Conductivité Optique

Ricardo Lobo

École Supérieure de Physique et Chimie Industrielles
de la Ville de Paris

lobo@espci.fr

Volume I. *July–August, 1893.* *Number 1.*

THE

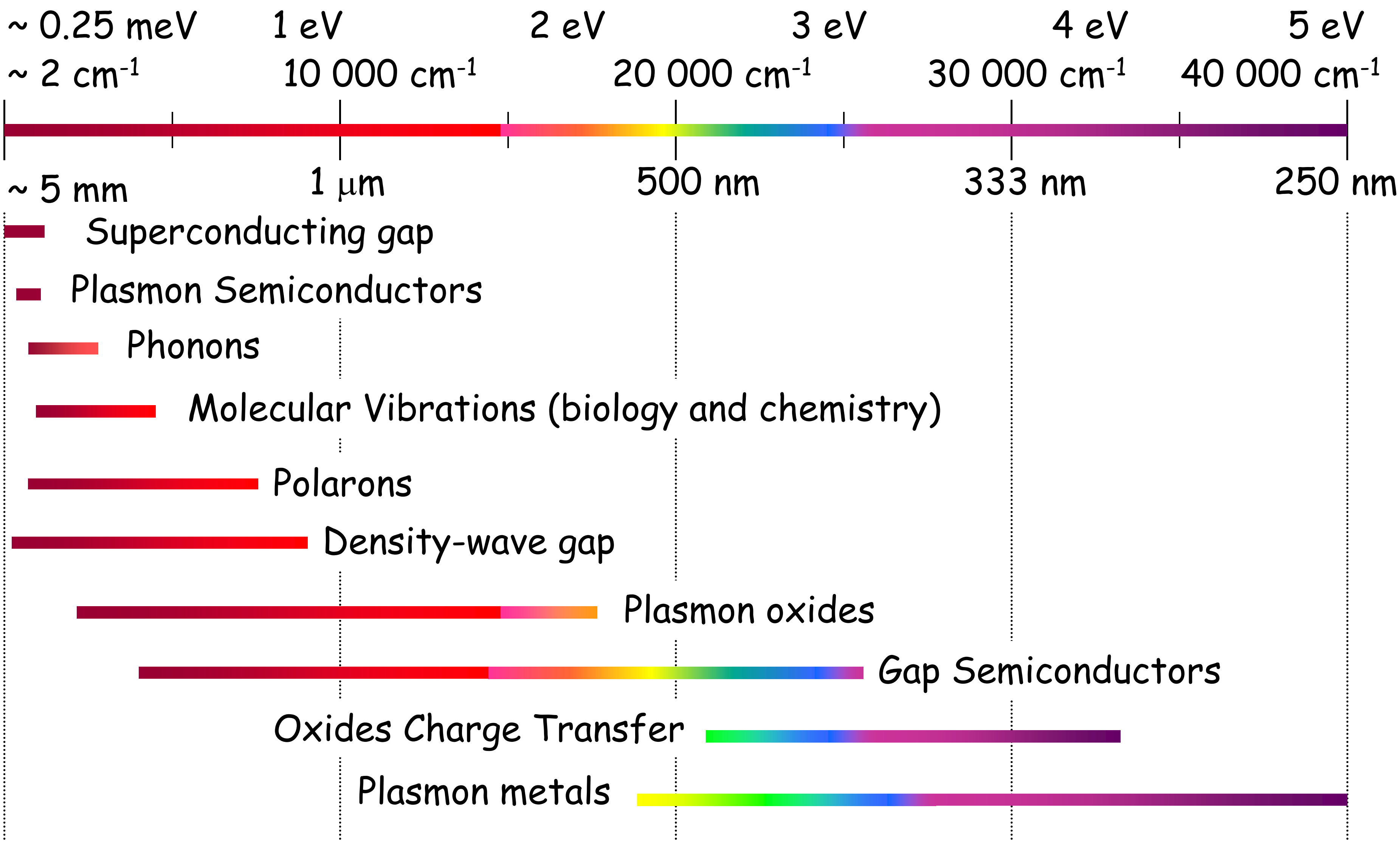
PHYSICAL REVIEW.

A STUDY OF THE TRANSMISSION SPECTRA OF CERTAIN SUBSTANCES IN THE INFRA-RED.

BY ERNEST F. NICHOLS.

Nichols, Phys. Rev. 1, 1 (1893).

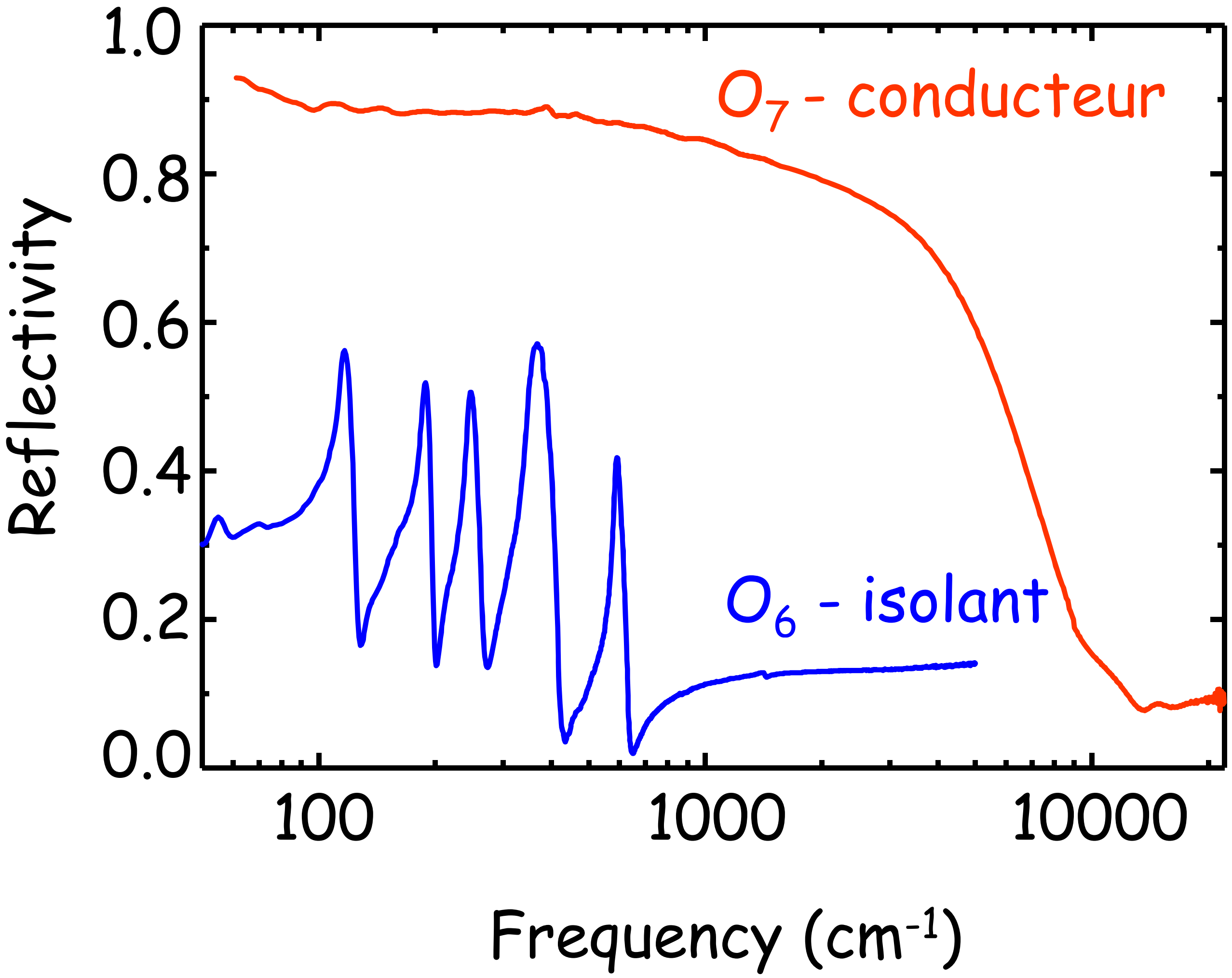
A few optical excitations in solids



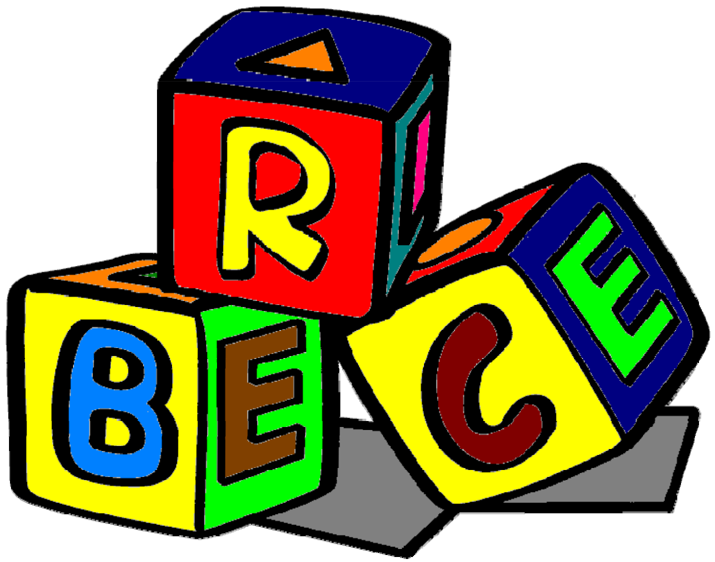
Outline

- ✓The basics of optical conductivity
- ✓The optical conductivity of metals
- ✓The f-sum rule and gaps
- ✓Fermi liquid signatures
- ✓Kinetic energy
- ✓Phonons and phase transitions
- ✓Disordered Materials

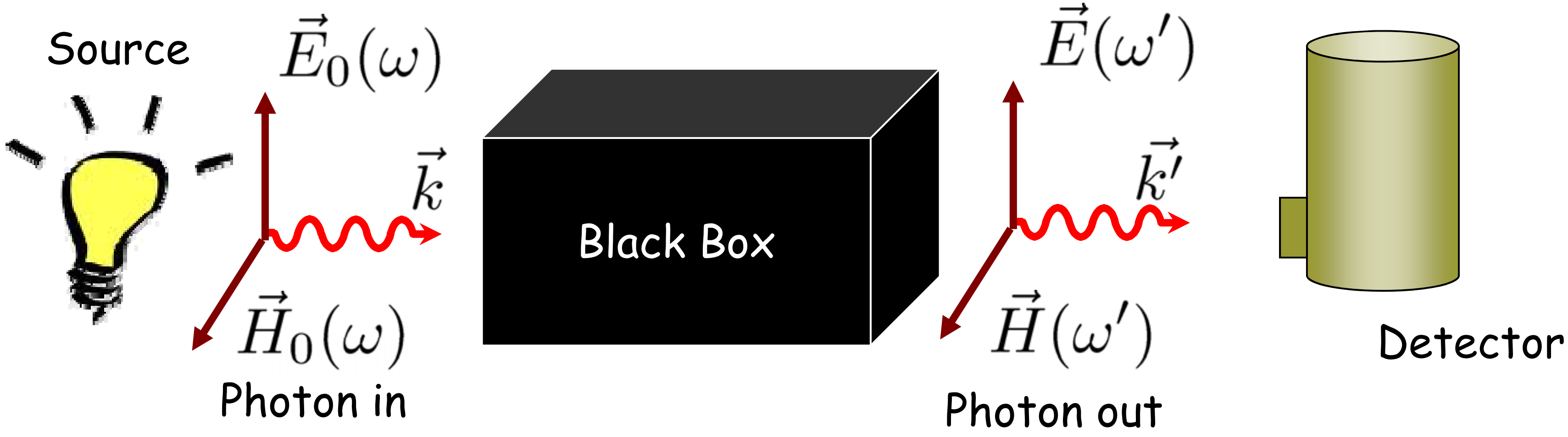
Preview: YBCO from an insulator to a conductor



Optical Conductivity Building Blocs



Light scattering



Optical conductivity

$\vec{k} \approx \vec{k}' \approx 0 \quad \left(\ll \vec{k}_{BZE}\right)$ Hence the misnomer "center of zone" technique

$\Delta\vec{k} \approx 0$ Better name: "momentum averaged" technique

$\omega = \omega'$ Elastic light scattering

$\omega \in [0, \infty]$ Broadband (whitelight) spectroscopy

$\chi_{Elec} \gg \chi_{Mag}$ The interaction with the electric field dominates

Maxwell Equations – The Message in the "Photon Out"

Maxwell Equations in matter

$$\nabla \cdot \vec{D} = \rho$$

$$\nabla \cdot \vec{B} = 0$$

$$\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}$$

$$\nabla \times \vec{B} = \mu_0 \vec{J} + \mu_0 \varepsilon_0 \frac{\partial \vec{E}}{\partial t}$$

Plane wave

$$\vec{E} \sim \vec{B} \sim \exp(-i\omega t)$$

In vacuum

$$\rho = 0 \quad \vec{J} = 0$$

$$c = (\mu_0 \varepsilon_0)^{-1/2}$$

Wave with speed "c"

$$-\nabla^2 \vec{E} - \omega^2 \frac{1}{c^2} \vec{E} = 0$$

In matter

$$\vec{\mathrm{j}}(\omega) = \sigma(\omega) \vec{E}(\omega)$$

Ohm's law

Wave with renormalized speed

$$-\nabla^2 \vec{E} - \omega^2 \frac{1}{c^2} \underbrace{\left[1 + \frac{i\sigma(\omega)}{\varepsilon_0 \omega}\right]}_{\varepsilon(\omega) = N(\omega)^2} \vec{E} = 0$$

The common optical functions

Refraction index: light propagation and attenuation

$$N = n + ik$$

Dielectric function: microscopic polarizability

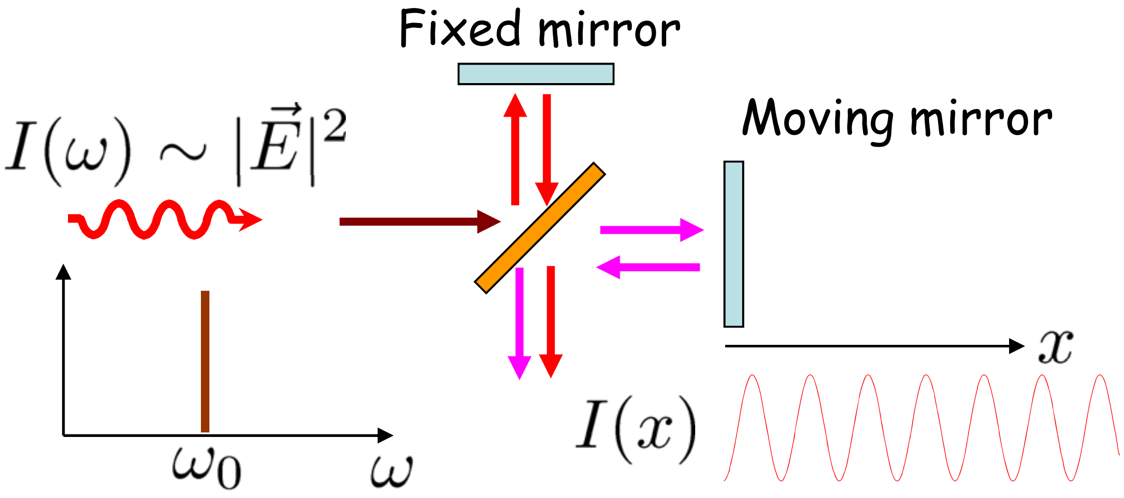
$$\varepsilon = \varepsilon' + i\varepsilon'' = N^2$$

Optical conductivity: high-frequency electrical transport

$$\sigma = \sigma_1 + i\sigma_2 = i\varepsilon_0\omega(1 - \varepsilon)$$

$$\varepsilon_0 = 8.85 \times 10^{-12} \mathrm{As/Vm}$$

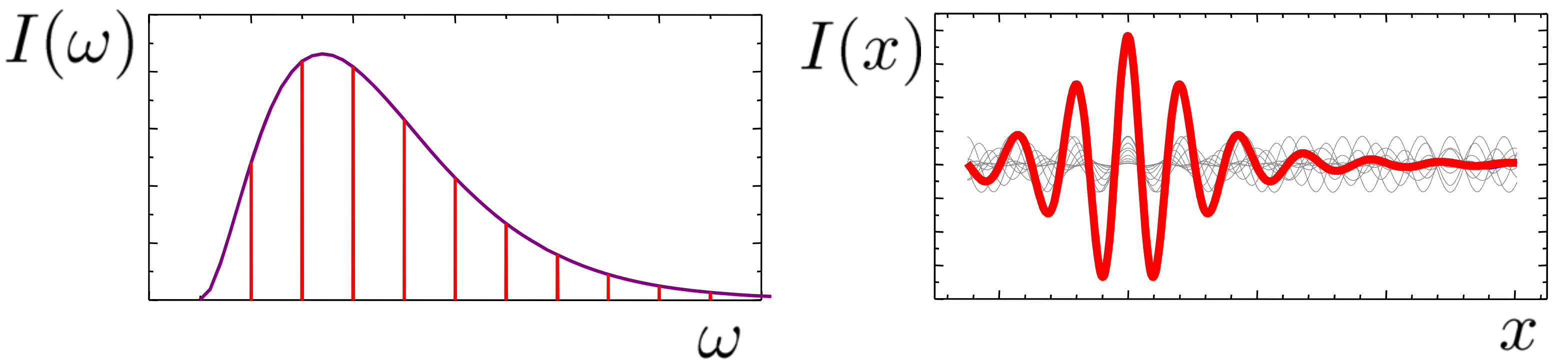
Inside the Black Box – Fourier Transform Spectroscopy



Fellgett advantage
Multiplex

Jacquinot advantage
Slitless spectrometer

Nyquist theorem
Discrete FT is fine

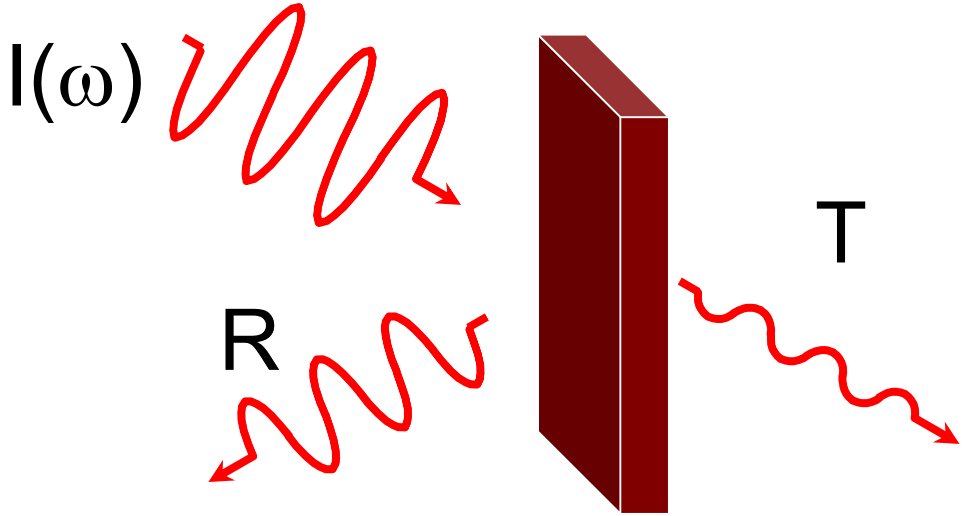


$$I(x) = \mathcal{FT}\left[I(\omega)\right]$$

A Strange Choice for Units

- ✓ Wavenumber $\omega = 1/\lambda = 2\pi c/\varpi \rightarrow$ cm^{-1}
- ✓ Energy $E = h\nu = hc\omega$
  1 eV ~ 8000 cm^{-1}
- ✓ Wavelength $\lambda = 1/\omega$
  1 μm = 10000 cm^{-1}
- ✓ Temperature $T = hc\omega/k_B$
  10000 K ~ 7000 cm^{-1}

What else is in the black box?



R(ω) and T(ω) ~ |E|²
are real functions.

$$R = \left|\frac{E_R}{E_0}\right|^2 = \left|\frac{1-\sqrt{\varepsilon}}{1+\sqrt{\varepsilon}}\right|^2 = \left|\frac{1-N}{1+N}\right|^2$$

Reflected Power (Real)

Kramers-Kronig, simulation; fit; R & T inversion...

