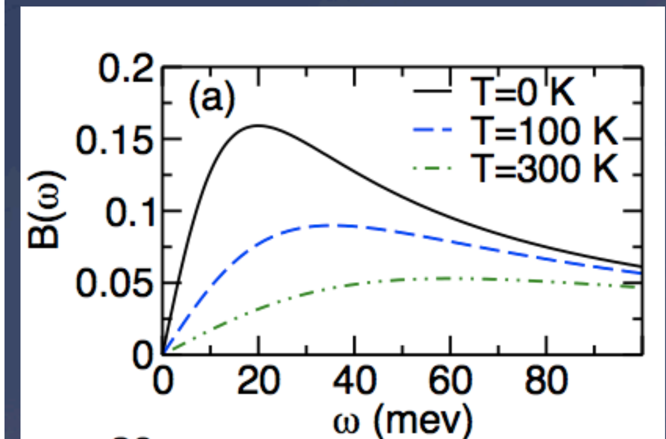
* We assume that the coupling to the bosonic mode decreases as T increases (as measured by neutrons)

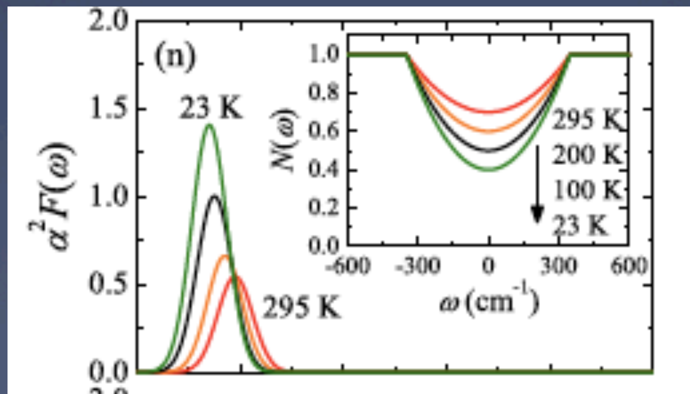


The coherent charge-carrier concentration increases with increasing temperature

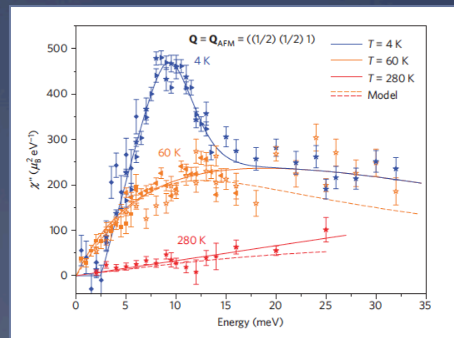
Optics

Neutron scattering





S.J.Moon.PRL 109, 027006 (2012)



Inosov et al. Nat. Phys. 6, 178 (10)

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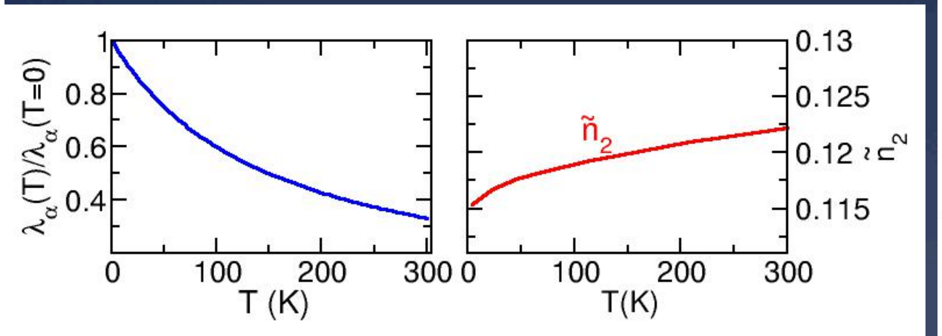
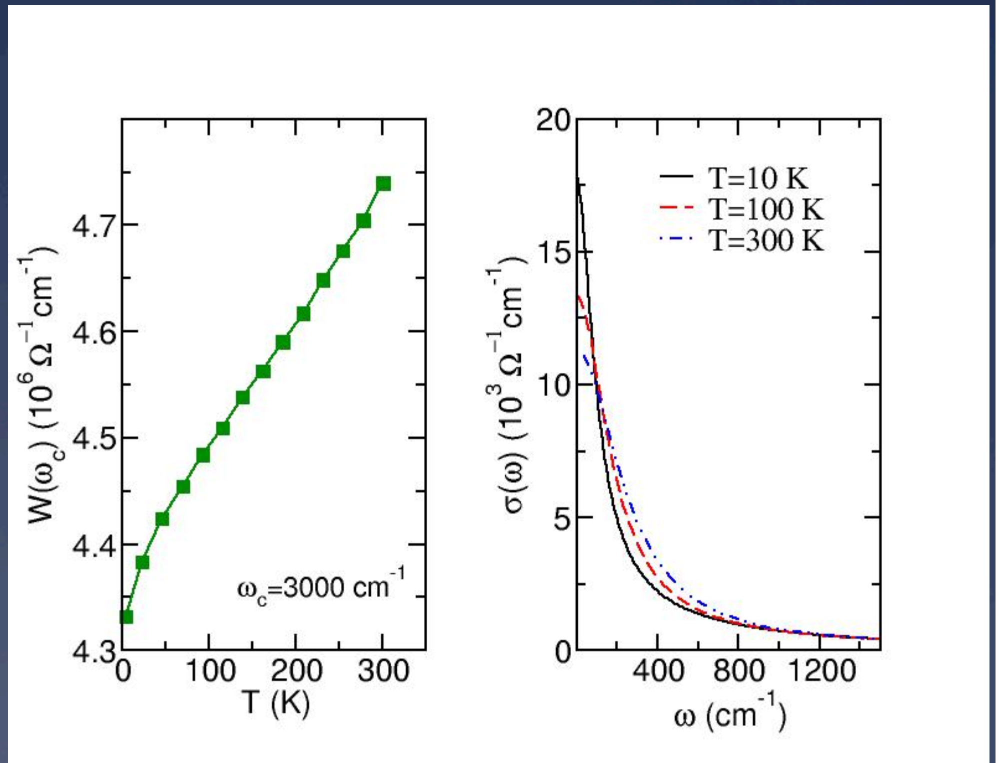