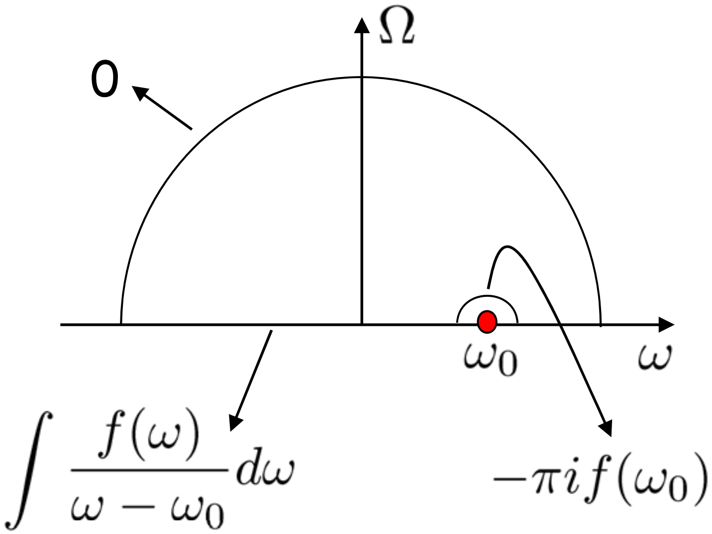
↓

Complex Optical Function

Other techniques using the same process:

- Ellipsometry
- Mach-Zehnder
- Coherent THz

Kramers-Kronig relations & the Reflectivity



Cauchy theorem

$$\oint \frac{f(z=\omega+i\Omega)}{z-\omega_0}dz = 0$$

Hilbert transform

$$f(\omega_0) = -\frac{i}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{f(\omega)}{\omega-\omega_0}d\omega$$

Kramers-Kronig

Causality: $f(\omega) = f^{\star}(-\omega)$ → $\mathrm{Im} f(\omega_0) = -\frac{2\omega_0}{\pi}\mathcal{P}\int_0^{\infty}\frac{\mathrm{Re} f(\omega)}{\omega^2-\omega_0^2}d\omega$

Reflectivity: $R = rr^{\star}$

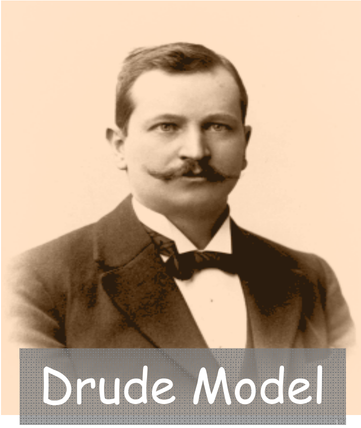
$r = \sqrt{R}e^{i\theta}$ → $\theta(\omega_0) = -\frac{\omega_0}{\pi}\mathcal{P}\int_0^{\infty}\frac{\log R(\omega)}{\omega^2-\omega_0^2}d\omega$

$$\log r = \frac{1}{2}\log R(\omega) + i\theta(\omega)$$

But we need R(ω) at all energies

The optical conductivity in metals

Drude Model



- ✓ Free electron gas
- ✓ No electron-electron interactions
- ✓ No electron-phonon interactions
- ✓ Scattering through elastic collisions

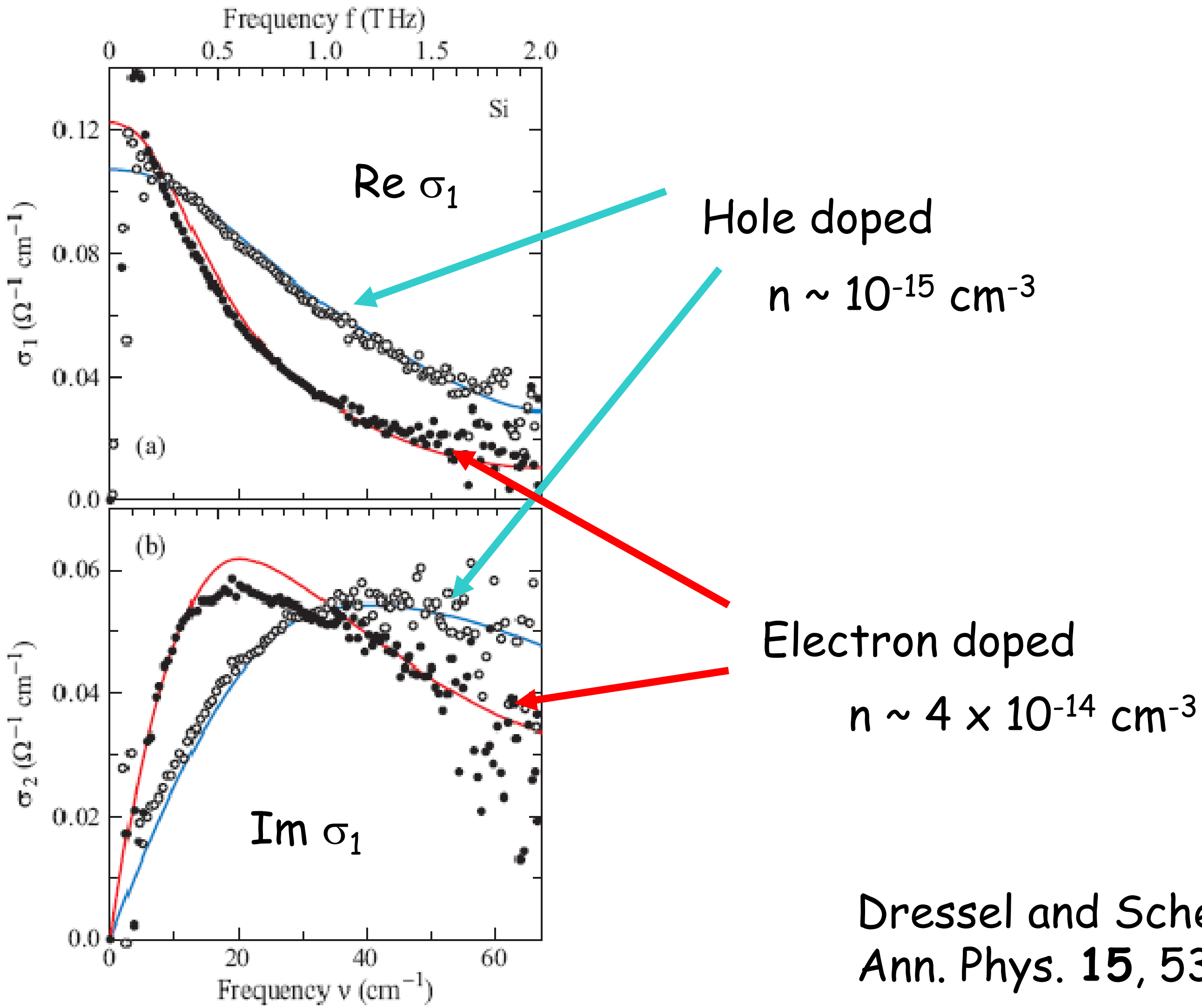
Drude, Ann. Physik **306**, 566 (1900)

$$\sigma(\omega) = \frac{2\pi}{Z_0} \frac{\Omega_p^2}{\tau^{-1} - i\omega} = \frac{\sigma_0}{1 - i\omega\tau}$$

$$\Omega_p^2 = \frac{Z_0}{2\pi} \frac{ne^2}{m}$$

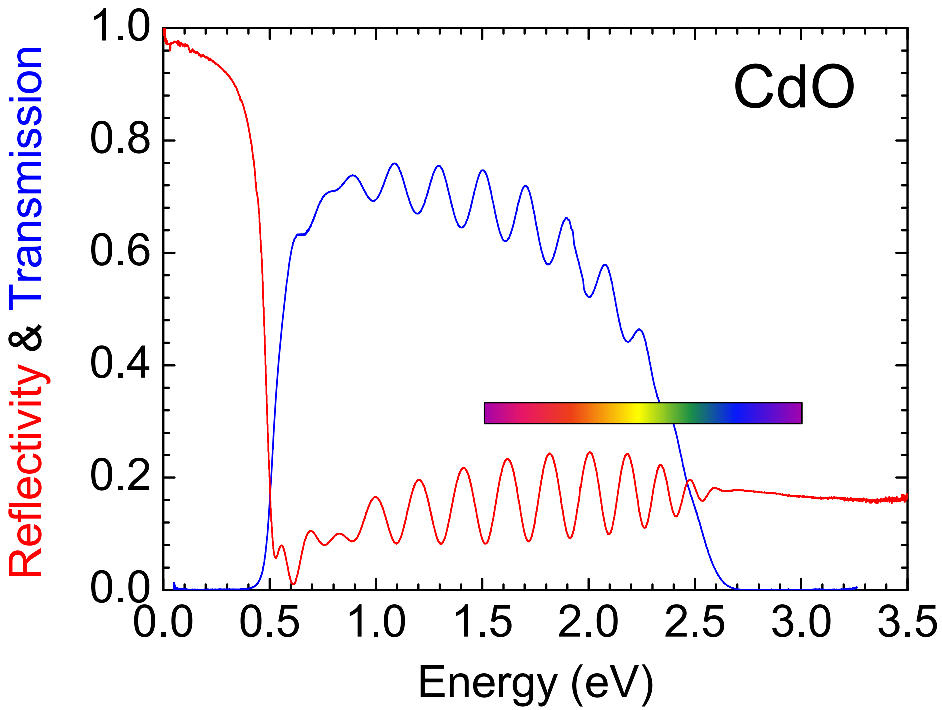
$$\sigma_0 = \frac{ne^2\tau}{m}$$

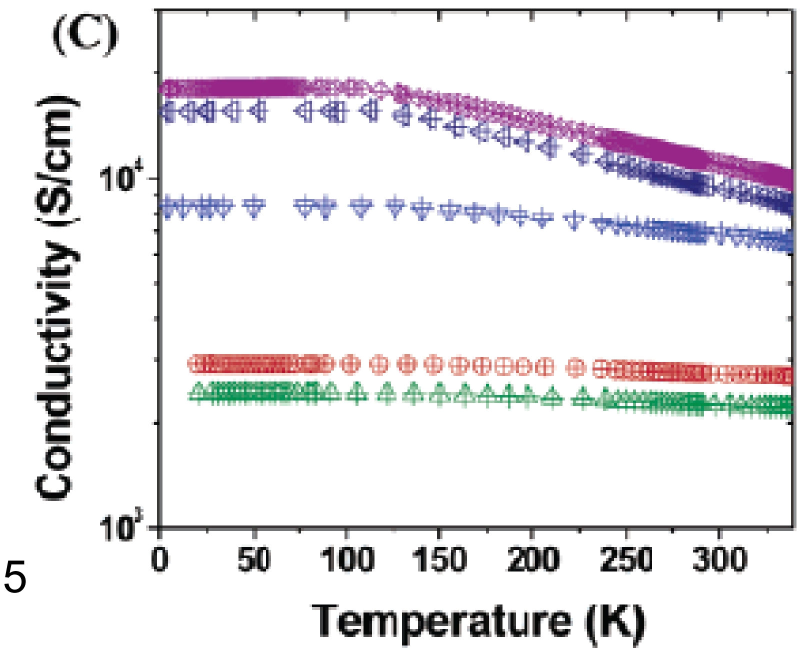
Drude in lightly doped Si



Dressel and Scheffler,
Ann. Phys. **15**, 535 (2006)

Transparent conducting oxides





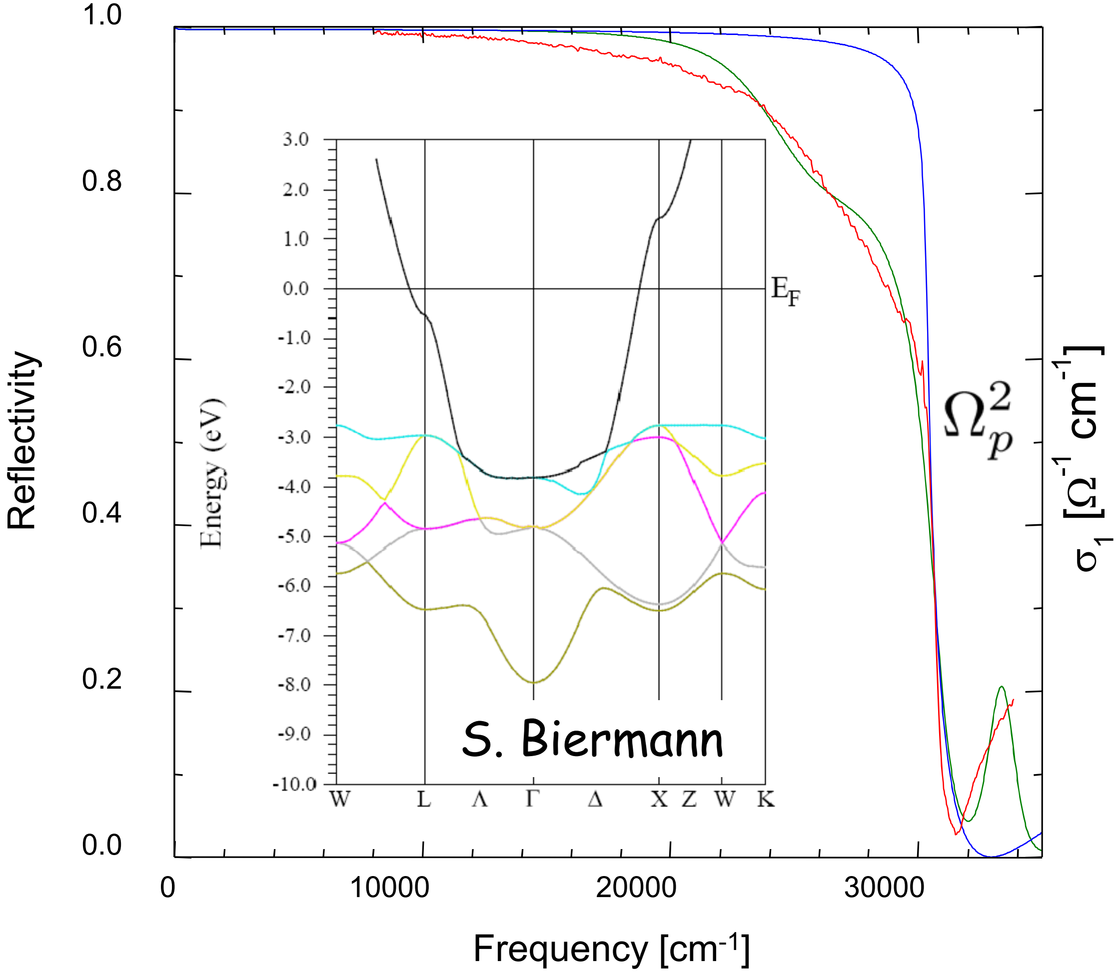
$$\Omega_p^2 = \frac{Z_0}{2\pi}\frac{ne^2}{m}$$

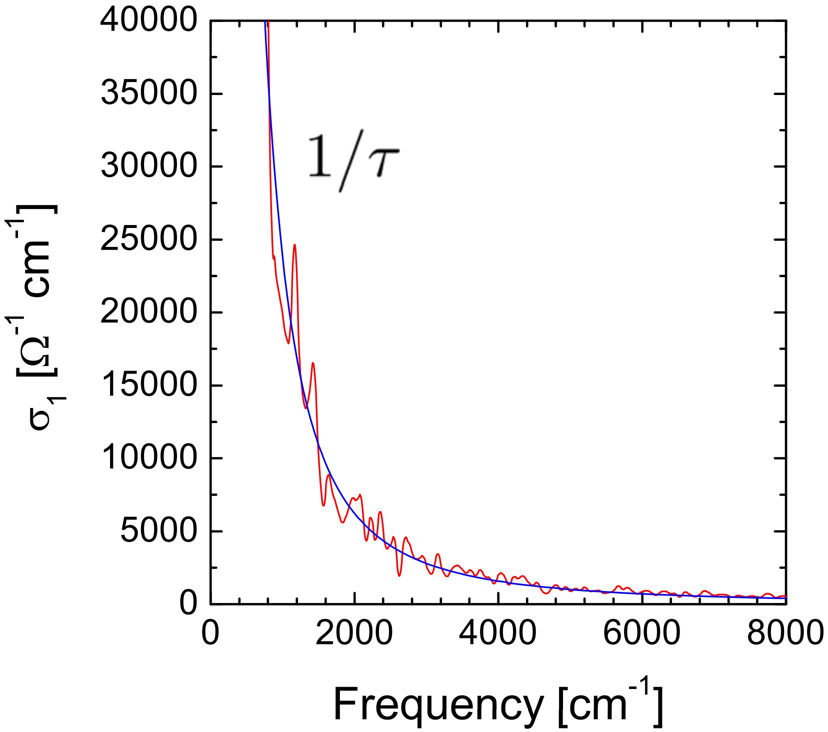
- ✓Keep n small (tweak materials)
- ✓Increase τ (improve materials)
- ✓Play with m* (create materials)

$$\sigma_0 = \frac{ne^2\tau}{m}$$

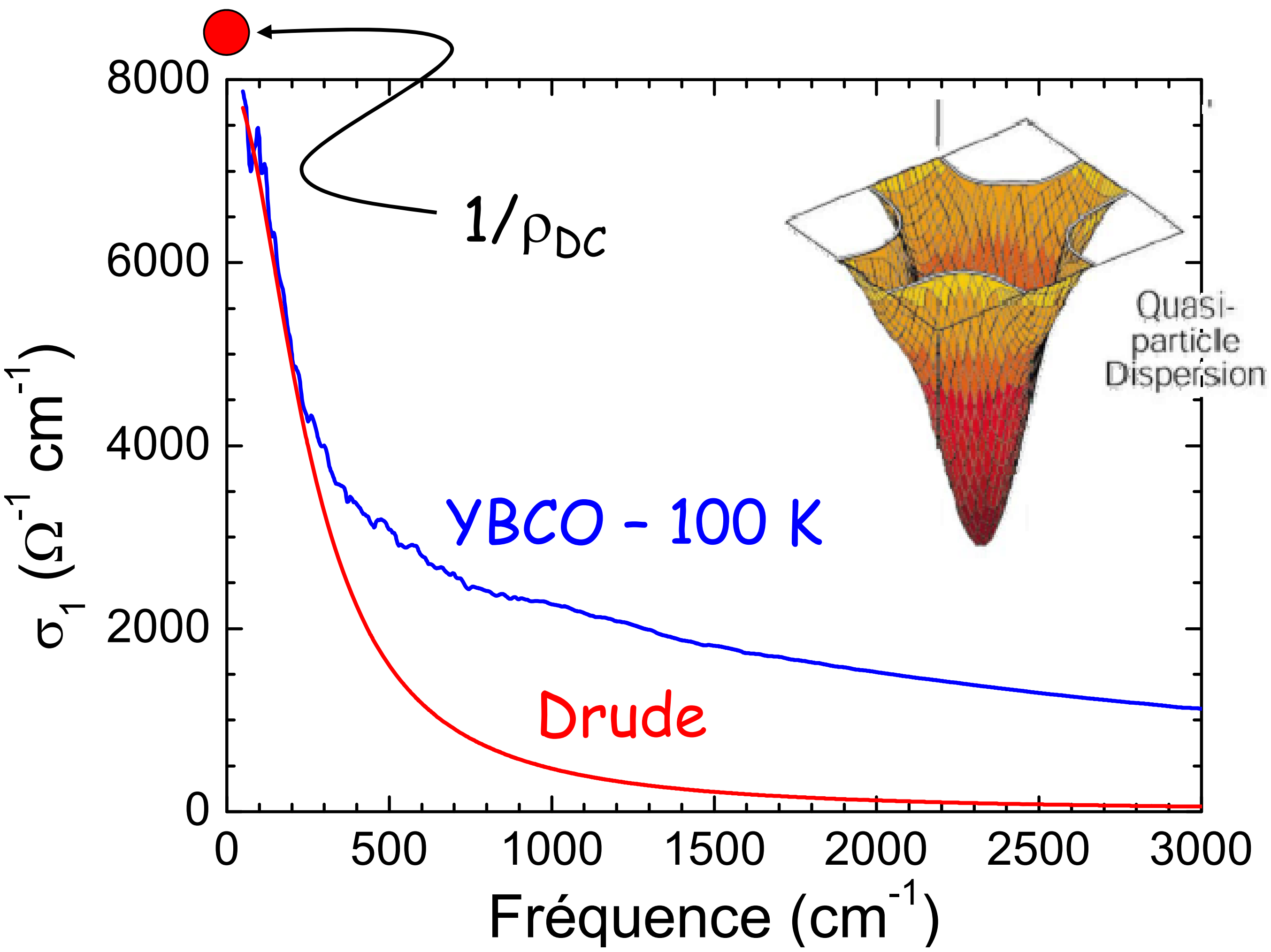
Metz et al., JACS 126, 8477 (2004)

Drude in metals (Silver)

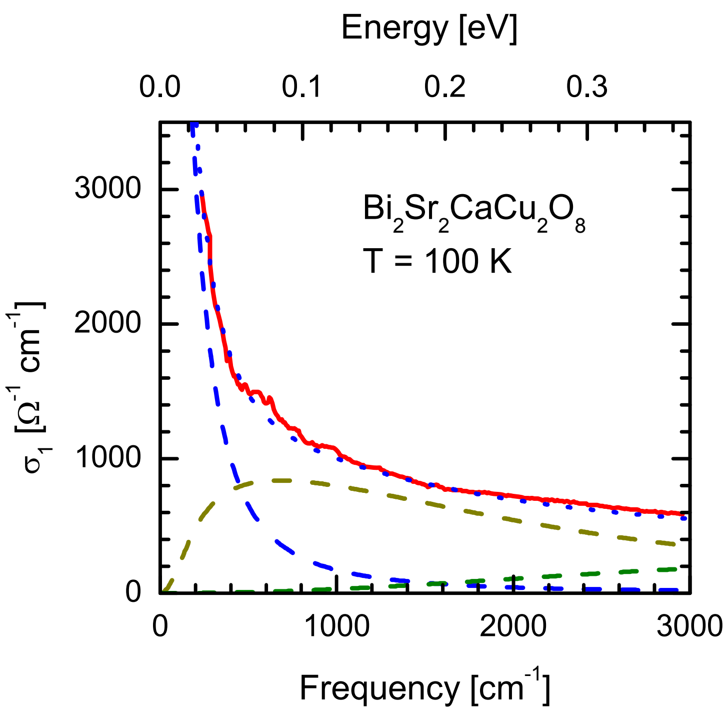


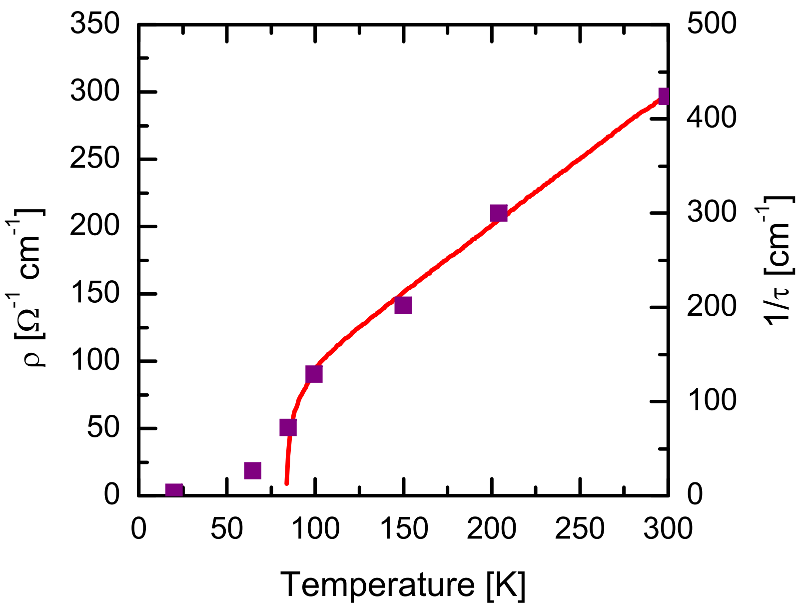


But Drude fails in correlated matter…



Ignoring the elephant



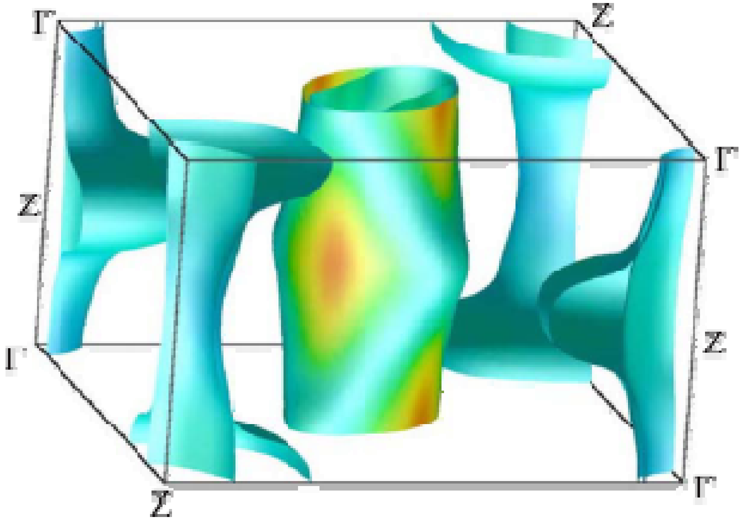


- ✓ Coherent peak described by Drude
- ✓ Mid-IR response simulated as Lorentz oscillators

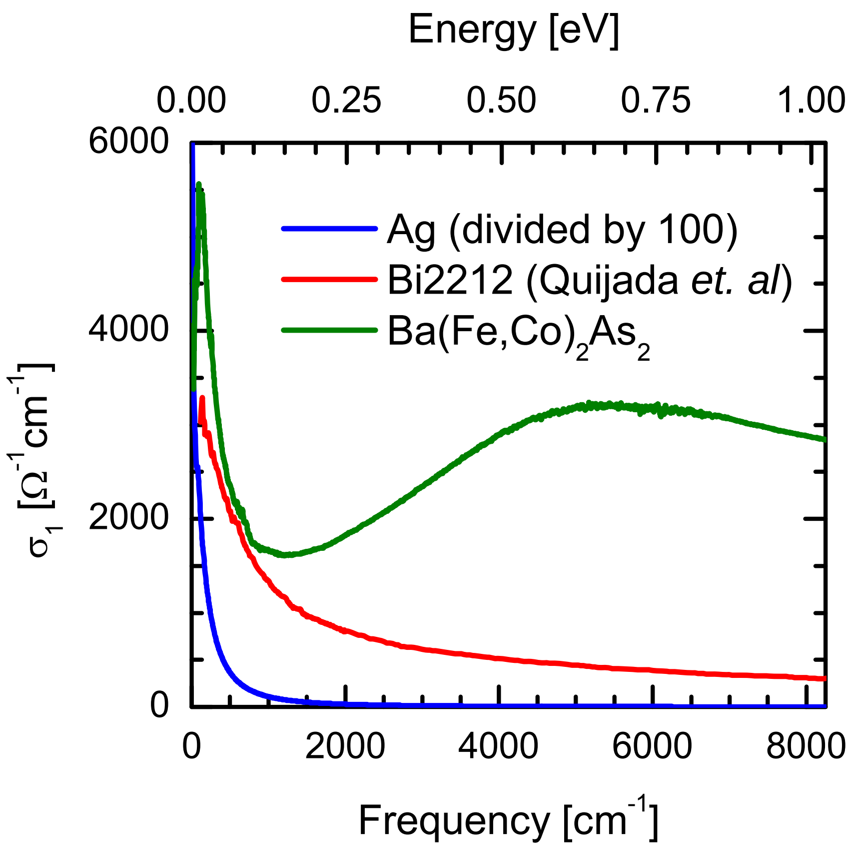
The Drude temperature dependent scattering rate follows the resistivity

Quijada *et al.*, PRB 60, 14917 (1999)

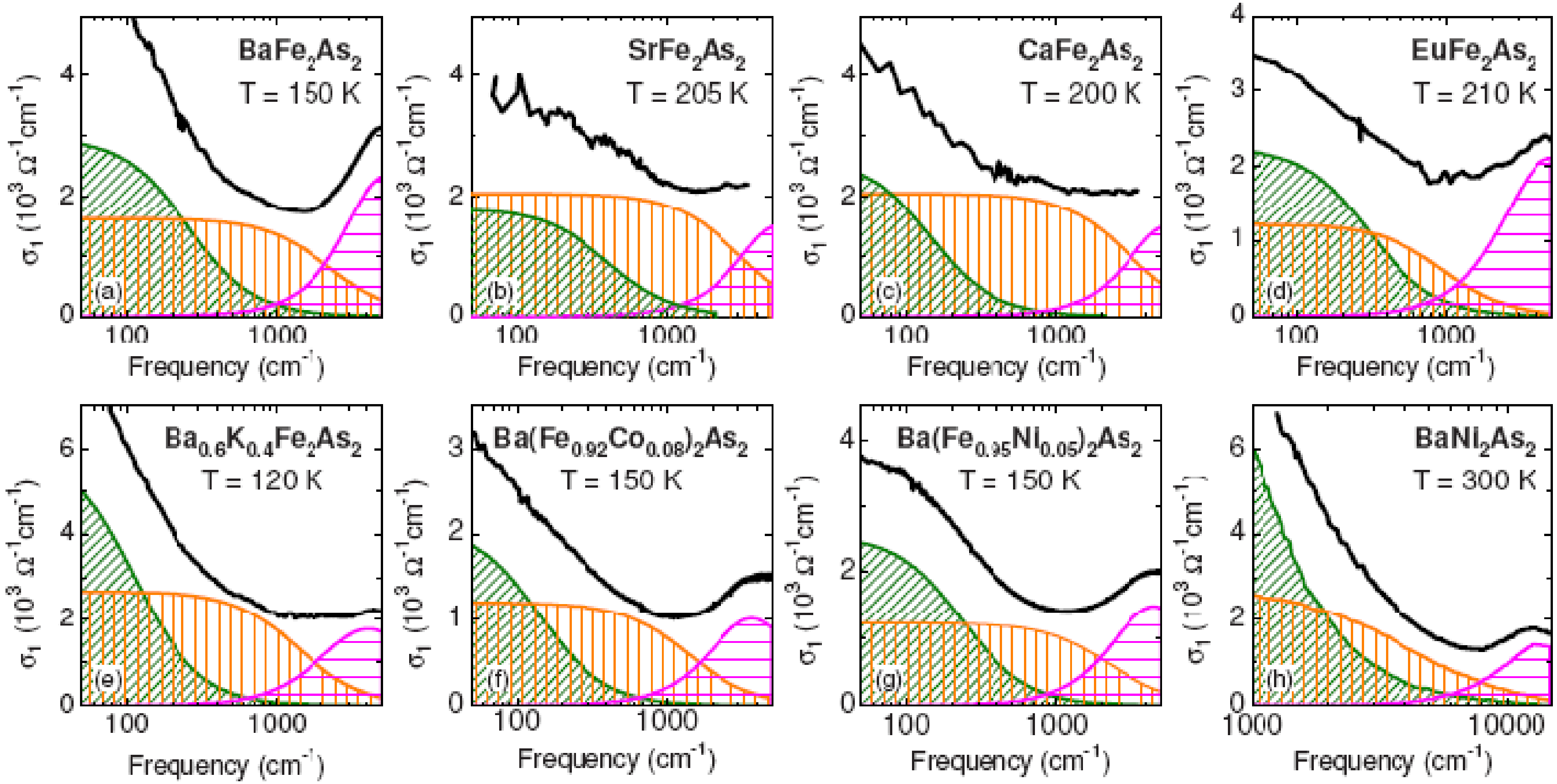
The optical conductivity and multiband pnictides



Singh, PRB (2008).



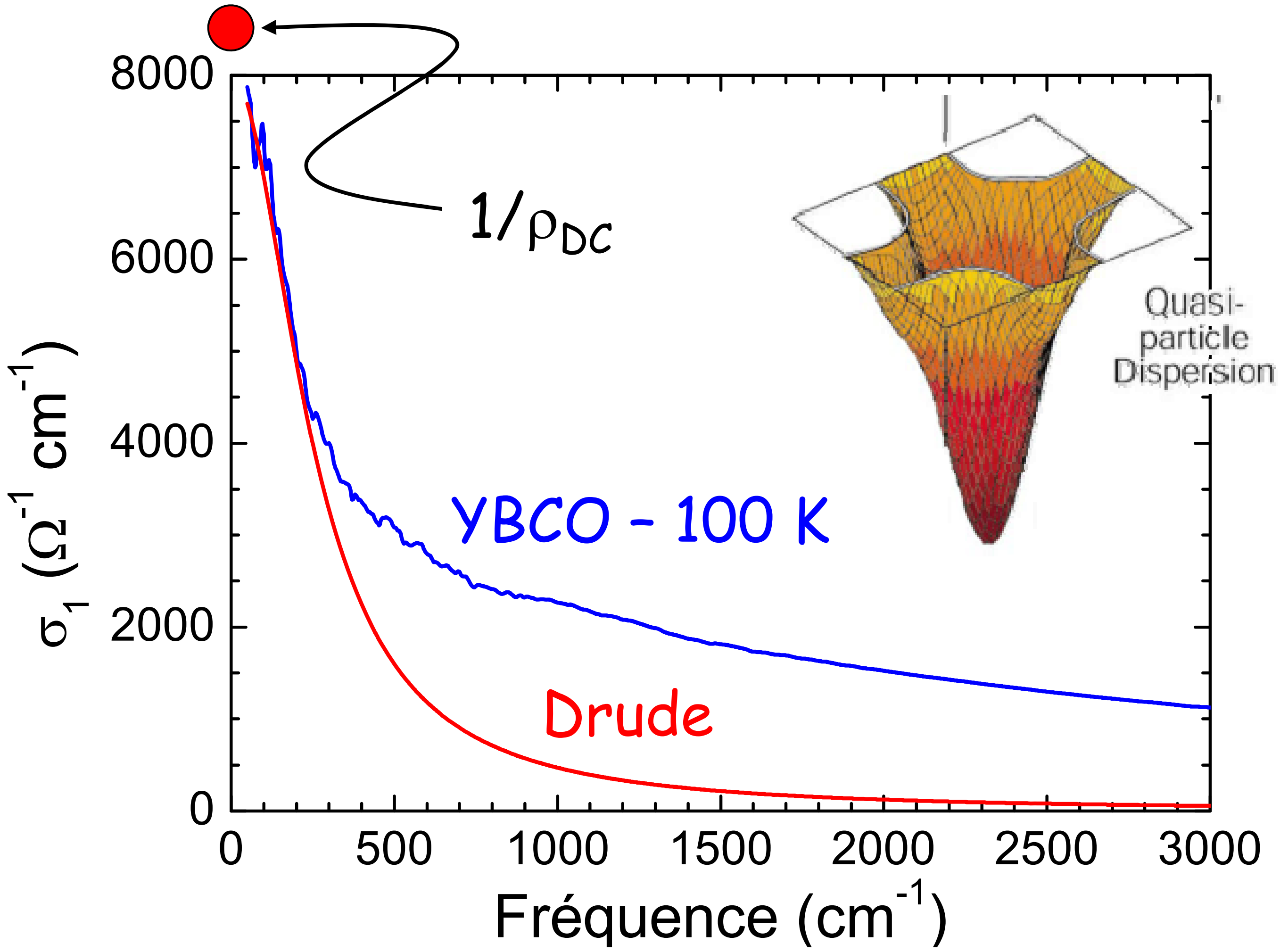
Multiband – pnictides Dressel



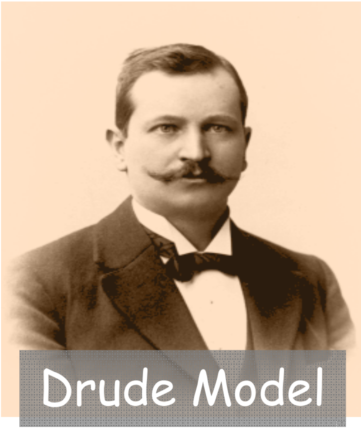
Using two Drude terms [two bands (??)] solves the problems

Wu et al., PRB **81**, 100512 (2010)

Can we take a look at the elephant?

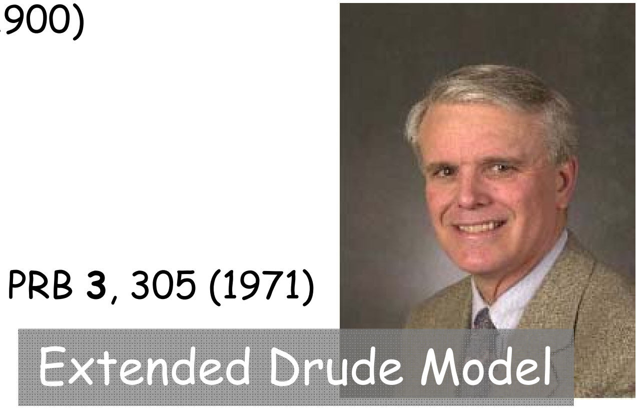


Patching the Drude Model



Drude, Ann. Physik **306**, 566 (1900)

Allen, PRB **3**, 305 (1971)



$$\sigma(\omega) = \frac{2\pi}{Z_0} \frac{\Omega_p^2}{\tau^{-1} - i\omega}$$

$$\sigma(\omega) = \frac{2\pi}{Z_0} \frac{\Omega_p^2}{\tau^{-1}(\omega) - i\omega\left[1 + \lambda(\omega)\right]}$$

Scattering rate

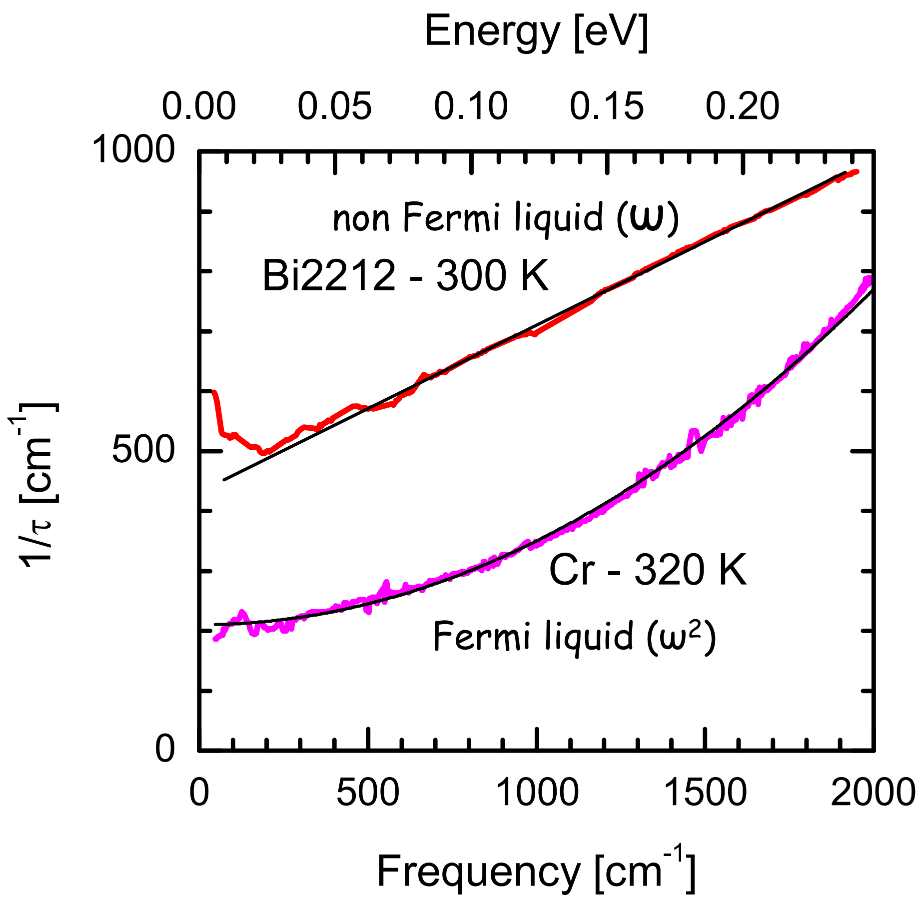
Mass enhancement

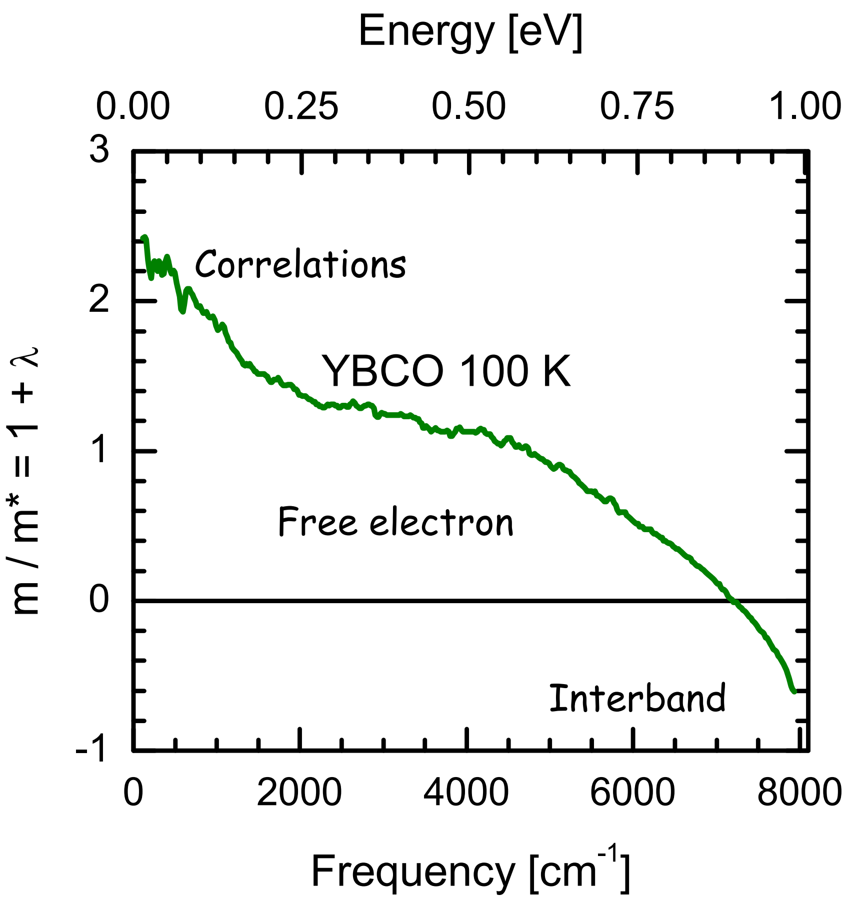
$$\frac{1}{\tau(\omega)} = \frac{2\pi}{Z_0}\Omega_p^2 \,\mathrm{Re}\left(\frac{1}{\sigma(\omega)}\right)$$

$$\frac{m}{m^\star} = 1 + \lambda(\omega) = \frac{1}{\omega}\frac{2\pi}{Z_0}\Omega_p^2 \,\mathrm{Im}\left(\frac{1}{\sigma(\omega)}\right)$$

"Optical self-energy"

Is this extended Drude mambo-jambo of any use?

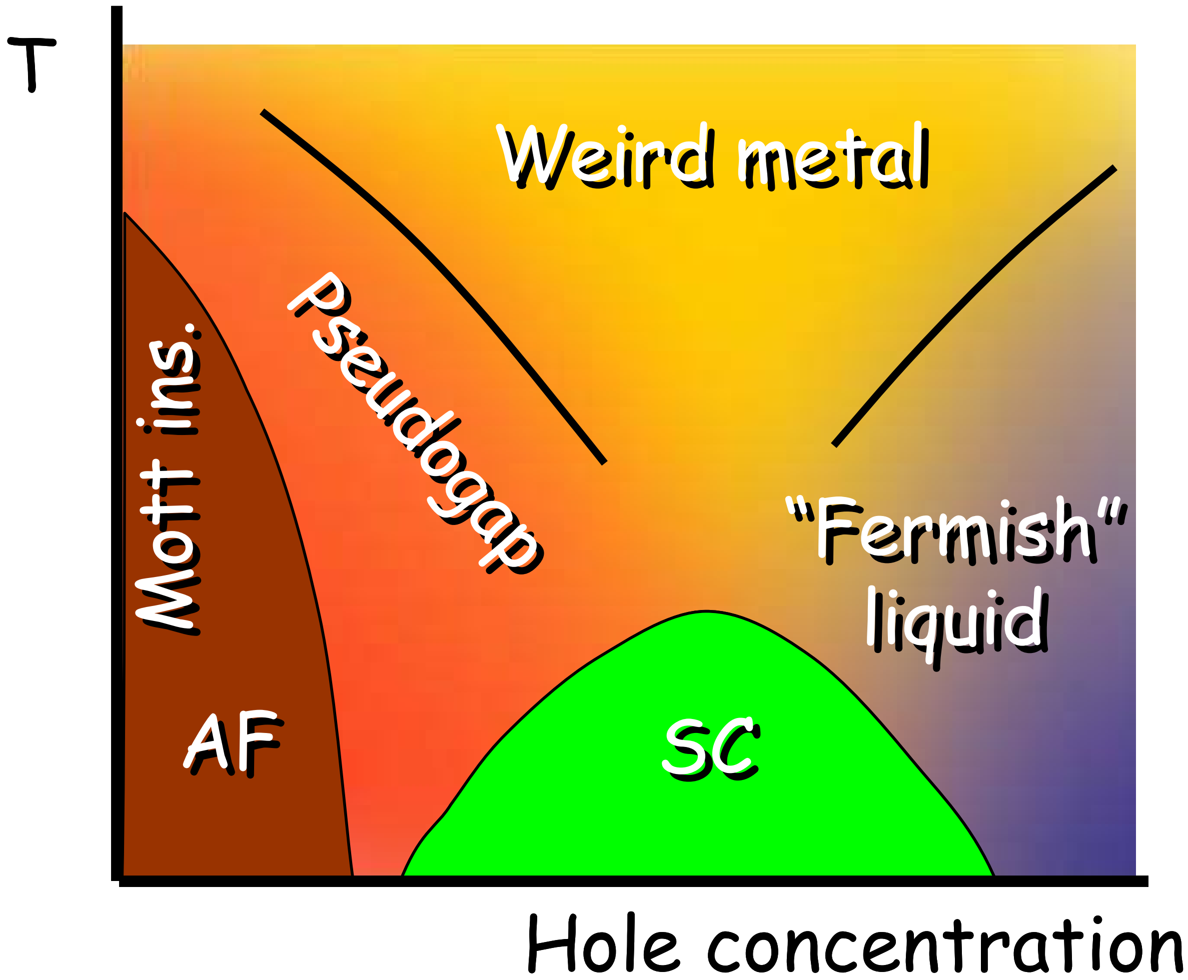




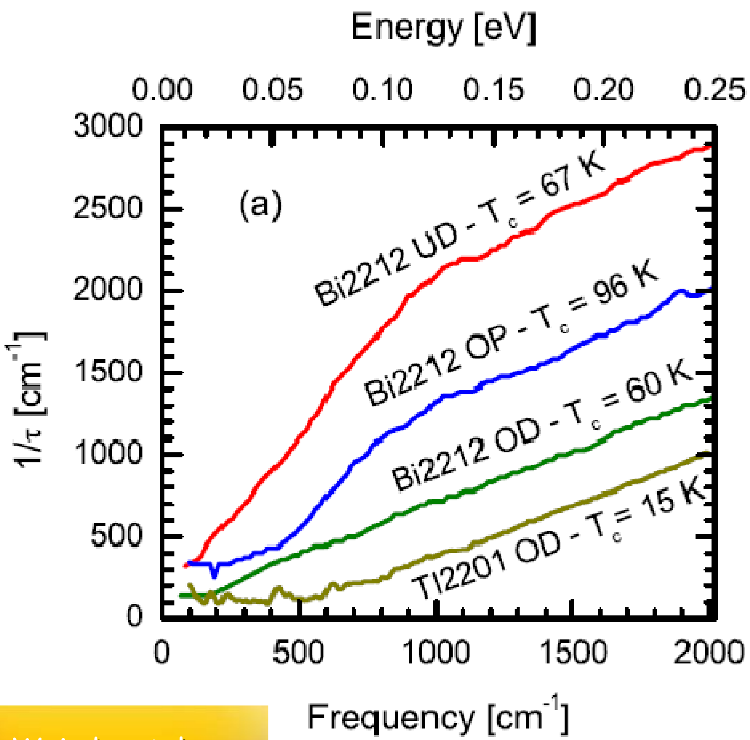
Basov *et al.*, PRB **65**, 054516 (2002)