The coherent charge-carrier concentration increases with increasing temperature

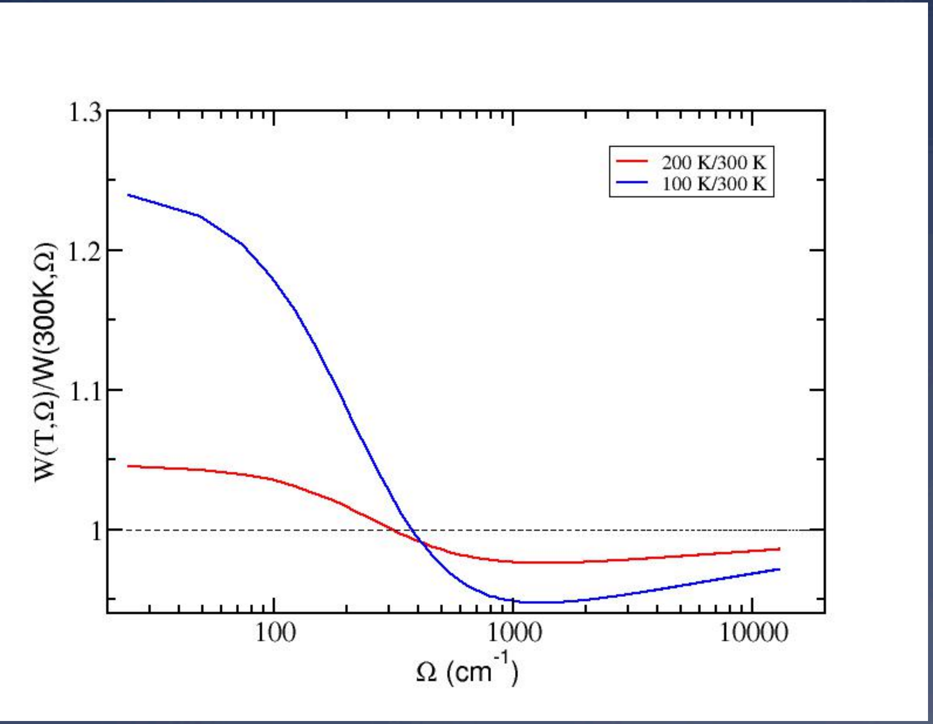
The low-energy spectral weight **increases**

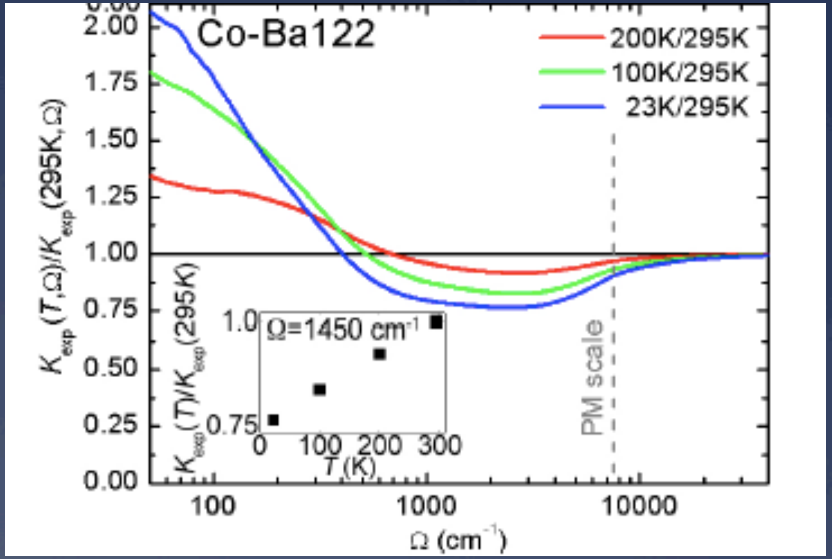
S.J.Moon.PRL 109, 027006 (2012)
L. Benfatto and E. Cappelluti, PRB 83, 104516 (11)

S.J.Moon.PRL 109, 027006 (2012)

Temperature dependence

* The spectral weight redistribution occurs over large energy scales, as in the experiments



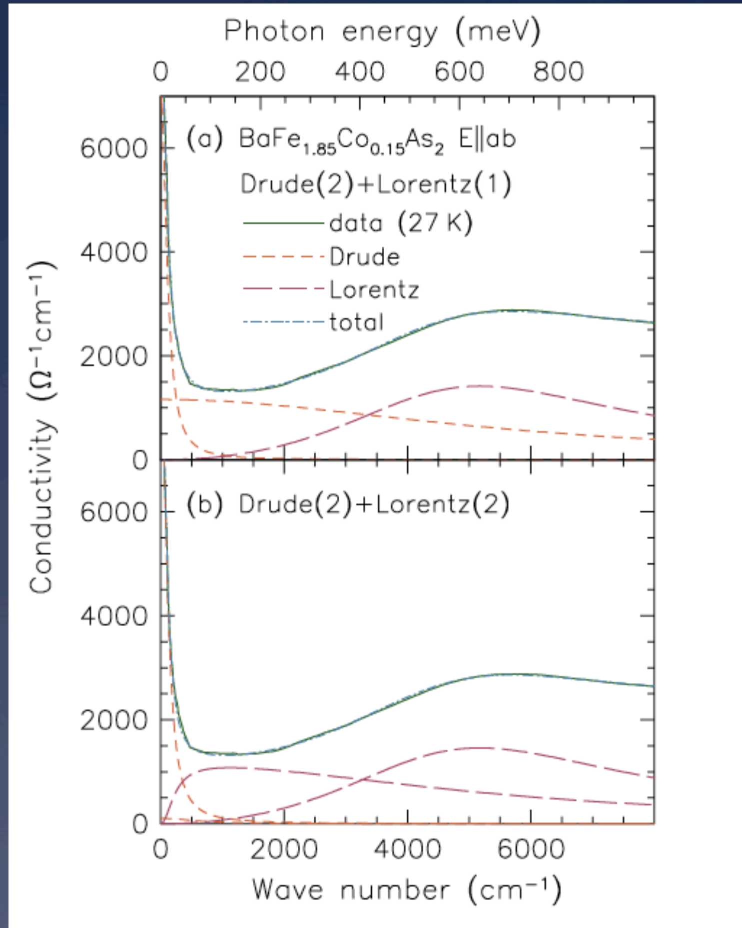


A.A.Schafgans et al. PRL 108, 147002

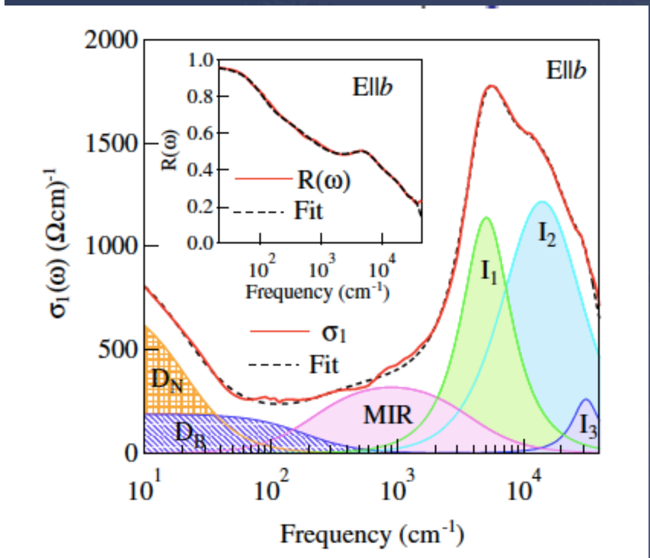
L. Benfatto and E. Cappelluti, PRB 83, 104516 (11)

Comments

* The experimental data for the optical conductivity are very "flat" even in the far-infrared:
  * large anisotropy of the scattering rates of hole/ electrons??
  * interband transitions??