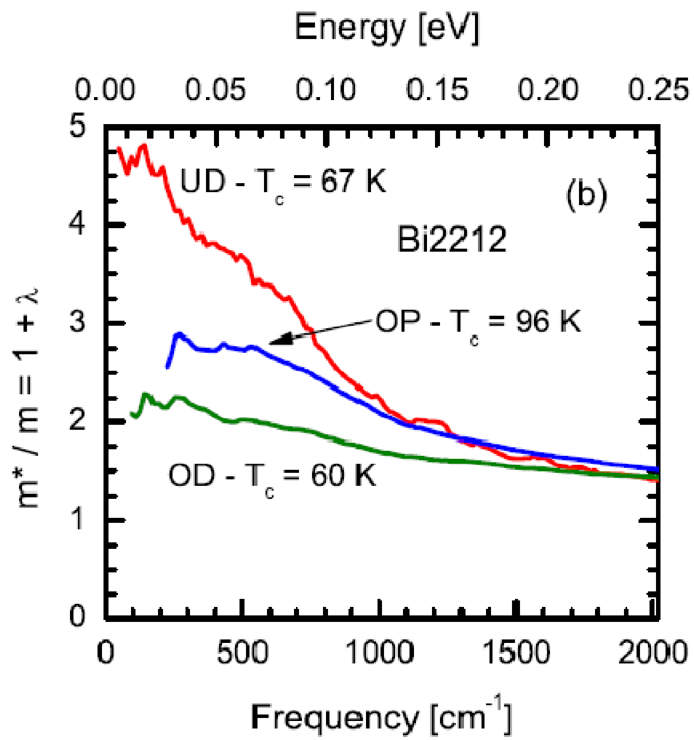
Puchkov *et al.*, JPCM **8**, 10049 (1996)

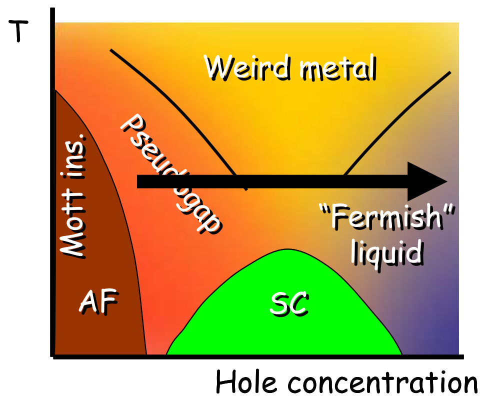
One of the multiple versions of the cuprates phase diagram



From a Nodal Metal to a Bad Metal to a Fermi Liquid







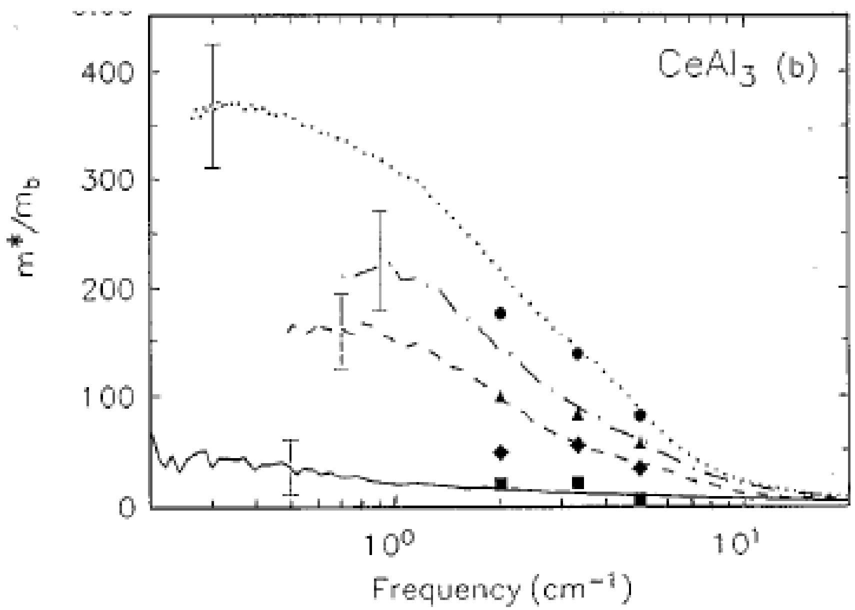
- ✓ $1/\tau$: Fermi liquidness increases with doping
- ✓ m*: Correlations decrease with increasing doping

Hwang, Nature 427, 714 (2004)
Ma, PRB 73, 144503 (2006).

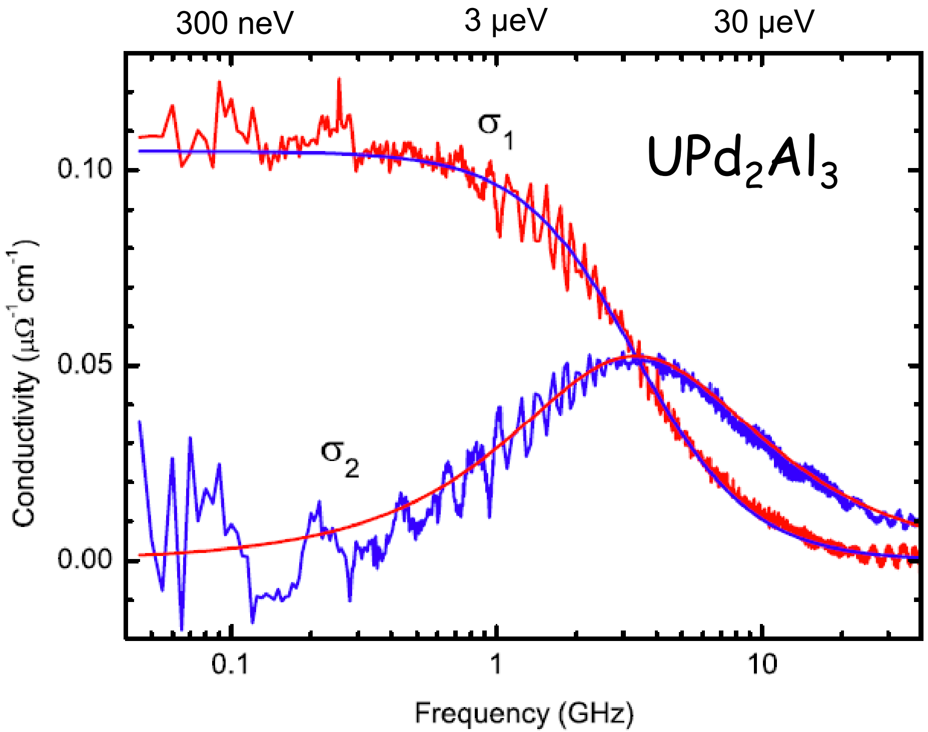
Heavy Fermions and LF Drude

$$m^\star \sim 20 - 100m$$

$$\frac{1}{\tau^\star} = \frac{m}{m^\star}\frac{1}{\tau}$$



Awasthiet al. PRB **48**, 10692 (1993).

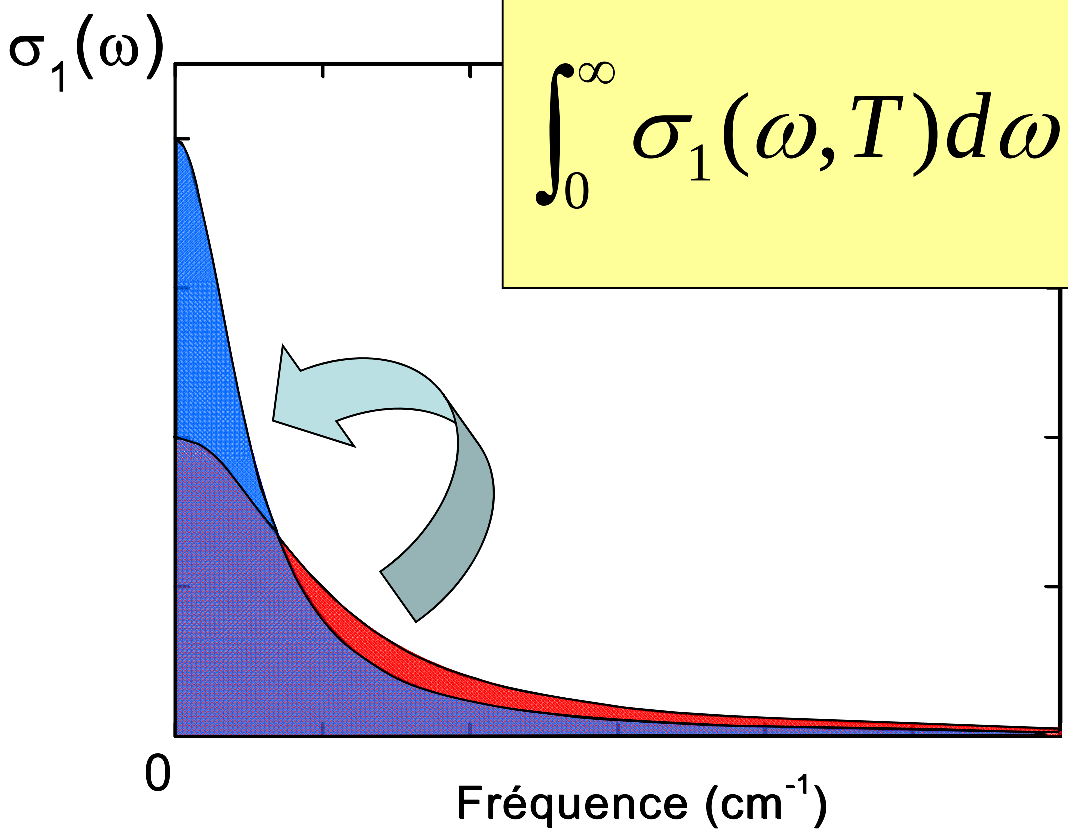


Dressel and Scheffler, Ann. Phys. **15**, 535 (2006)

Mass enhancement leads to scattering rate decrease

Gaps and the sum rule

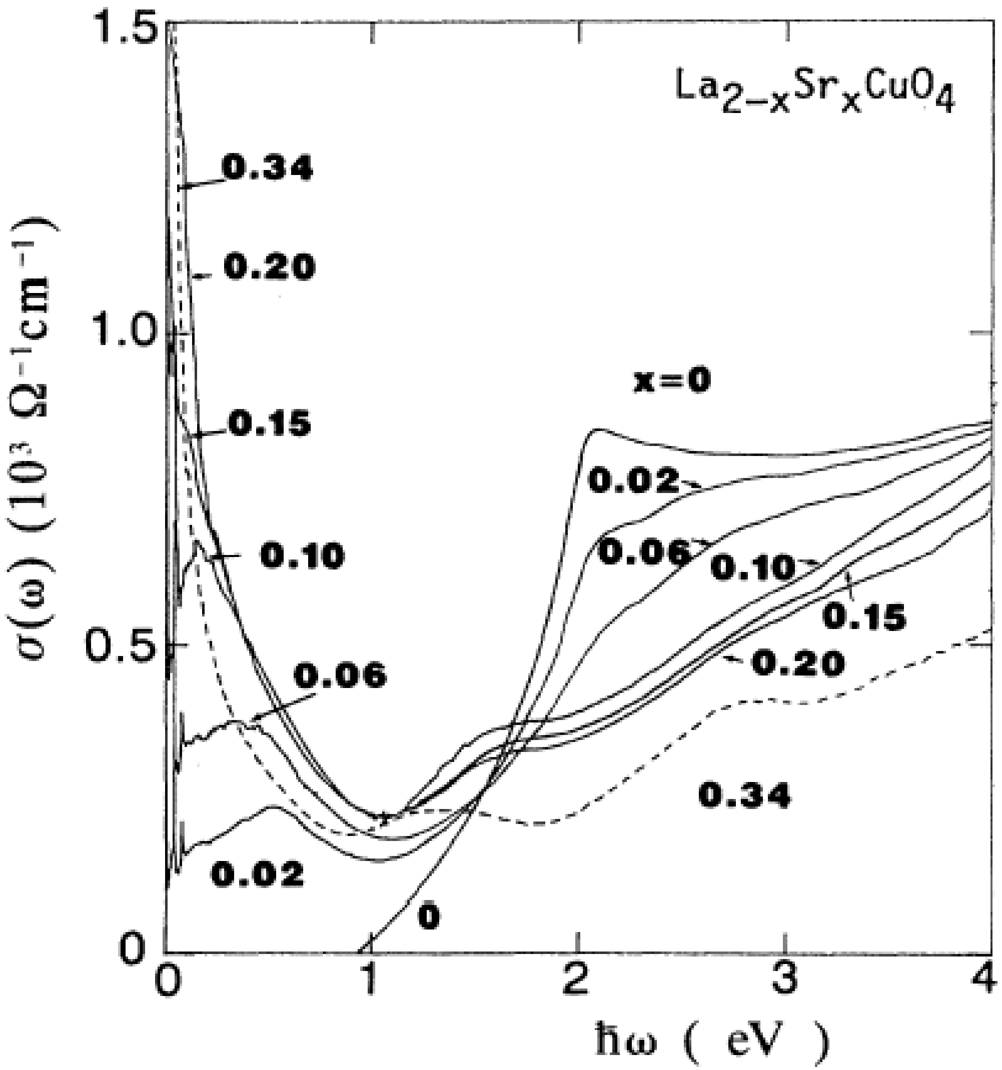
The Optical Conductivity Sum Rule



Sum rule

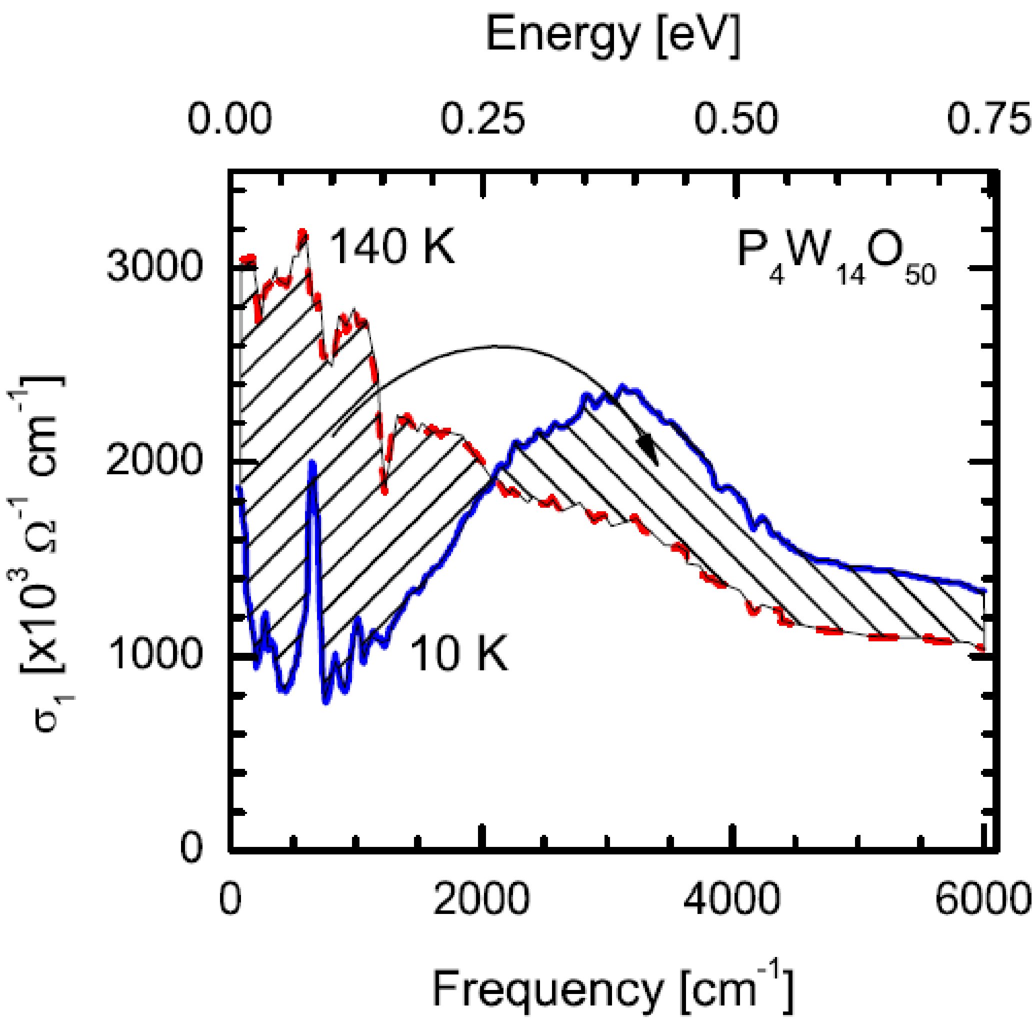
$$\int_0^\infty \sigma_1(\omega, T)\,d\omega = \frac{\pi}{2}\frac{ne^2}{m}$$

The charge transfer gap of $(La,Sr)_2CuO_4$



Uchida et al. PRB 43, 7942 (1991).

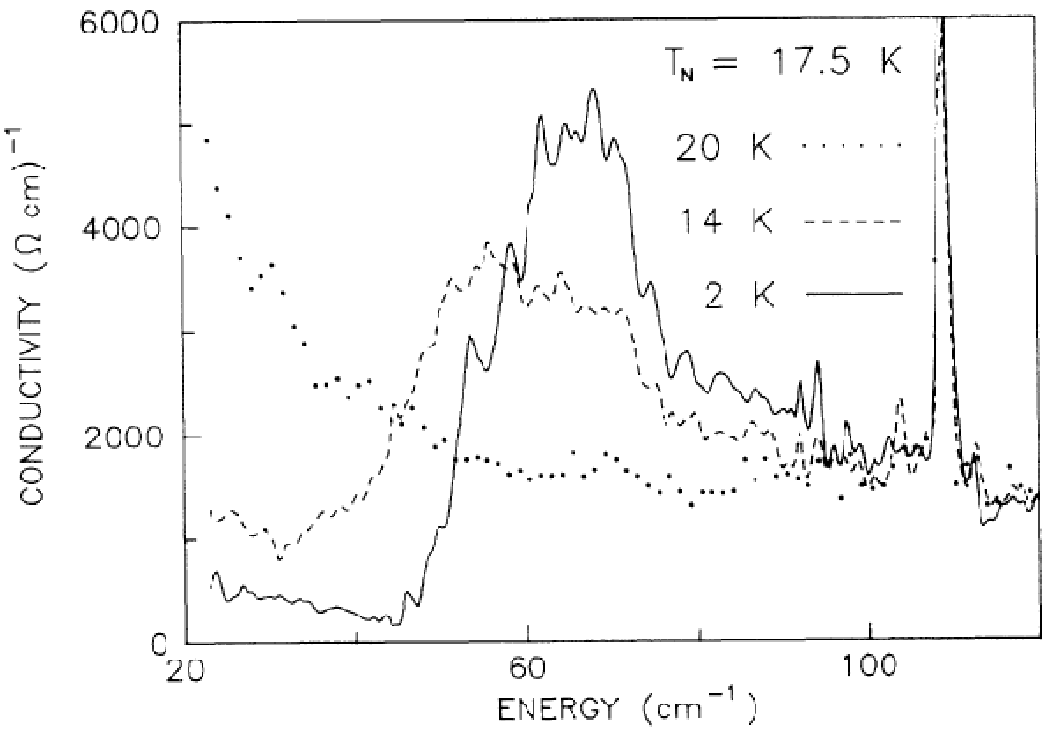
Density-wave like gap



- ✓ $P_4W_{14}O_{50}$
- ✓ CDW transition at T~140 K
- ✓ CDW gap at 1400 cm^{-1}
- ✓ Spectral weight transfer from low to high frequencies

Zhu *et al.*, PRB **65**, 214519 (2002)