J.J. Tu, PRB 82, 174509 (10)



A. Dusza et al. NJP 14, 023020 (12)

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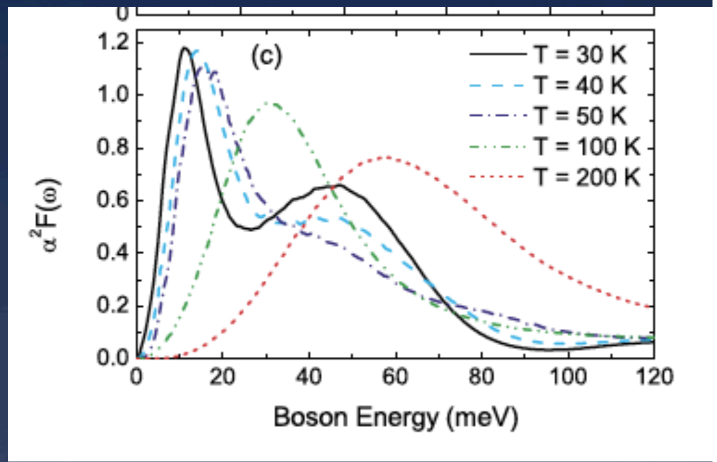
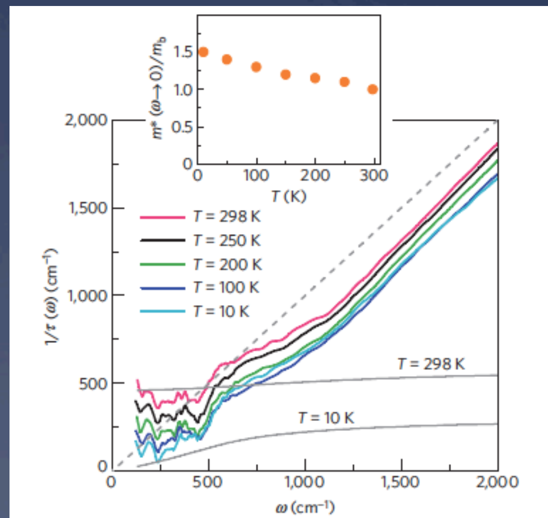
  - why the extended Drude Model analysis leads to a coupling λ~ 4 much larger than obtained by other probes??



D.Wu et al, PRB 82, 144519 (10)



M. M. Qazilbash et al. Nat. Phys. 5, 647 (2009)

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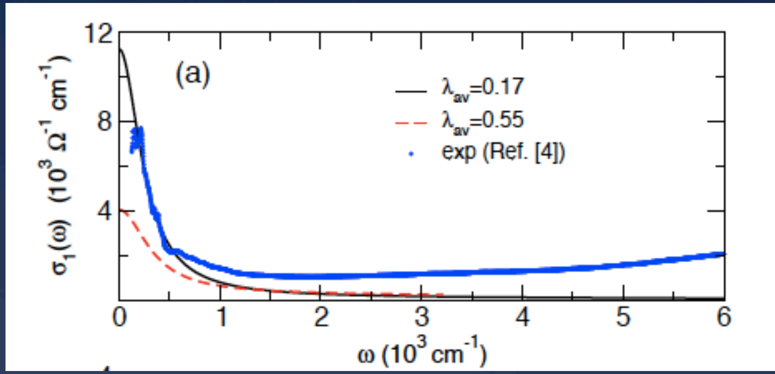

L. Benfatto et al. PRB 83, 224514 (11)

Even our mechanism of large transfer of INTRABAND spectral weight is not enough
Most likely candidate are INTERBAND transitions

see also
E. van Heumen, EPL 10
S.L.Drechsler et al., PRL 08, arXiv:0904.0827
A. Charnukha et al. PRB 11

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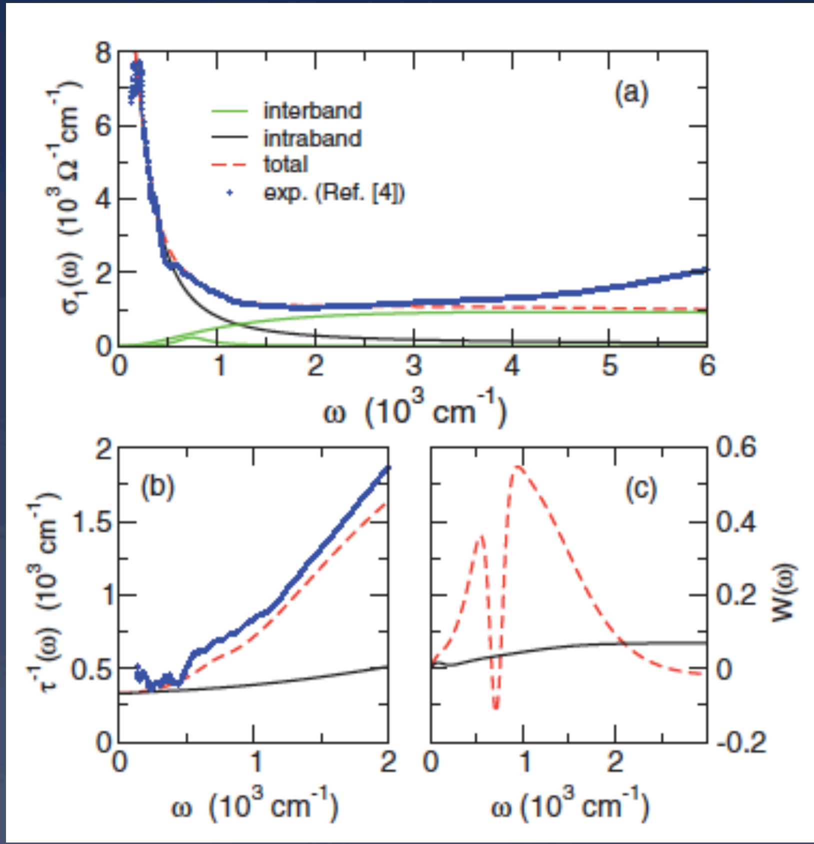