Hidden order in URu_2Si_2

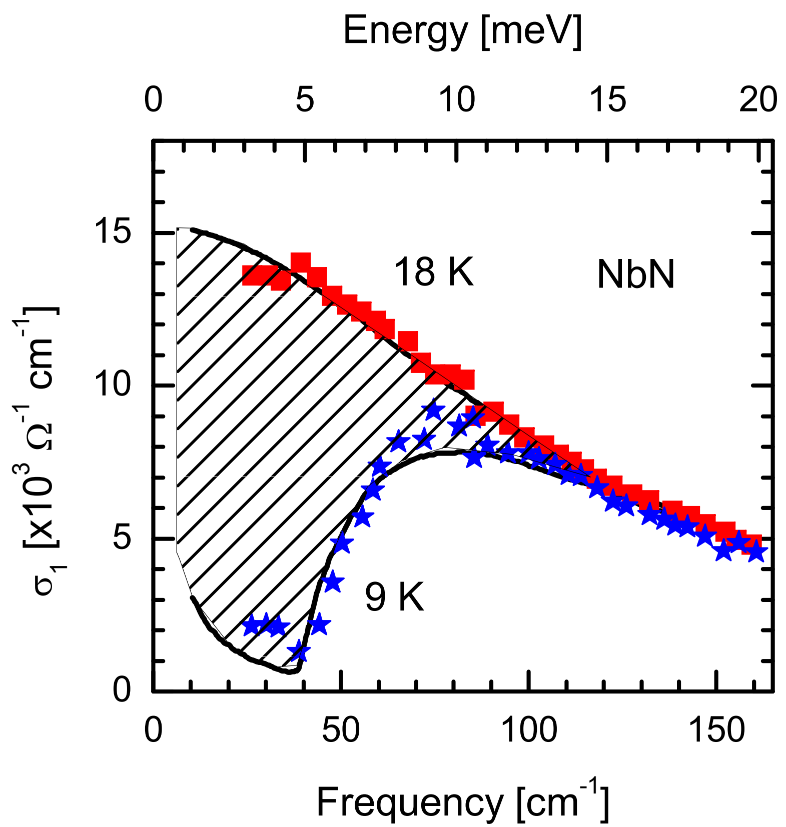


Bonn, PRL 61, 1305 (1988)

- ✓ Phase transition at 17K to an unknown order parameter
- ✓ Possible renormalization of the band structure
- ✓ Order closely related to magnetism
- ✓ Gap opening at the Fermi surface

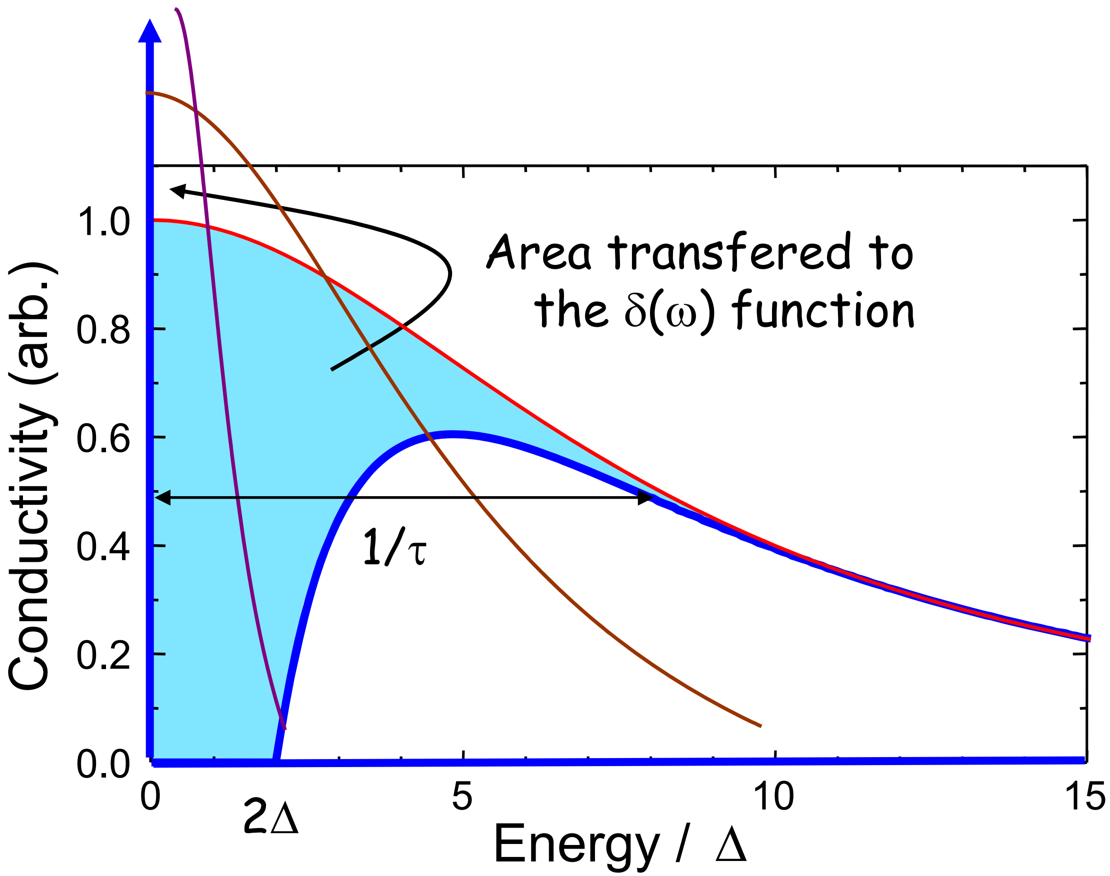
Even when we do not know what the crap is the gap, it still creates a spectral weight transfer

And let's go superconducting



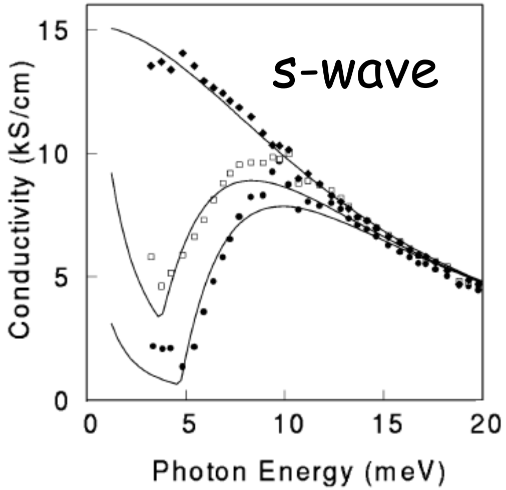
Is the sum rule violated???

Superconducting gap – Back to Drude



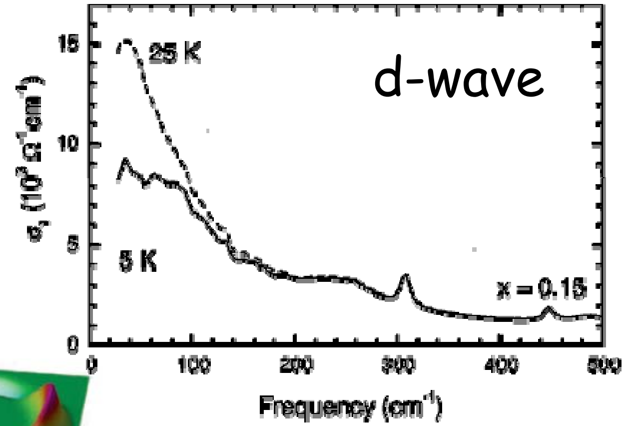
- ✓ ω = 0 not accessible to optics
- ✓ The "Lost area" gives superfluid density

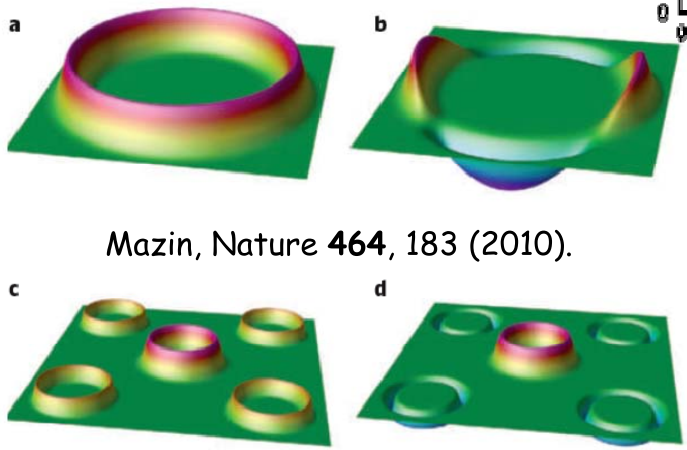
Manifestation of the superconducting gap in the infrared



NbN – T_c = 16.5 K
Somal *et al.*, PRL **76**, 1525 (1996)

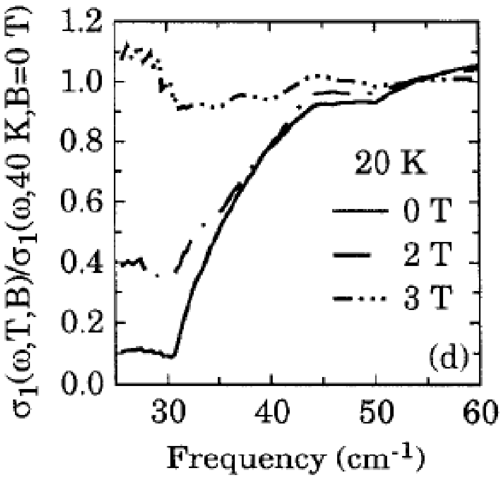
$(Pr,Ce)_2CuO_4$ - T_c = 21 K
Zimmers *et al.*, PRB **70**, 132502 (2004)





Mazin, Nature **464**, 183 (2010).

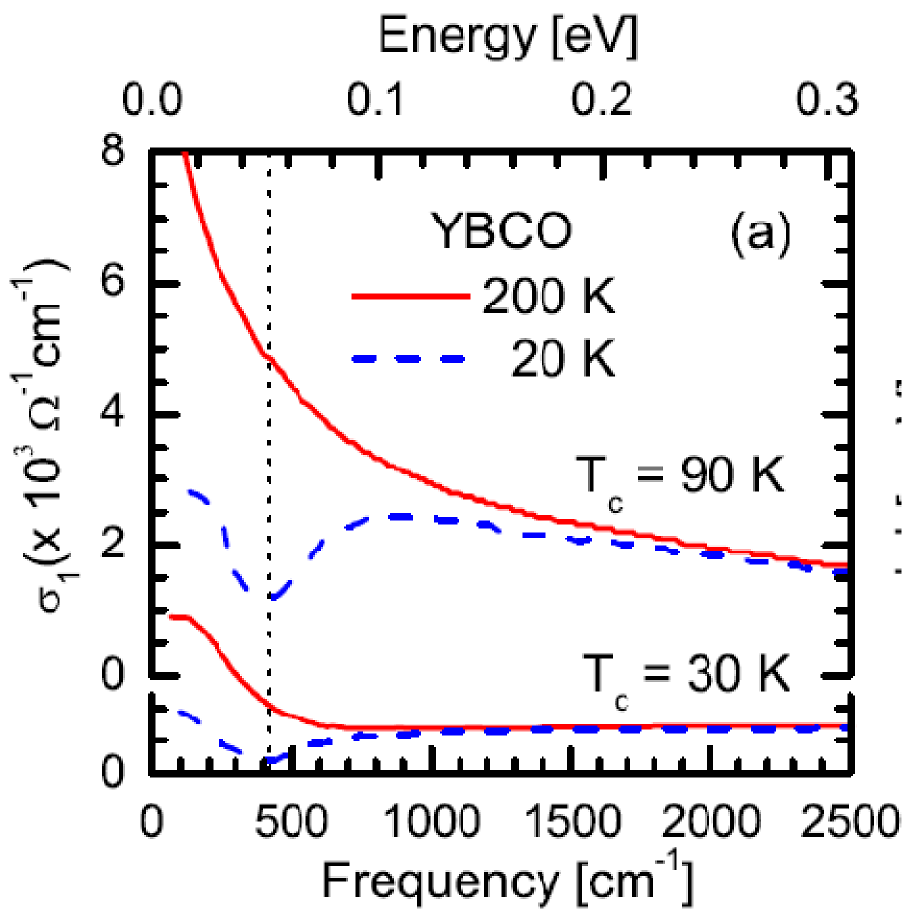
s-multiband



MgB_2 – T_c = 39 K
Perucchi *et al.*, PRL **89**, 097001 (2002).

Pnictides:
$s_{\pm}$? d? Multiband?
Extended s?

The gap in the cuprates



T_c changes by a factor 3 and the thing that looks like the gap stays put ?!?!?!?!

Clean and Dirty

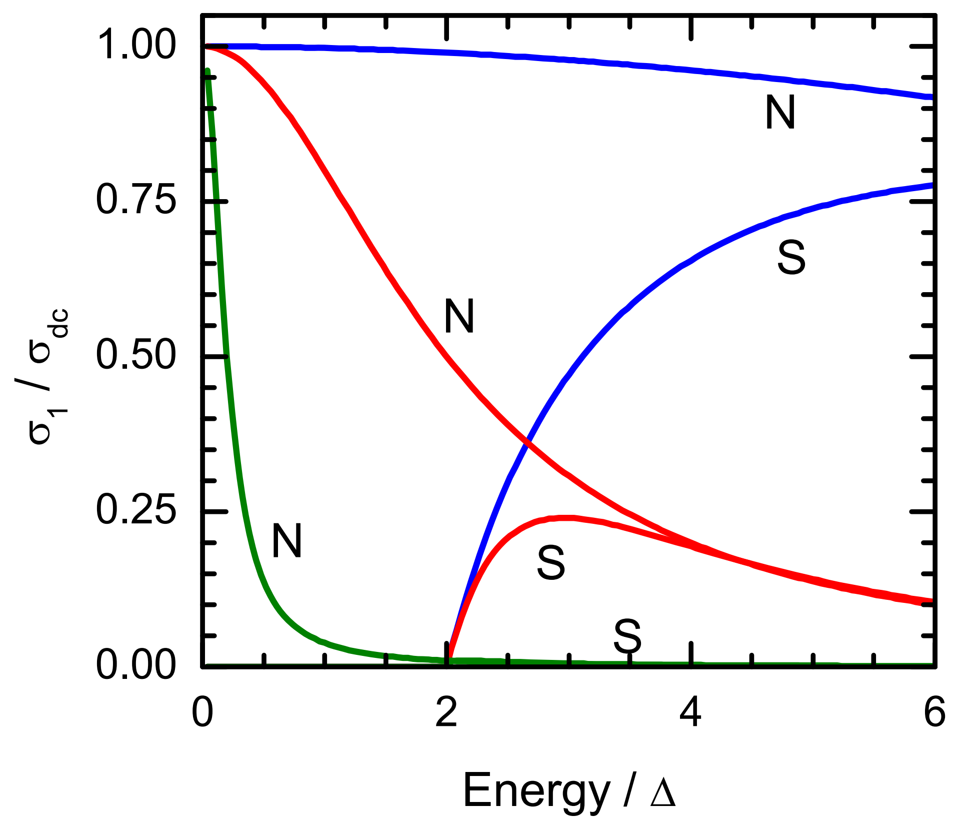
Clean superconductor

$$l \gg \xi_0$$

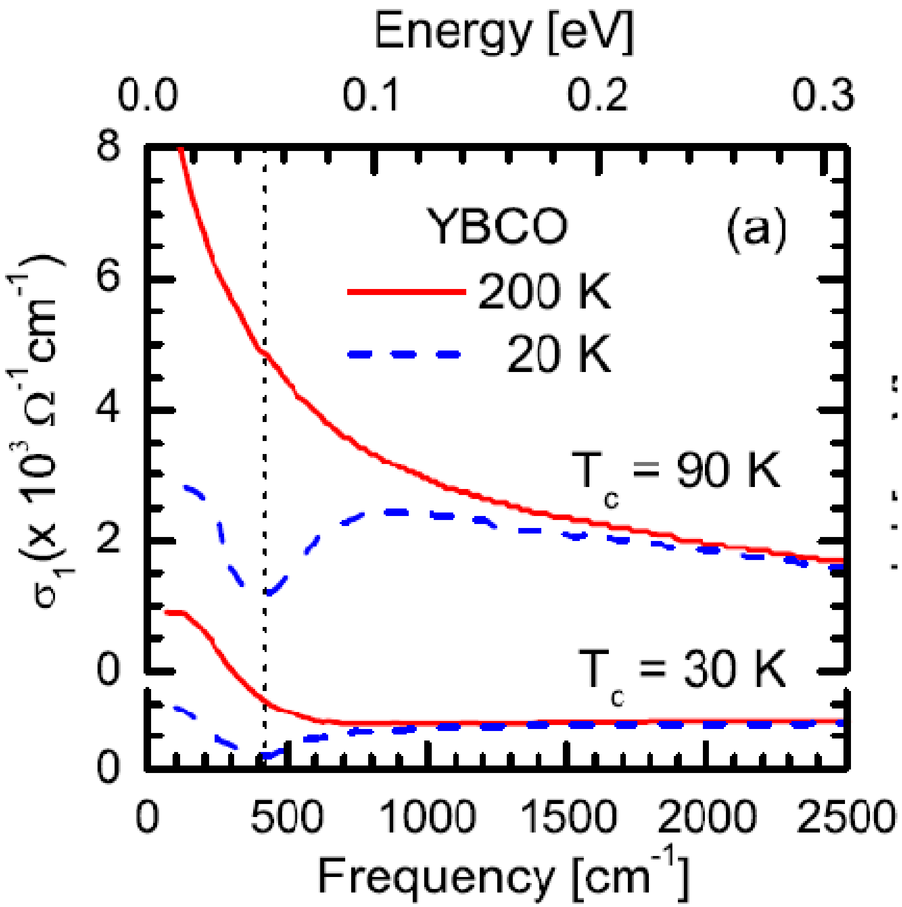
$$v_f \tau \gg \frac{v_f}{2\Delta}$$

$$\frac{1}{\tau} \ll 2\Delta$$

- ✓In a clean superconductor you do not see the gap because there is no optical conductivity left at that frequency
- ✓The important feature in the optical conductivity is the determination of the superfluid density.
- ✓The gap is a bonus!!!



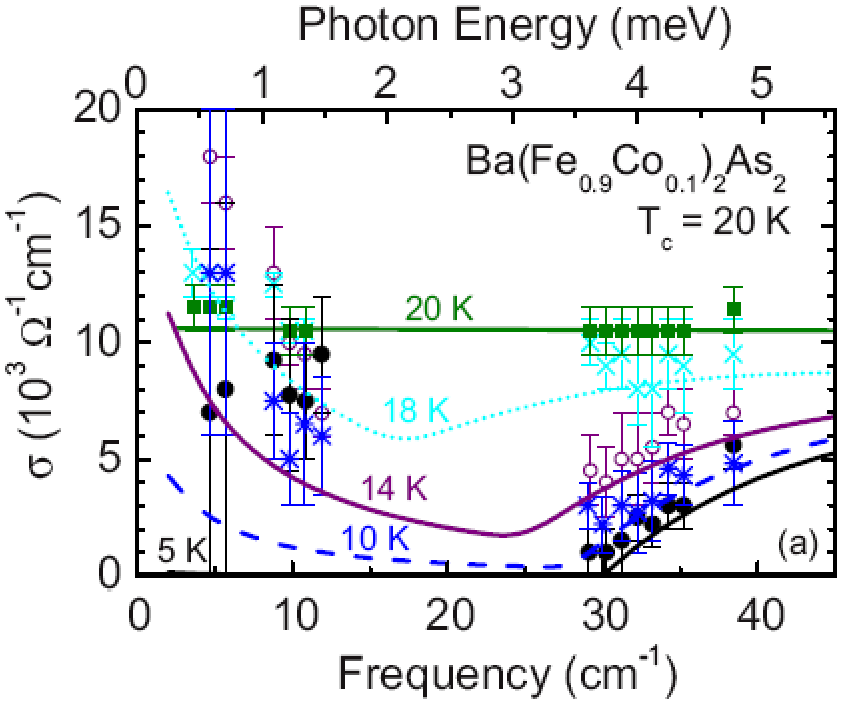
The gap in the cuprates



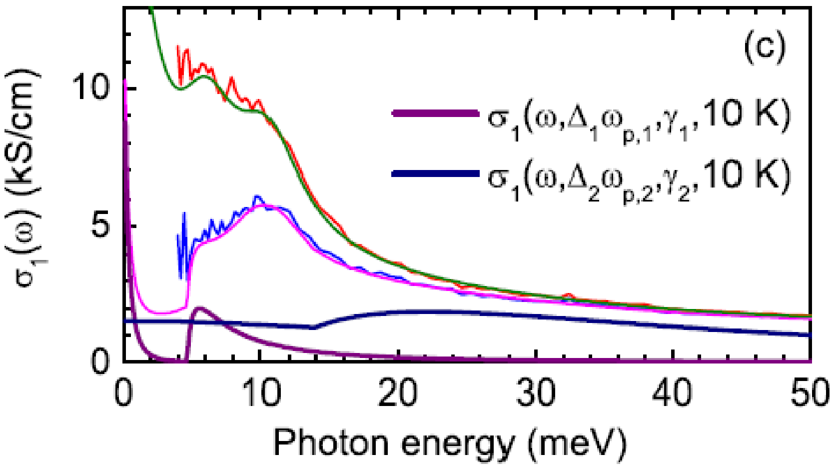
T_c changes by a factor 3 and the thing that looks like the gap stays put ?!?!?!?!

Yep! Cause it ain't the gap, stupid!!