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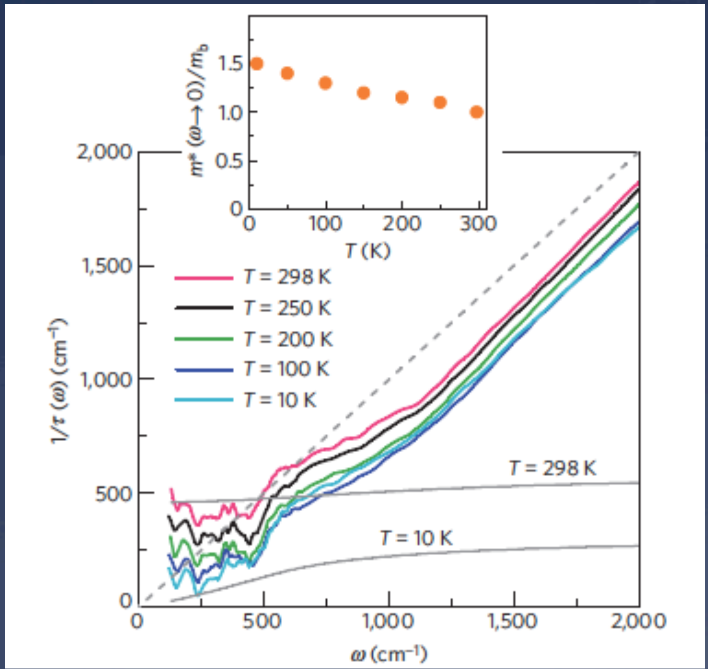
Comments

* So I'm glad that we now agree that intraband is only below ~1500 cm^{-1}



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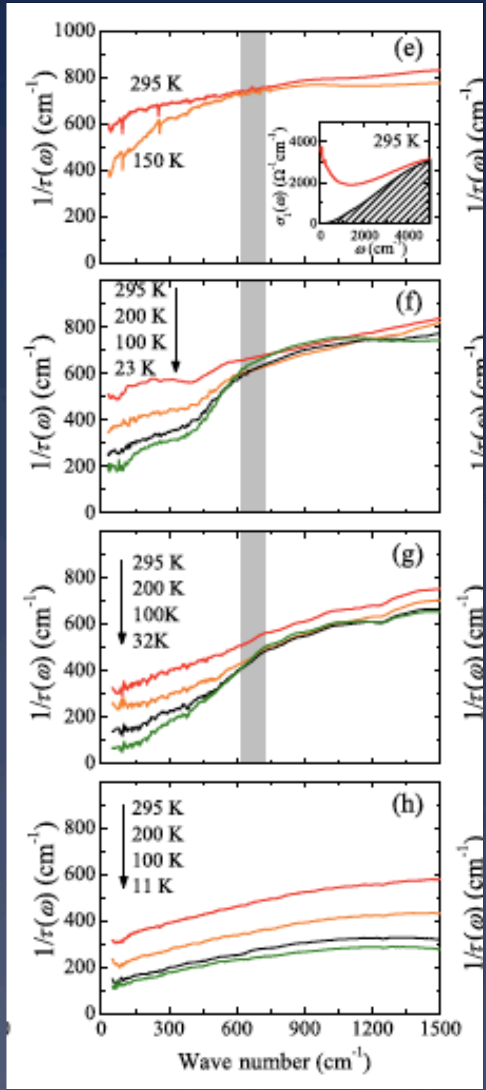
2009

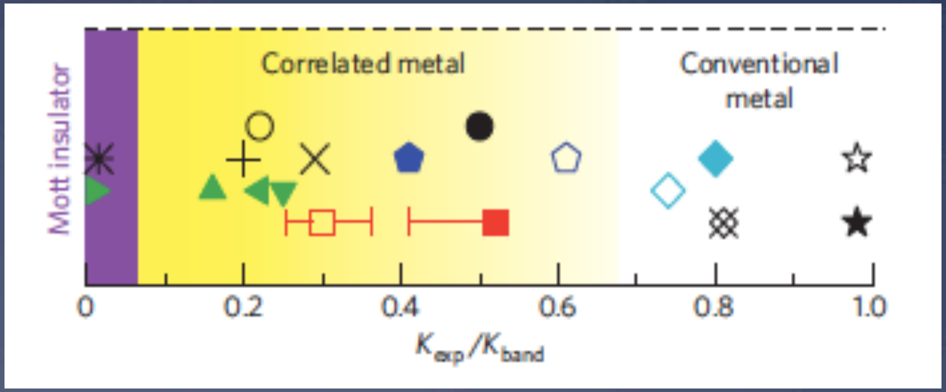


S.J.Moon.PRL 109, 027006 (2012)

2012

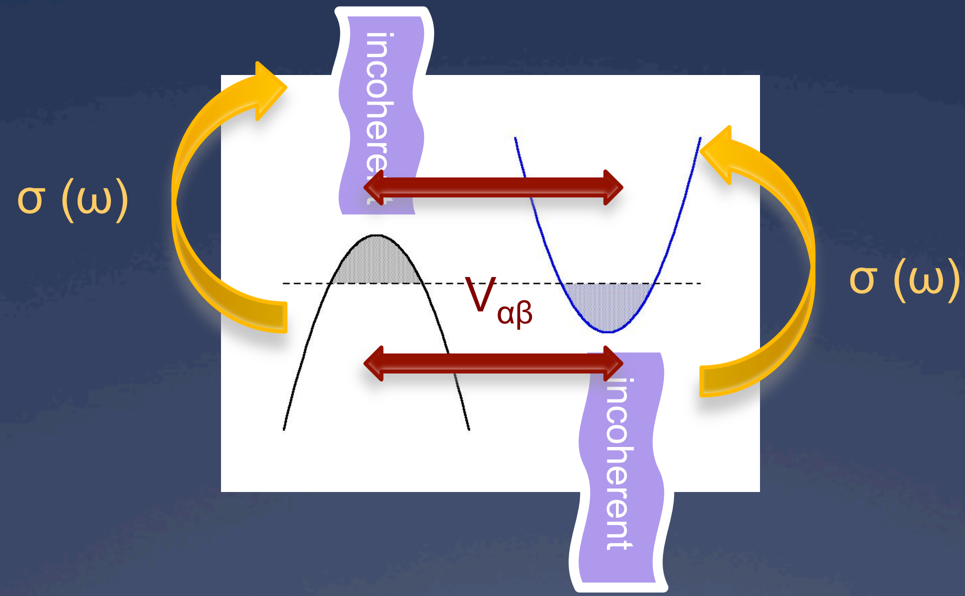
Comments

- So I'm glad that we now agree that intraband is only below ~1500 cm^{-1}
- Including some interband transition in the intraband part does not change considerably the sum-rule analysis, it affects mainly the extended-Drude model analysis



M. M. Qazilbash et al. Nat. Phys. 5, 647 (2009)

* The anomalous sum rule is crucially related to the mixing between hole and electron character induced by interband interactions



* An other interesting effect occurs in the Hall conductivity

(see poster by Laura Fanfarillo)

Hall conductivity

* Hall coefficient in an interacting multiband system

$$R_H = \frac{\sum_i \sigma^i_{xy}}{(\sum_i \sigma^i_{xx})^2 H_z}.$$

* Conserving approximation: the current J differs from the band velocity v due to interactions

$$\sigma^\alpha_{xx} \sim \mathbf{J}^\alpha \cdot \mathbf{v}^\alpha \tau^\alpha$$

$$\frac{\sigma^\alpha_{xy}}{H} \sim \mp(|J^\alpha|\tau^\alpha)^2$$

* How to do? We need to compute explicitly vertex corrections

Vertex corrections in a nutshell

- Usual single-band case: vertex corrections lead to a **transport scattering time** τ_{tr} different from the **quasiparticle lifetime** τ (Boltzmann approach)

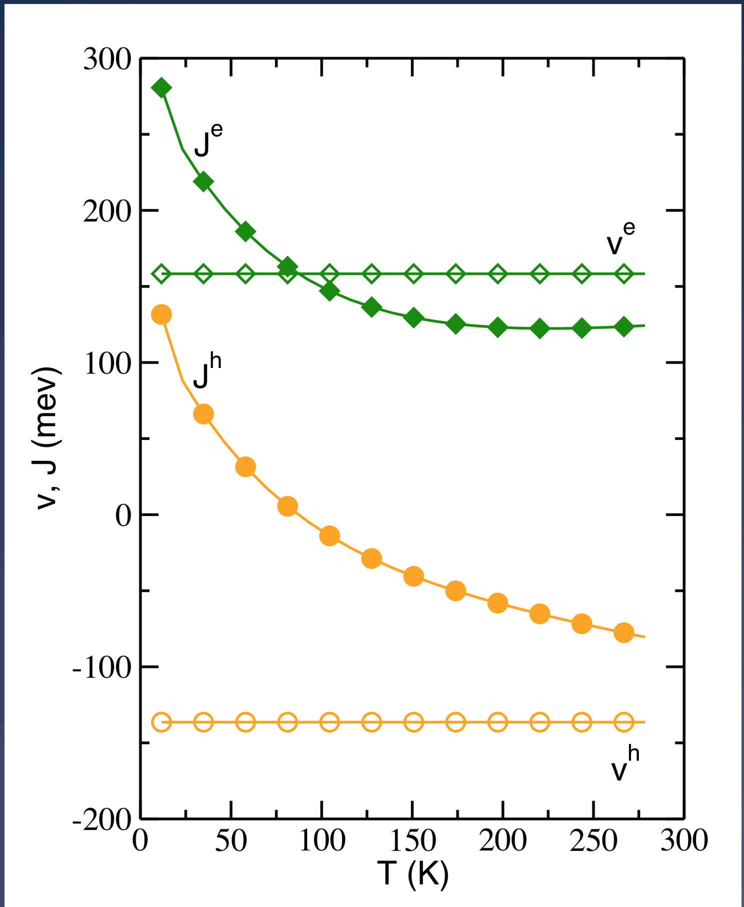
$$\mathbf{J} = \Lambda \mathbf{v} \qquad \sigma_{xx} \sim \mathbf{v} \cdot \mathbf{J}\, \tau = \mathbf{v}^2 \, (\Lambda \tau) = \mathbf{v}^2 \, \tau_{\mathrm{tr}}$$