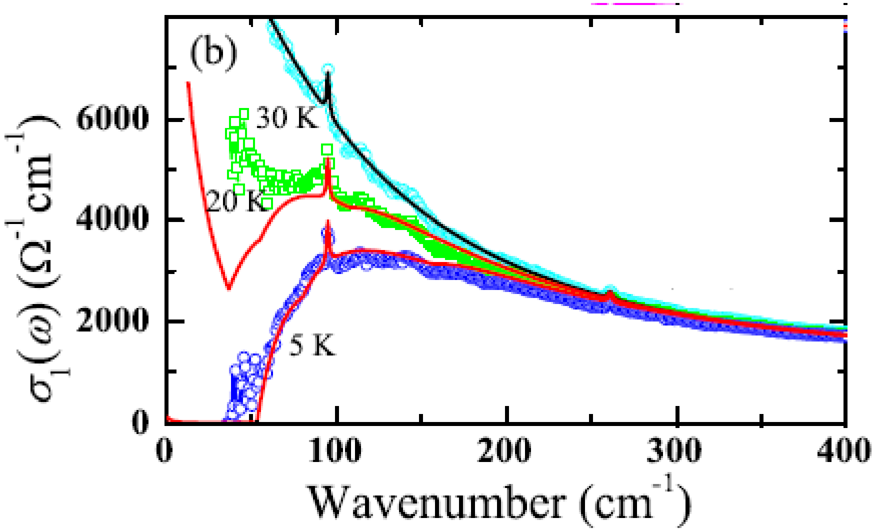
Pnictides Gap Review



Claim: Good agreement with s-wave BCS
Gorshunov *et al.*, PRB **81**, 060509 (2010)

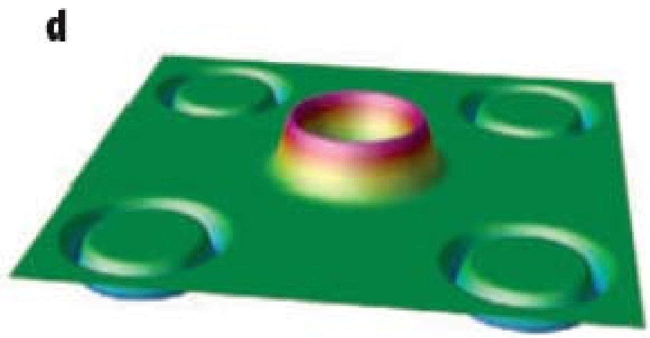


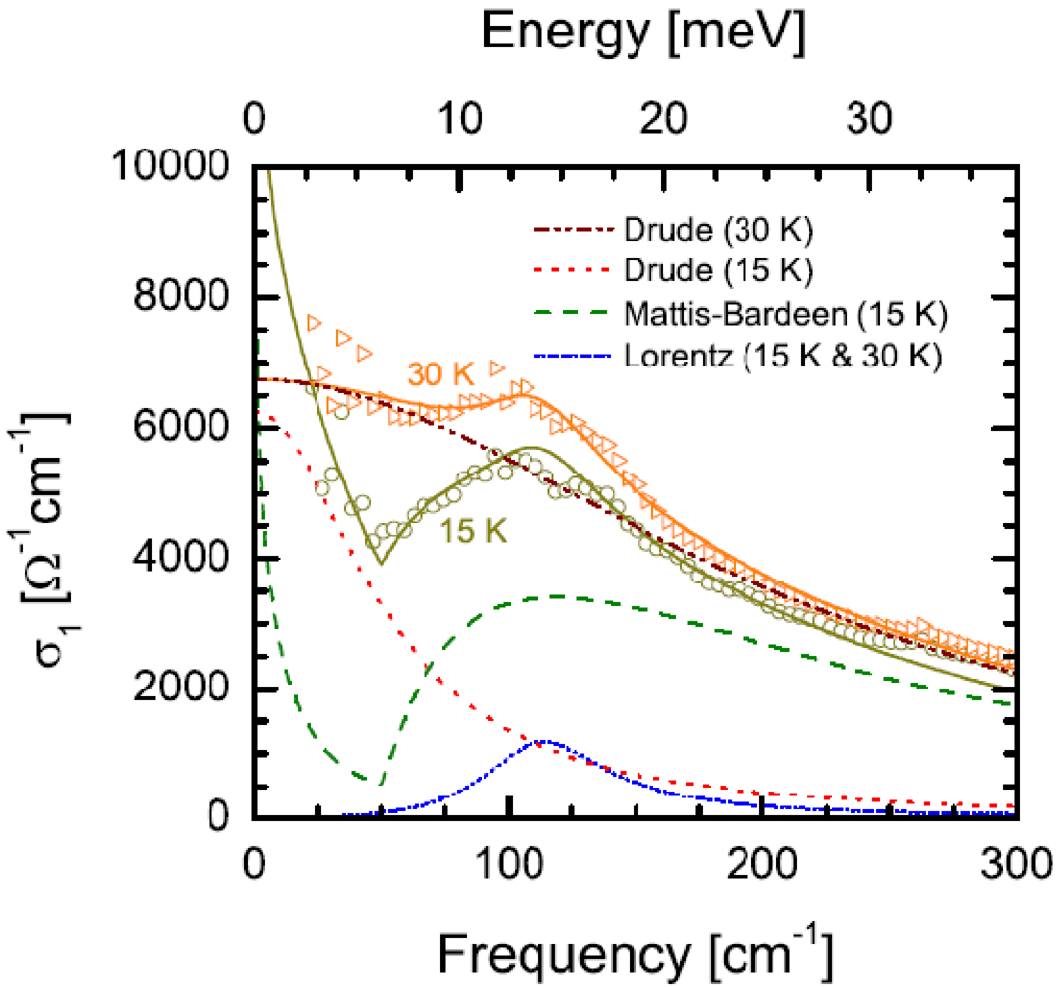
Claim: 2 gap BCS
van Heuman *et al.*, arXiv:0912.0636v1



Claim: 3 gap BCS
Kim *et al.*, arXiv:0912.0140

s± gap: Interband scattering is pair breaking





- ✓ Interband scattering annihilates Cooper pairs
- ✓ Residual Drude peak in the superconducting state

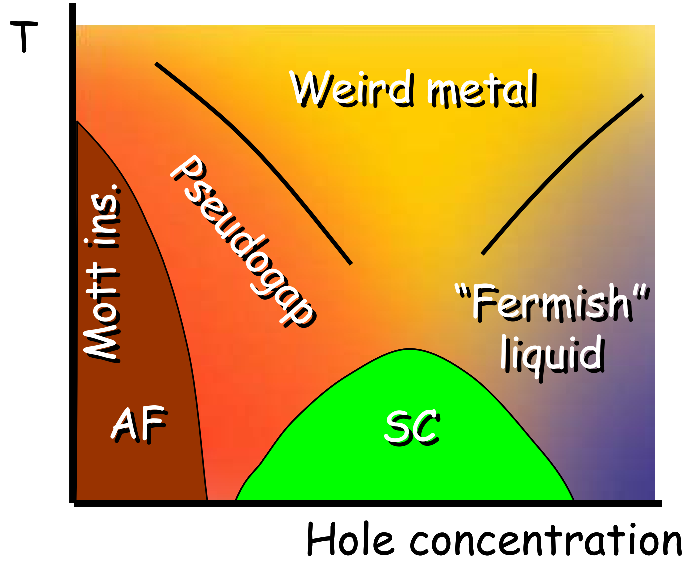
Lobo et al. (2010)

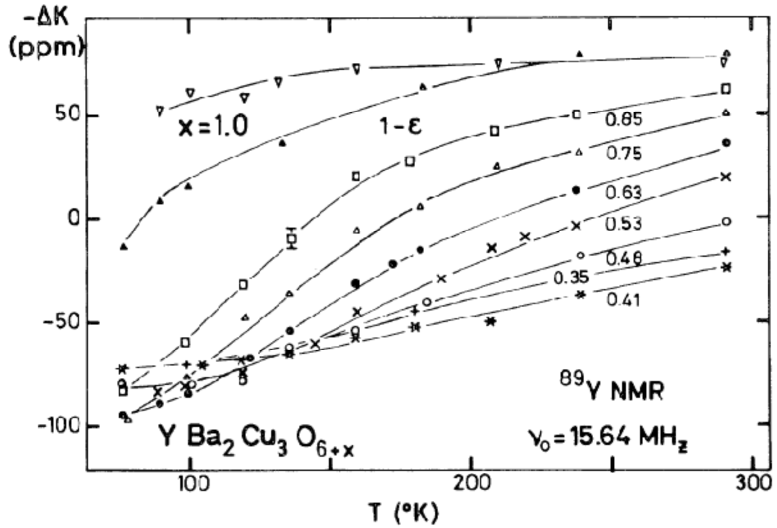
The infamous cuprate pseudogap

CNRS

ESPCI ParisTech

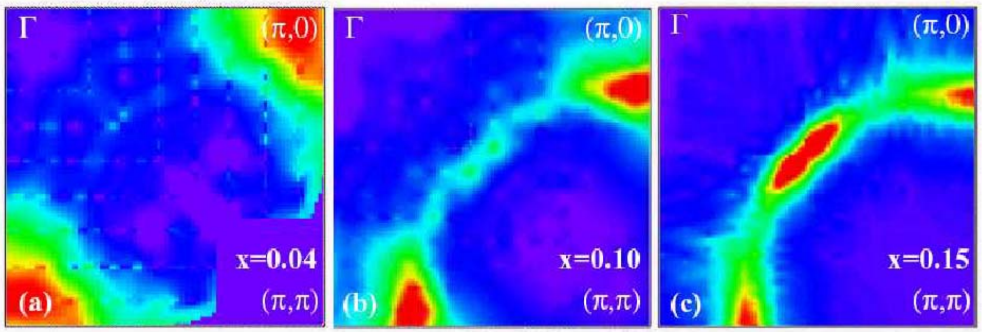
The pseudogap





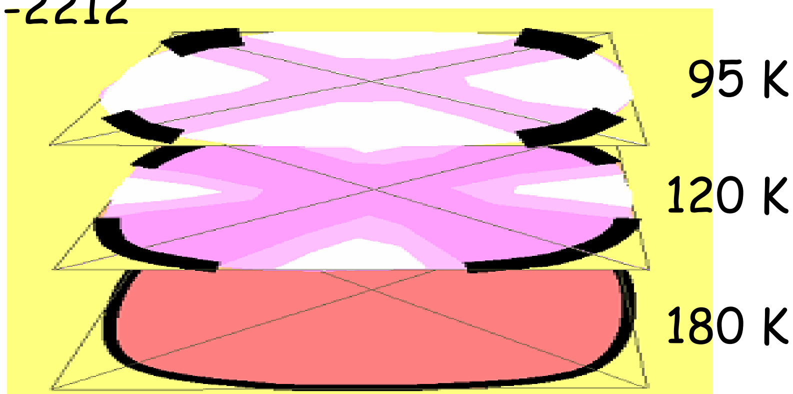
Alloul, PRL 63, 1700 (1989)

$Nd_{2-x}Ce_xCuO_4$



Armitage et al. PRL 81, 257001 (2002).

Bi-2212



Norman et al. Nature 392, 157 (1998).

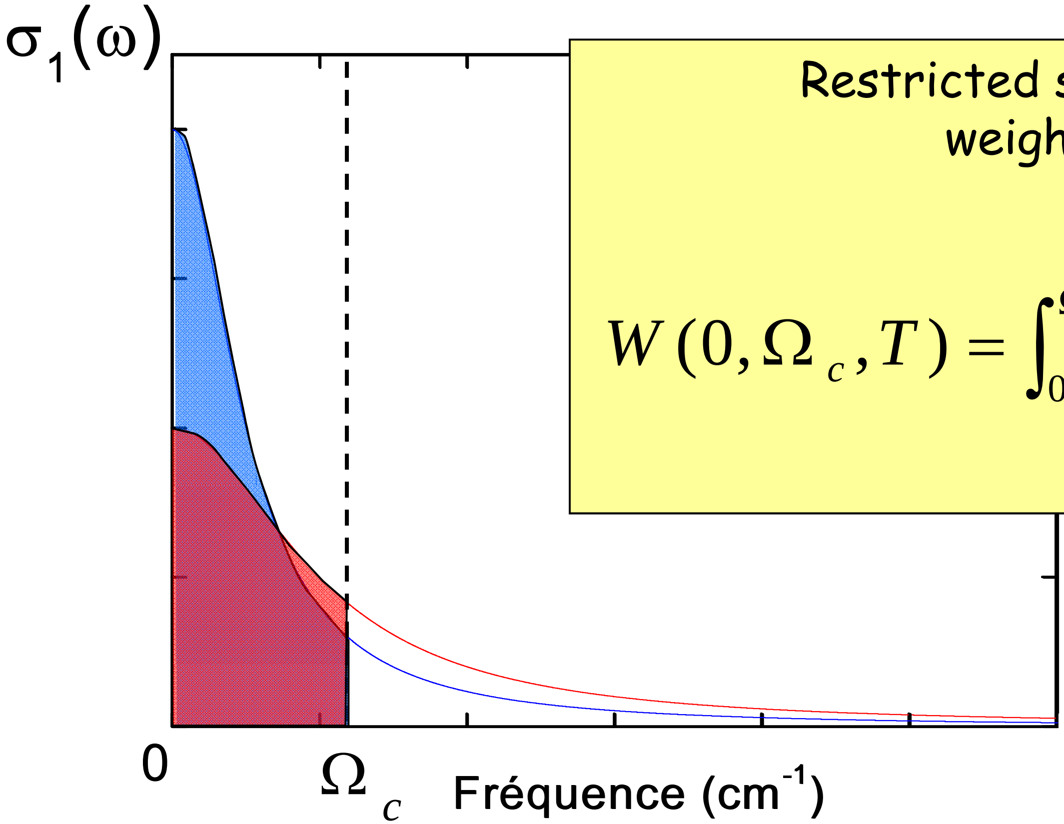
Restricted Spectral Weight or Partial Sum Rule

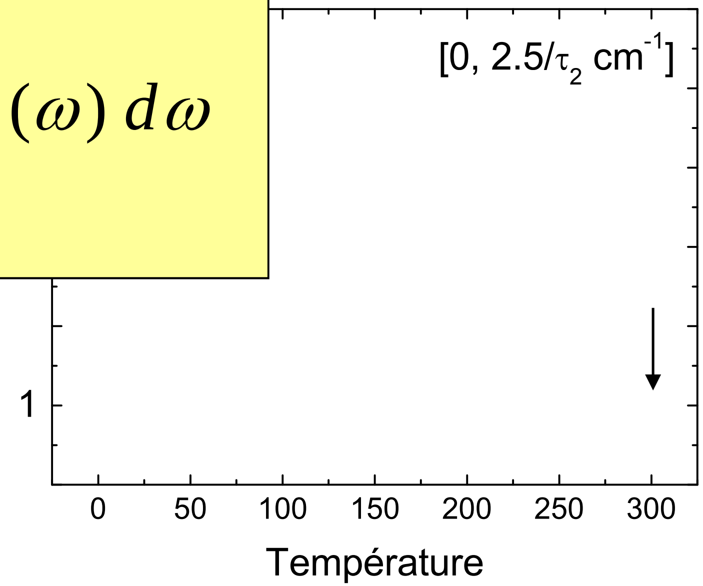
Sum rule

$$\int_0^{\infty} \sigma_1(\omega,T)d\omega = \frac{\pi}{2}\frac{ne^2}{m}$$

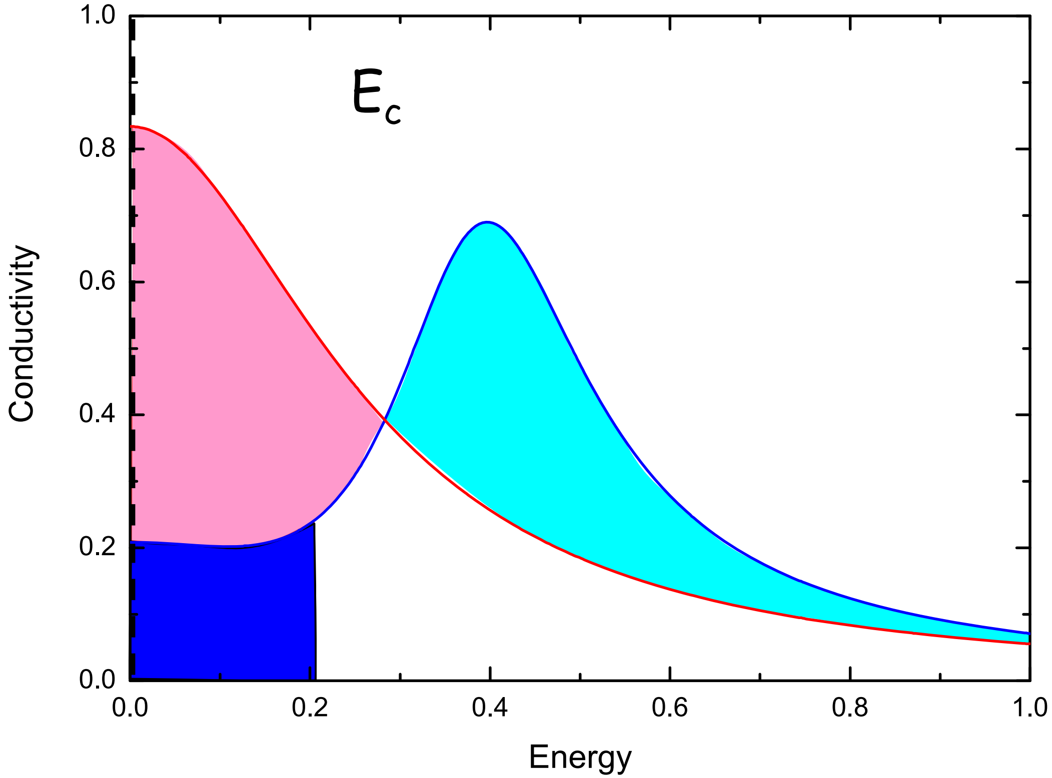
Restricted spectral weight

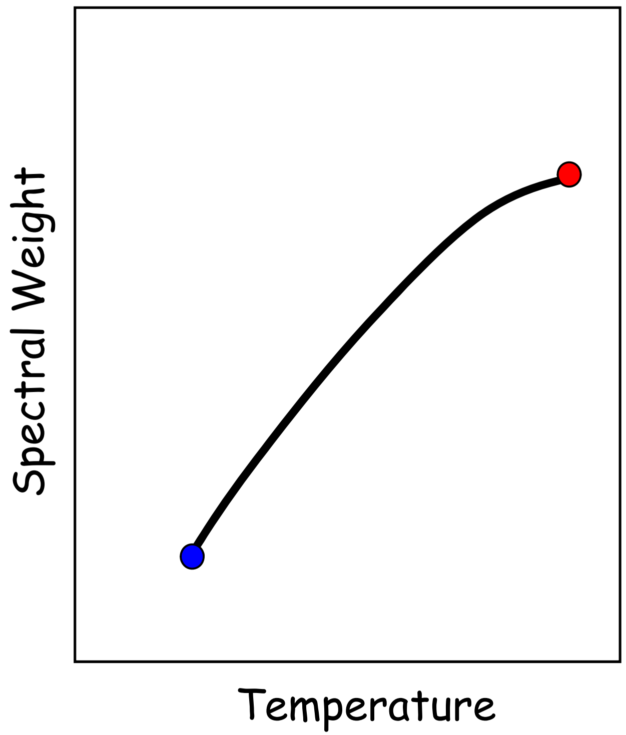
$$W(0,\Omega_c,T) = \int_0^{\Omega_c} \sigma_1(\omega)\, d\omega$$



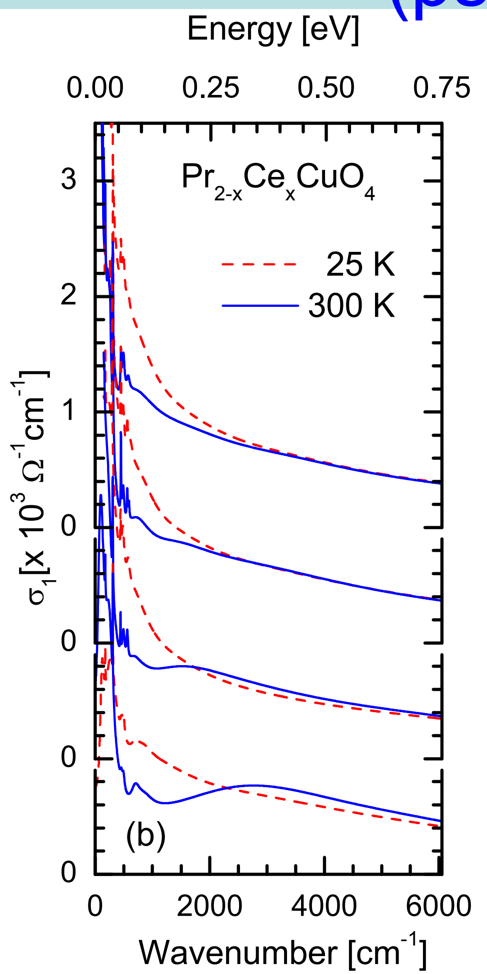


The Sum Rule when a Gap Opens

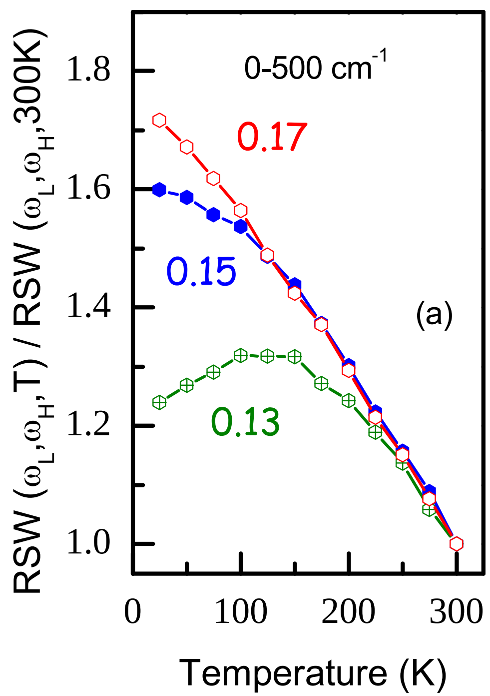




$(Pr,Ce)_2CuO_4$ - A well behaved normal state (pseudo) gap



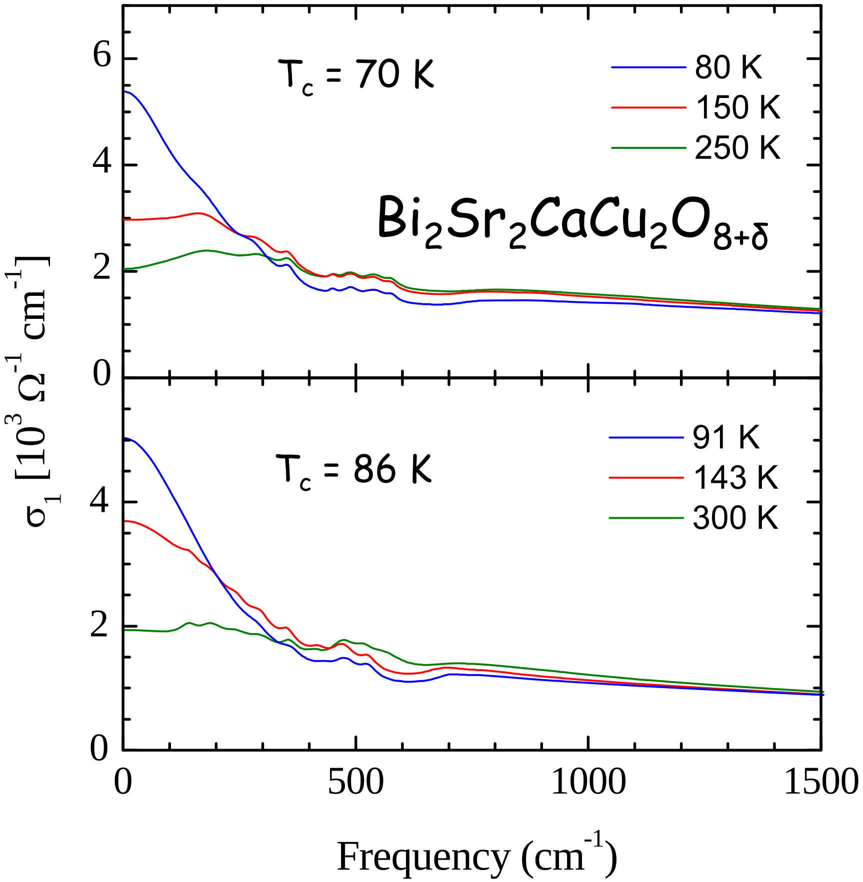
Partial Sum Rule



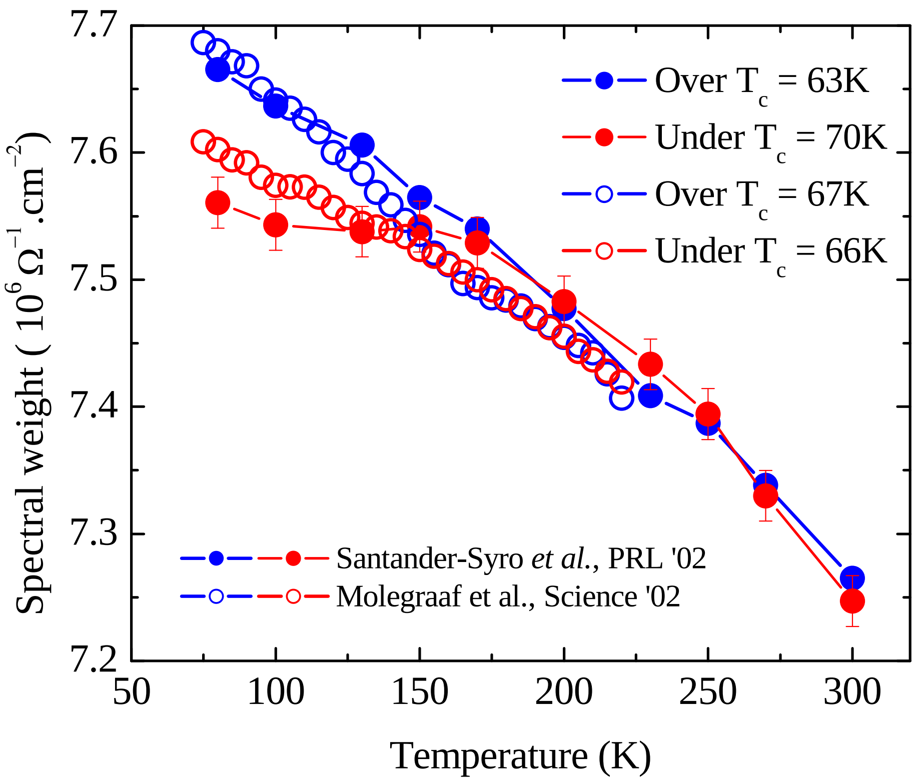
*Zimmers et al., EPL **70**, 225 (2005).*