- However when hole and electron are mixed J can even have different sign from v and in a multiband case

$$\mathbf{J}^e \sim \Lambda_{ee} \mathbf{v}^e + \Lambda_{eh} \mathbf{v}^h, \quad \mathbf{v}^e > 0, \, \mathbf{v}^h < 0$$

- Vertex corrections **cannot** be recast in a re-definition of the transport scattering time, Boltzmann-like picture fails

R_H in a slightly e-doped compound



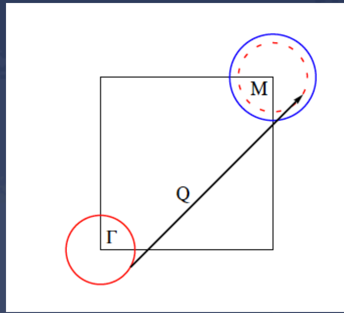
- One e-like and one h-like band
- It is crucial to retain the momentum dependence of the spin fuctuations

$$\chi(\mathbf{q},\omega) = \frac{\chi_Q}{1+\xi_T^2(\mathbf{q}-\mathbf{Q})^2 + i\omega/\omega_{sf}},$$

- Slightly e-doped case $v_e > |v_h|$

$$J^h = \frac{v^h + \lambda_{he}\, v^e}{1-\lambda_{he}\lambda_{eh}}$$

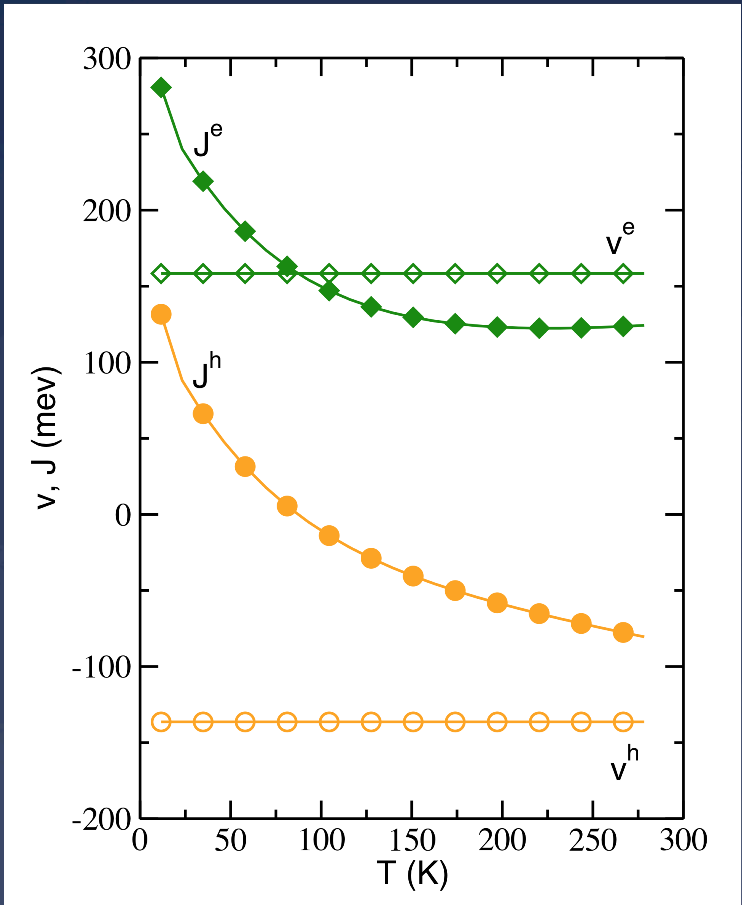
$$J^e = \frac{v^e + \lambda_{eh}\, v^h}{1-\lambda_{he}\lambda_{eh}}$$

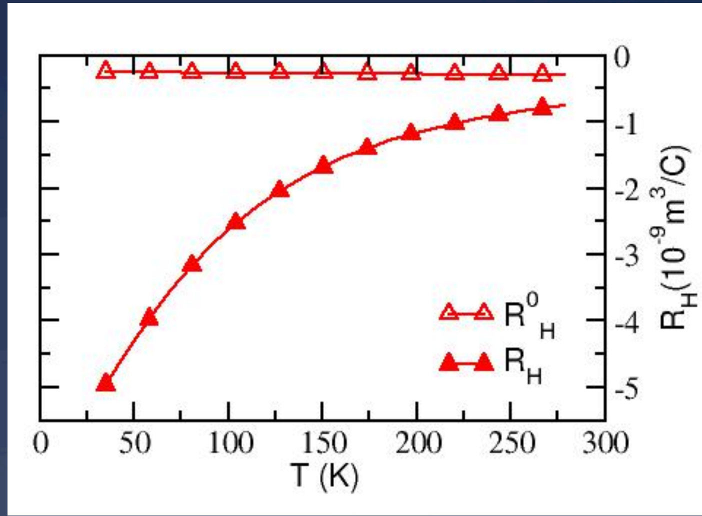


- The λ coefficients increase as T decrease due to larger SF

$$|J^e| \gg |J^h|$$

R_H in a slightly e-doped compound





$$\sigma^e_{xy} \sim -|J^e|^2$$
$$\sigma^h_{xy} \sim |J^h|^2$$

$$|J^e| \gg |J^h| \Rightarrow R_H << 0$$

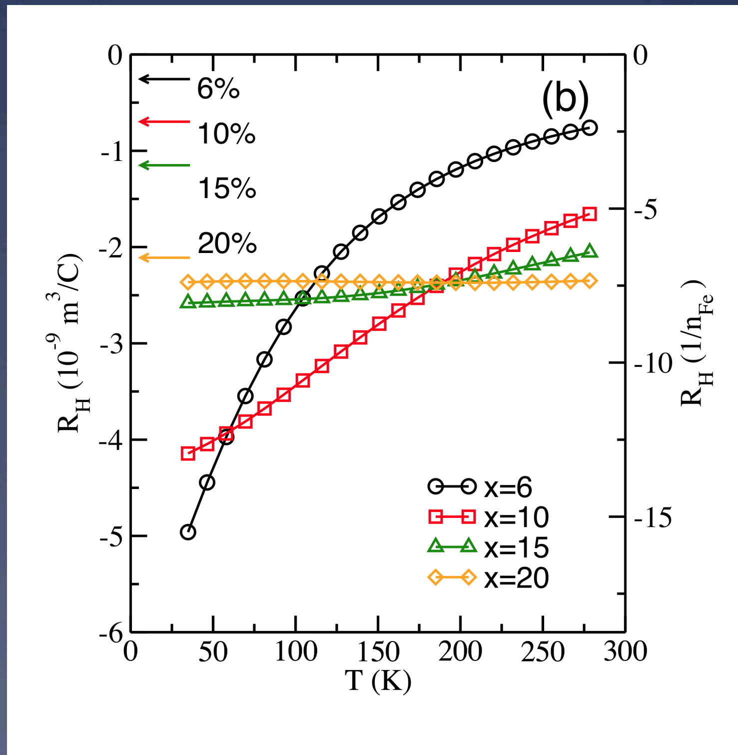
L. Fanfarillo et al. arXiv:1205.2242, PRL (2012)

A single mechanism for the whole phase diagram

* In e-doped J_e >> $|J_h|$ ➡ negative R_H
* In h-doped $|J_h|$>>J_e ➡ positive R_H

L. Fanfarillo et al. arXiv:1205.2242, PRL (2012)

* When doping increases and one moves away from nesting spin fluctuations are less effective, J≈v and one approaches the Boltzmann result



Take-home message for pnictides

**More
(interacting
electron-hole bands)
is different!**

- Interaction-induced Fermi-surface shrinking with respect to DFT
- Large transfer of optical spectral weight to incoherent states and anomalous temperature dependence of the sum rule
- The extended Drude model analysis can be dangerous since low-energy interband transition are very likely to be present
- Anomalous Hall effect due to large vertex corrections

Conclusions part I and part II

* It is not so easy to make calculations, especially if one wants to describe at the same time optical spectra and the corresponding optical sum rule

* Still, it is worth doing that, and we learned a lot in the last ten years of intense work on correlated systems

* Optical conductivity contains several information about the system, but we should not be paradigmatic in reading them out (i.e. by using only our pet model)

* (seventh Matthia's rule) … stay (*away??*) in contact with experimentalists

Acknowledgments and References

* Sum rule:

  Emmanuele Cappelluti (ISC-CNR)

  L. Benfatto and E. Cappelluti, PRB 83, 104516 (2011)

* Fermi-surface shrinking and interband transitions

  L. Pietronero (Physics Dep. Sapienza), L. Ortenzi (MPI Stuttgart), L. Boeri (MPI Stuttgart)

  L.Ortenzi, E. Cappelluti, L. Benfatto and L. Pietronero. Phys Rev Lett 103, 046404 (2009)

  L. Benfatto, E. Cappelluti, L. Ortenzi and L. Boeri, PRB 83, 224514 (11)