Bi-2212 – Where is the pseudogap?

Optical conductivity

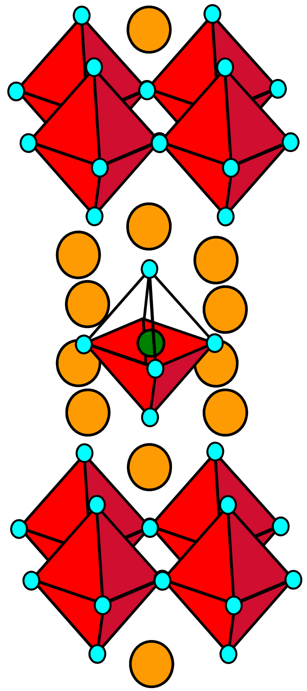


Partial sum rule

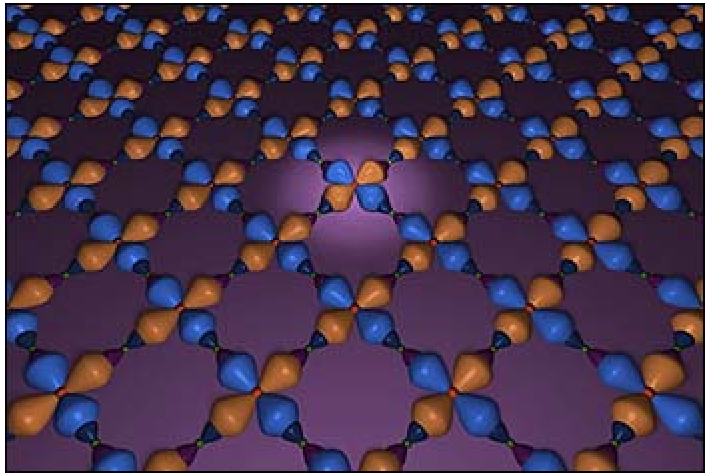


*Santander-Syro et al., PRL **88**, 097005(2002).*

Why the difference?

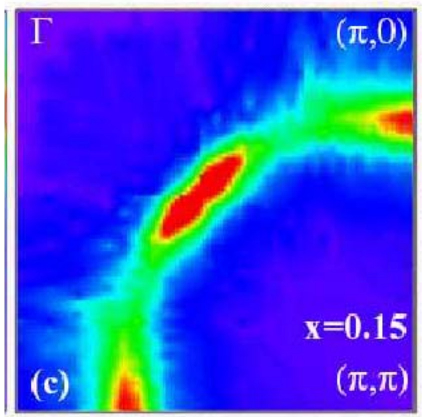


$(La,Sr)_2CuO_4$



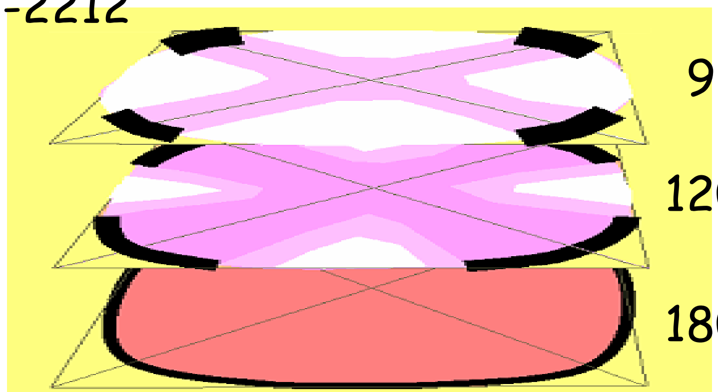
The ab-plane optical probes mostly the nodal (π,π) directions

$Nd_{2-x}Ce_xCuO_4$



Armitage et al. PRL 81, 257001 (2002).

Bi-2212



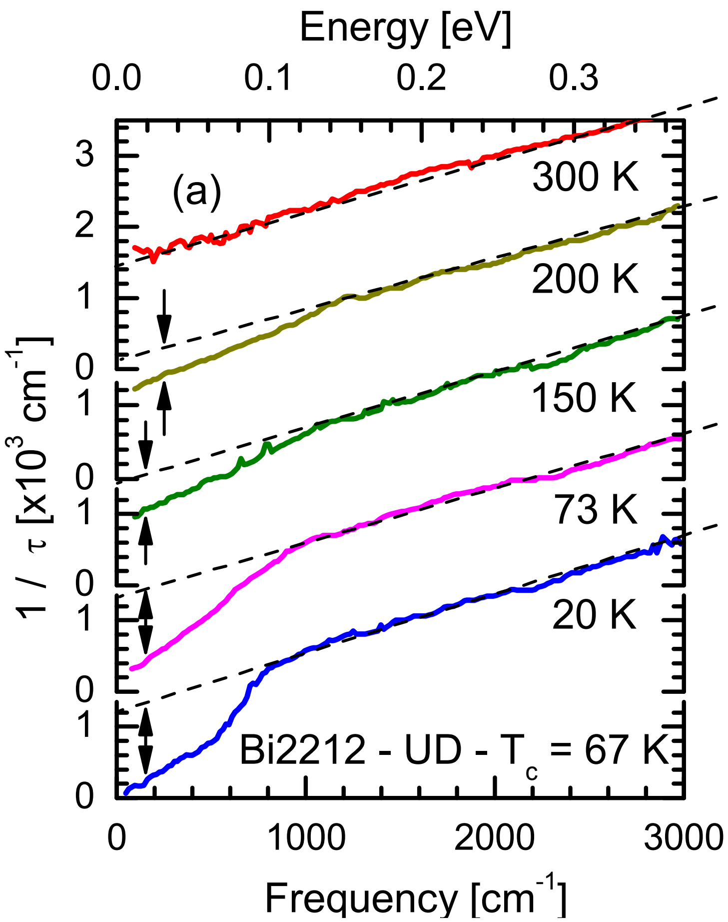
95 K

120 K

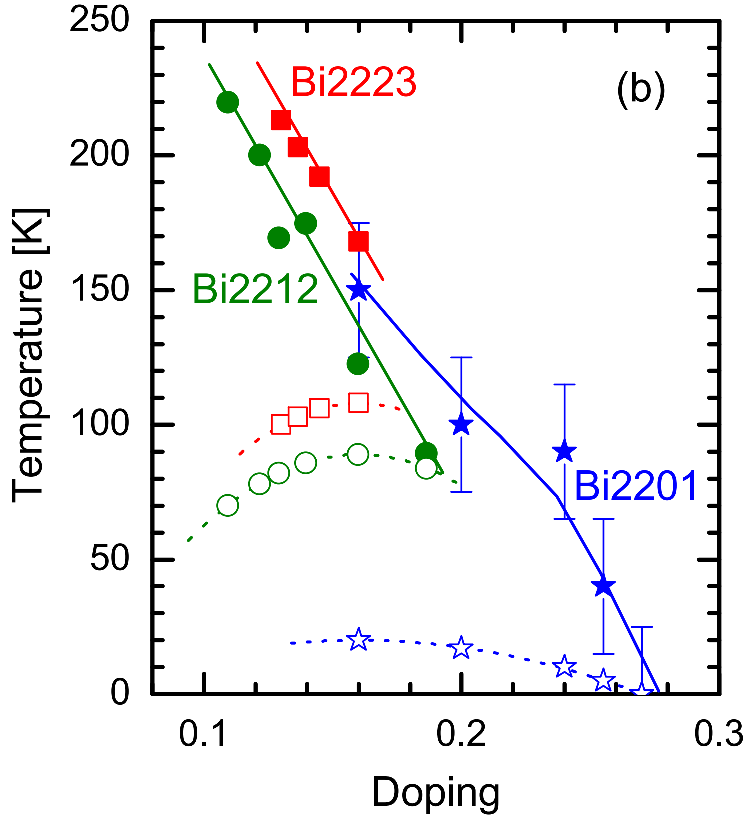
180 K

Norman et al. Nature 392, 157 (1998).

The pseudogap in the scattering rate

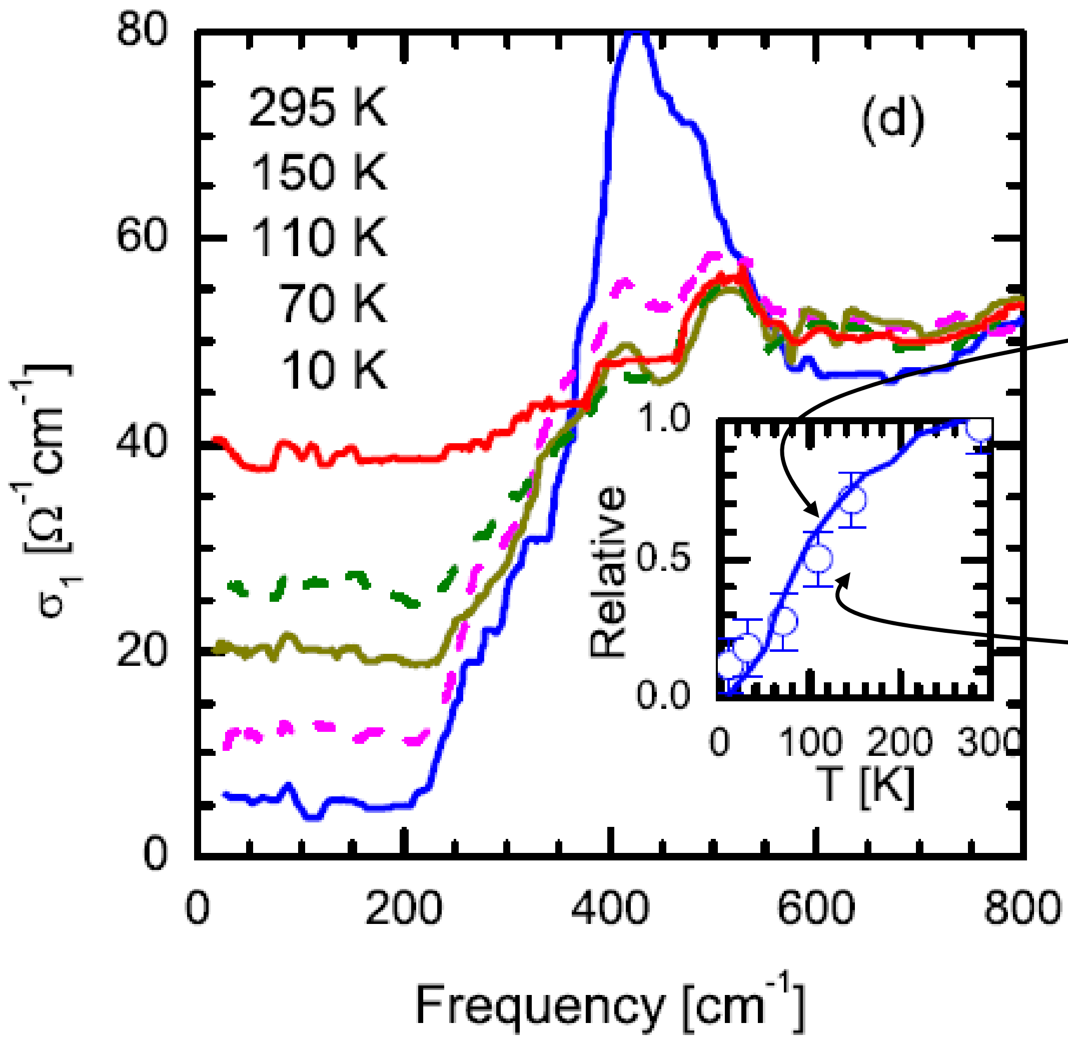


Hwang et al., Nature **427**, 714 (2004)



Lobo *et al.* (2009)

The pseudogap along the c-axis of YBCO (T_c = 63 K)



Spectral weight decrease

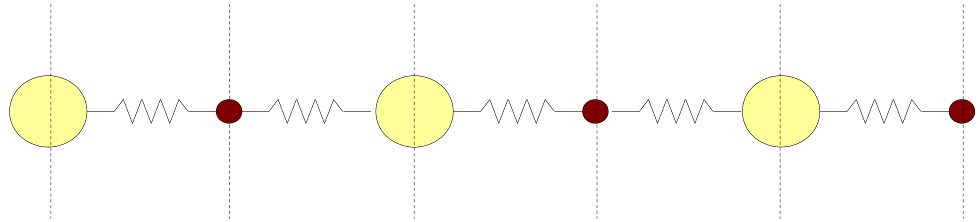
Knight shift

Homes et al., PRL 71, 1645 (1993).


AND NOW FOR SOMETHING
COMPLETELY DIFFERENT

Light and Matter interaction in an Insulator

Phonons and the Harmonic Approximation



$$m\ddot{x} + m\gamma\dot{x} + m\Omega_0^2 x = -qE$$

$$P = nqx = (\varepsilon - 1)E = \chi E$$

$$\chi = \frac{\Delta\varepsilon\Omega_0^2}{\Omega_0^2 - \omega^2 - i\gamma\omega}$$

And for many phonons

$$\varepsilon = \varepsilon_\infty + \sum_j \frac{\Delta\varepsilon_j \Omega_{TOj}^2}{\Omega_{TOj}^2 - \omega^2 - i\gamma_j\omega}$$

$$\Delta\varepsilon_j \Omega_{0j}^2 = \frac{n_j q_j^2}{m_j} \qquad \varepsilon(0) = \varepsilon_\infty + \sum_j \Delta\varepsilon_j$$

Phonons & the f-sum rule

The f-sum rule (particle conservation):

$$\sigma = \frac{2\pi i\omega}{Z_0}\chi$$

$$\int_0^\infty \mathrm{Re}\left[\sigma(\omega)\right] d\omega = const \times \left(\frac{n_e}{m_e} + \frac{n_p}{m_p}\right)$$

The f-sum rule for phonons:

$$\sum_j \Delta\varepsilon_j \Omega_{0j}^2 = \sum_j \frac{n_j q_j^2}{m_j} = const$$

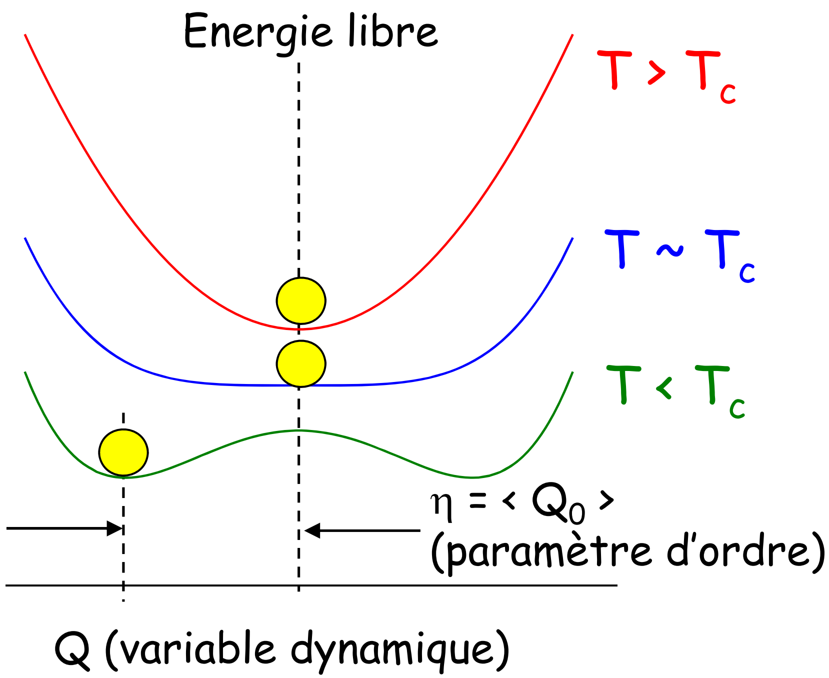
The f-sum rule for decoupled phonons:

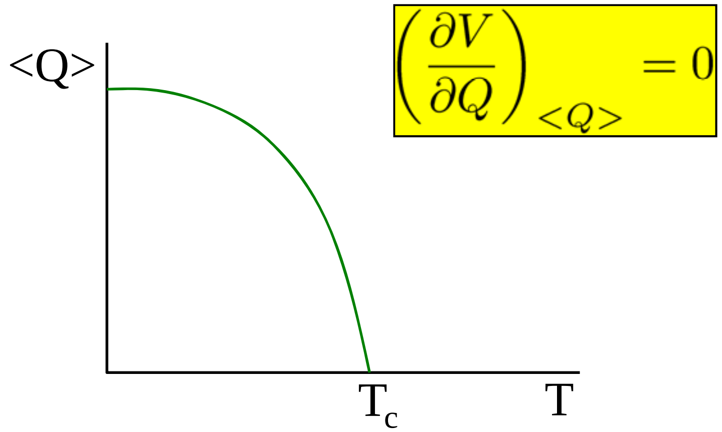
$$\Delta\varepsilon_j \Omega_{0j}^2 = \frac{n_j q_j^2}{m_j} = const$$

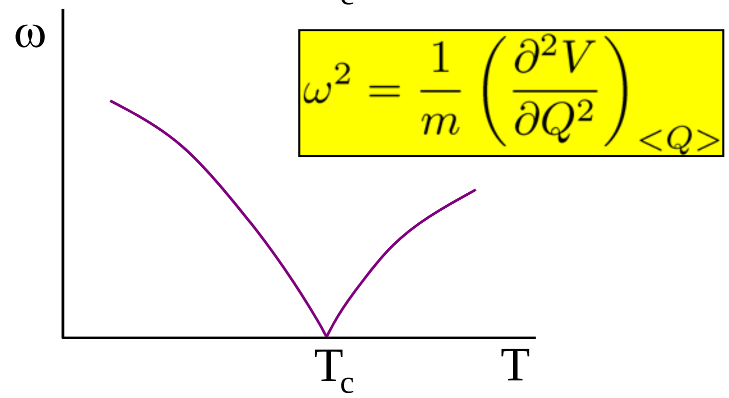
Soft Mode & Phase Transitions

Q is a dynamic variable of the system

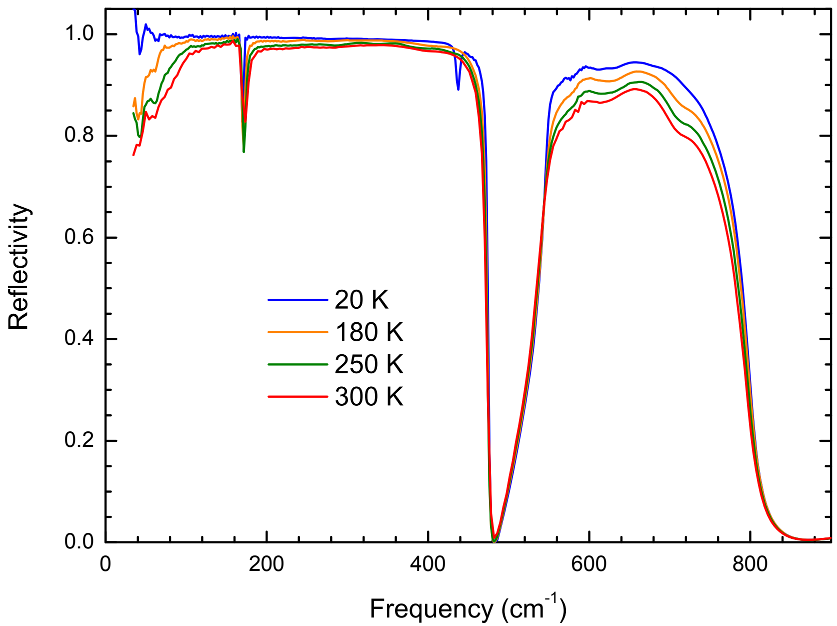
$$\text{Order parameter } \eta = \langle Q \rangle \begin{cases} = 0, & T > T_c \\ \neq 0, & T < T_c \end{cases}$$

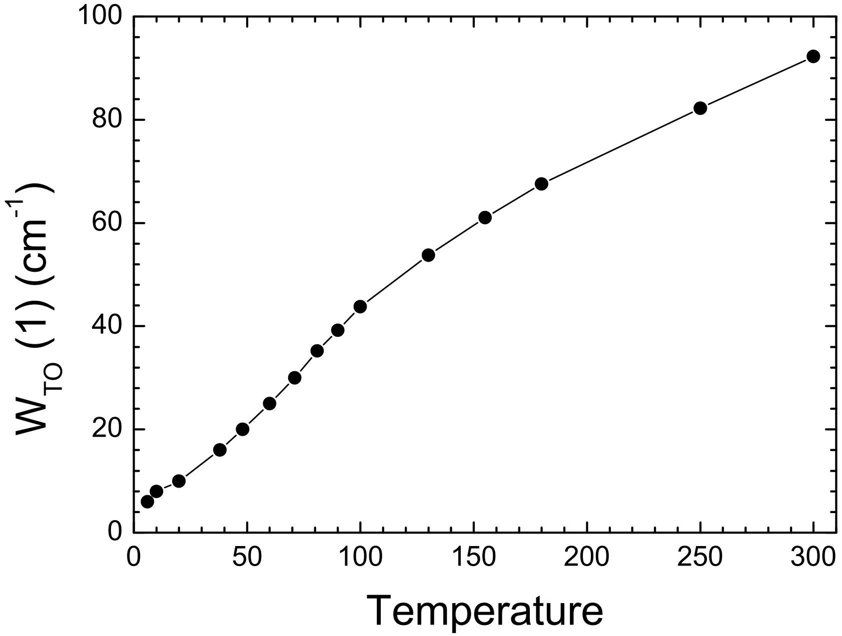






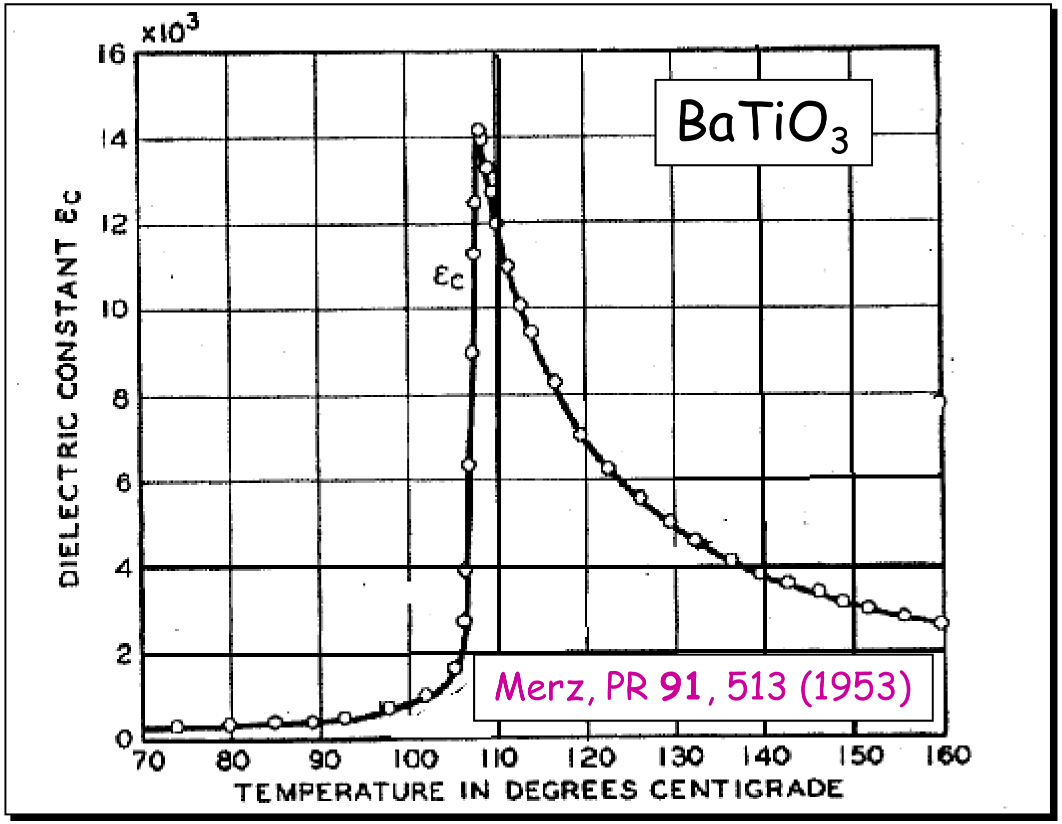
$SrTiO_3$ – A wannabe ferroelectric





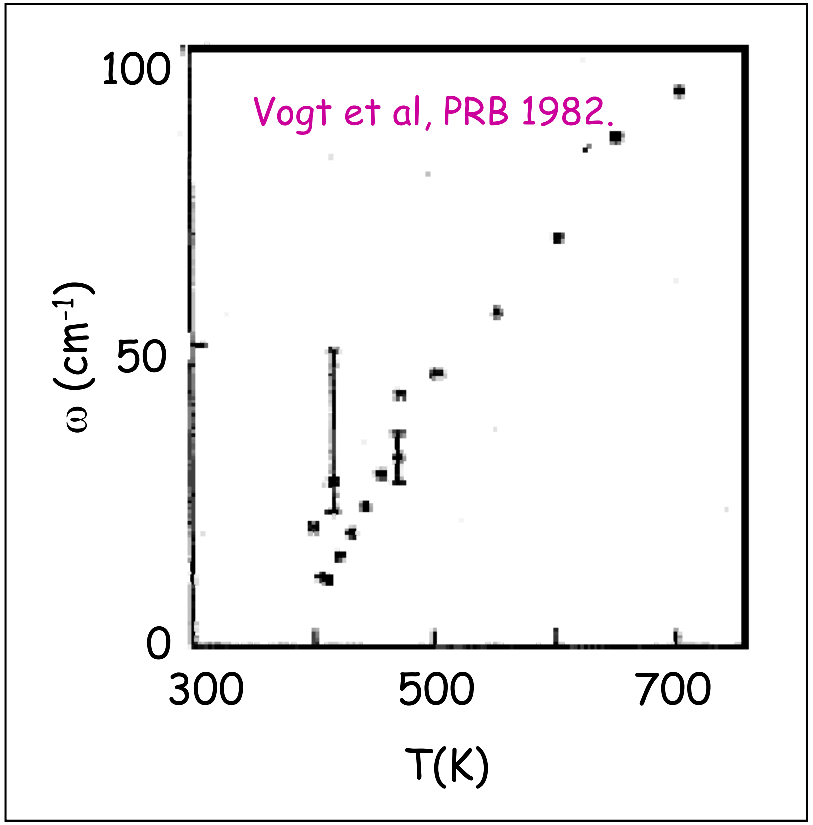
- ✓ Incipient soft mode driven ferroelectric transition
- ✓ T_C ~ -15 K

Soft mode and Ferroelectrics



$$\varepsilon(0) = 1 + \sum_j \Delta\varepsilon_j$$

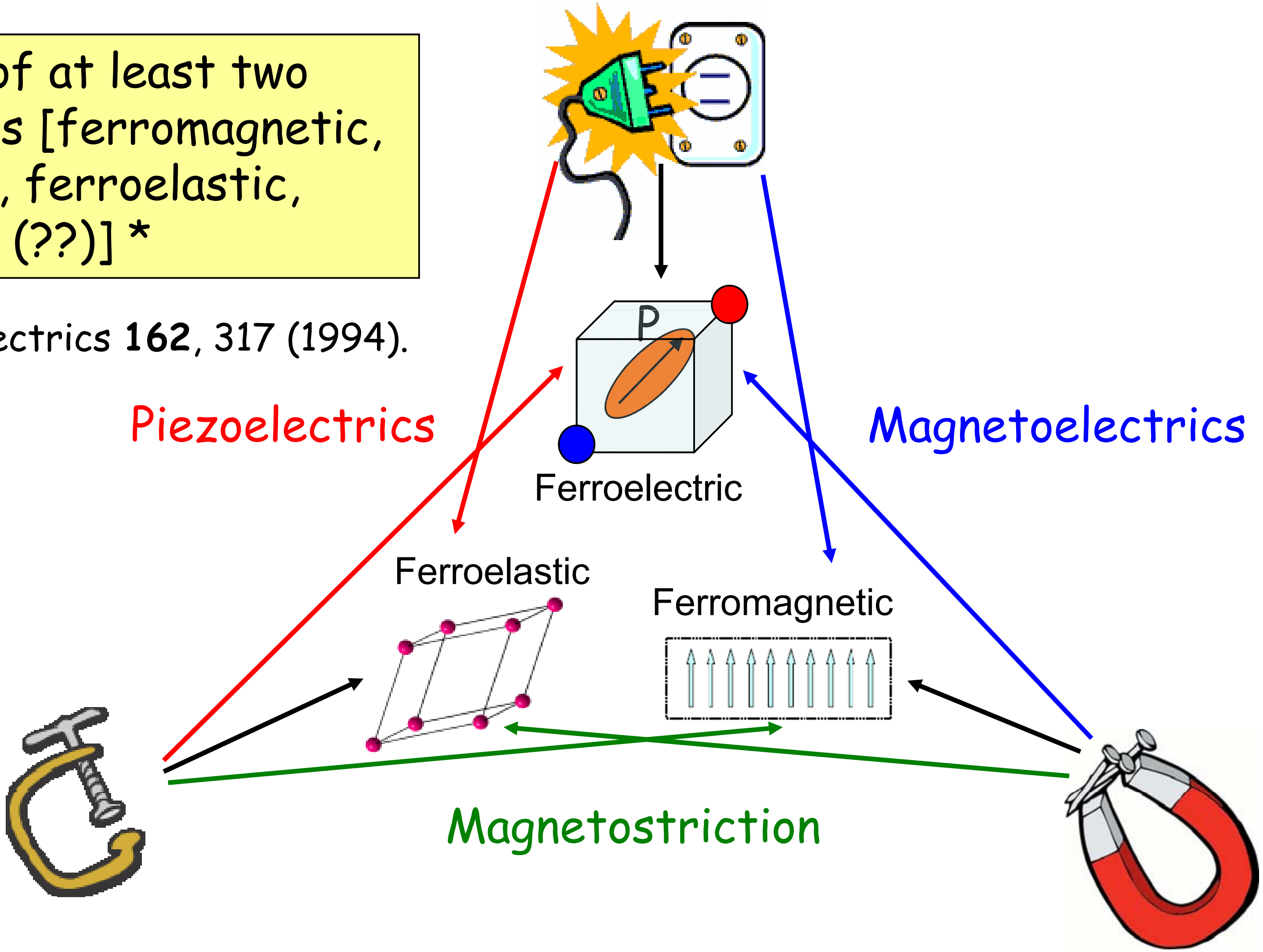
$$\Delta\varepsilon_j \Omega_{0j}^2 = \frac{n_j q_j^2}{m_j} = const$$



The Multiferroic Materials Totem

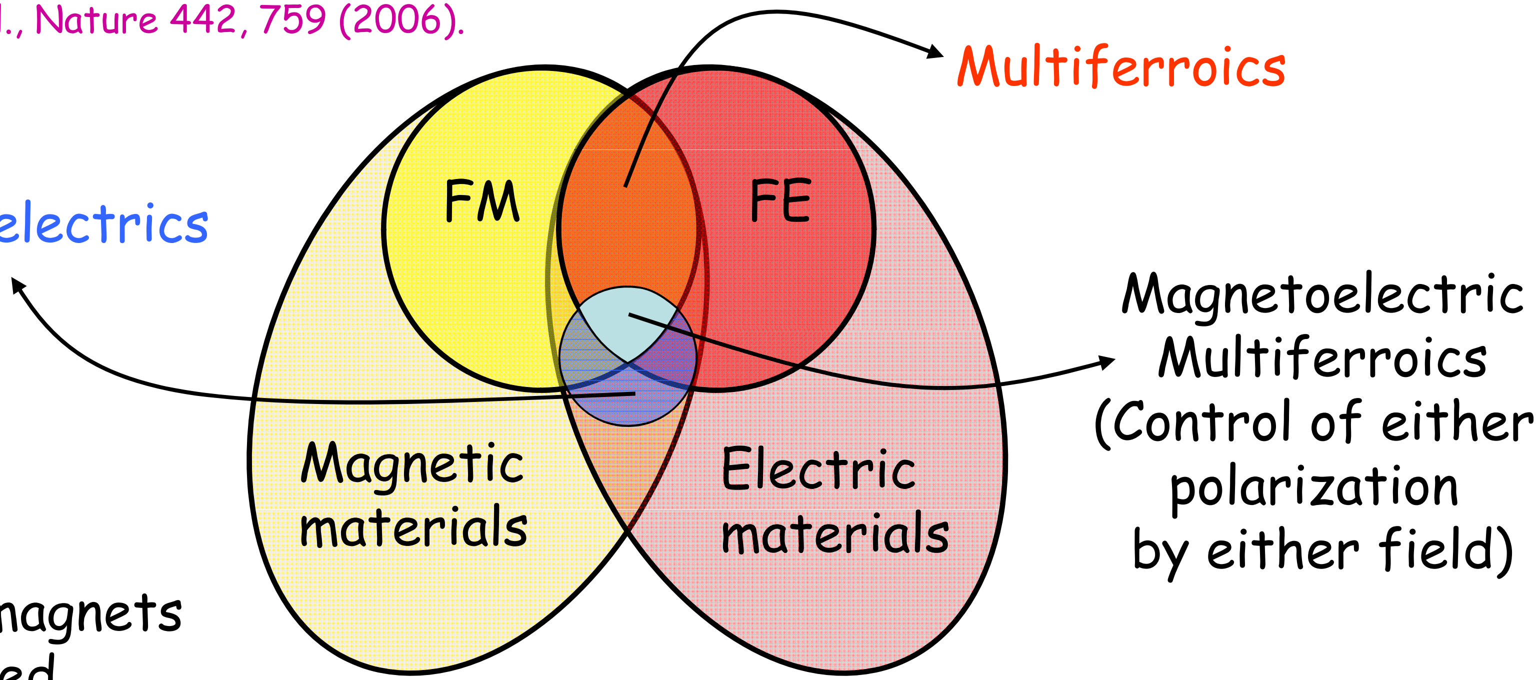
Coexistence of at least two ferroic orders [ferromagnetic, ferroelectric, ferroelastic, ferrotoroidal (??)] *

Schmid, Ferroelectrics **162**, 317 (1994).



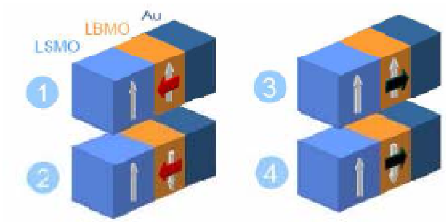
Magneto-electric Multiferroics

Eerenstein et al., Nature 442, 759 (2006).



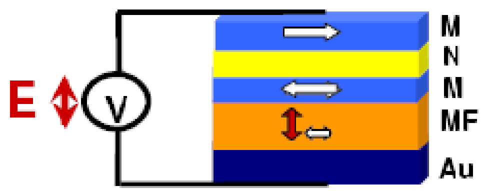
*antiferromagnets also accepted.

Small coupling – 4 state memory



1 (+P,+ M)
2 (+P,- M)
3 (-P,+ M)
4 (-P,- M)

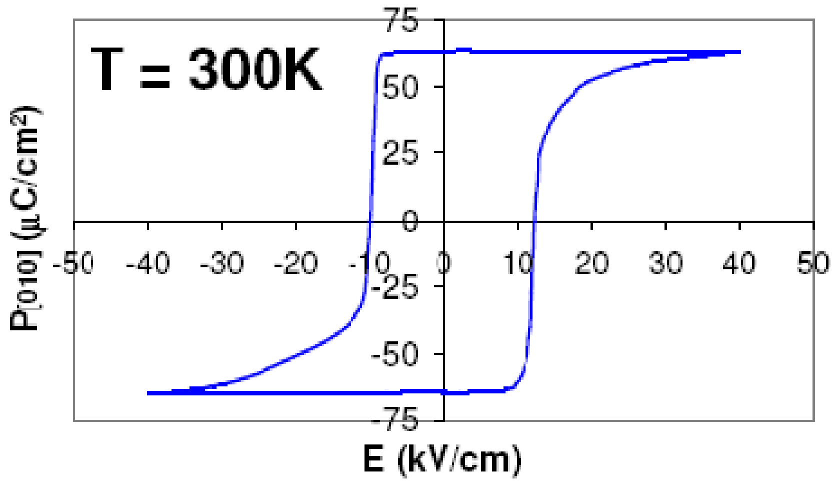
Large coupling – "E" write / "M" read



Ying & Yang

Ferroelectricity & Magnetism coexist

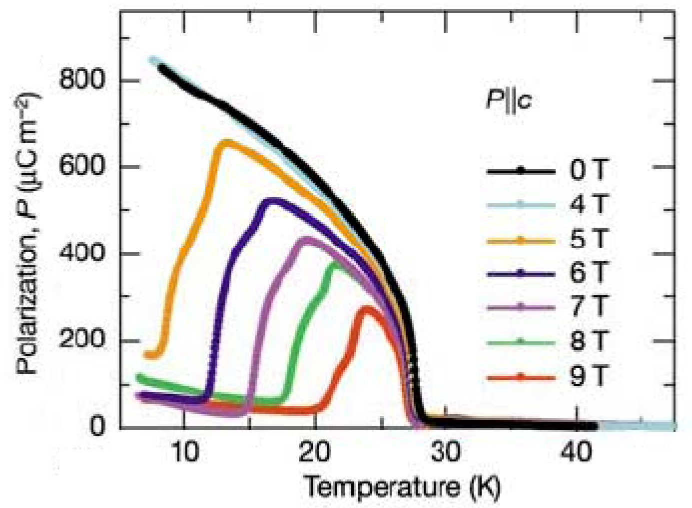
- ✓ $BiFeO_3$
- ✓ Large moments
- ✓ High temperature transitions
- ✓ Weak coupling



Lebeugle et al. APL 2007

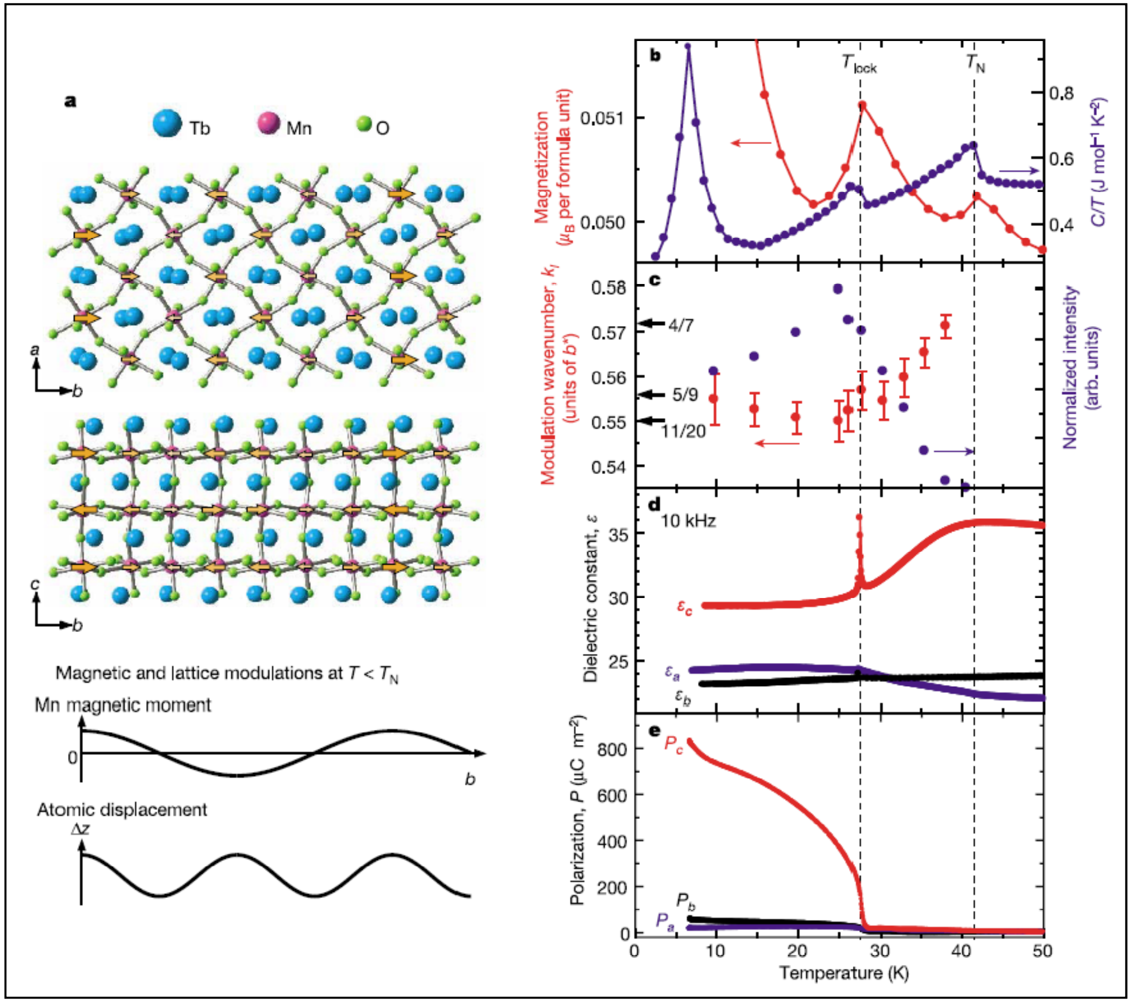
Magnetism causes Ferroelectricity

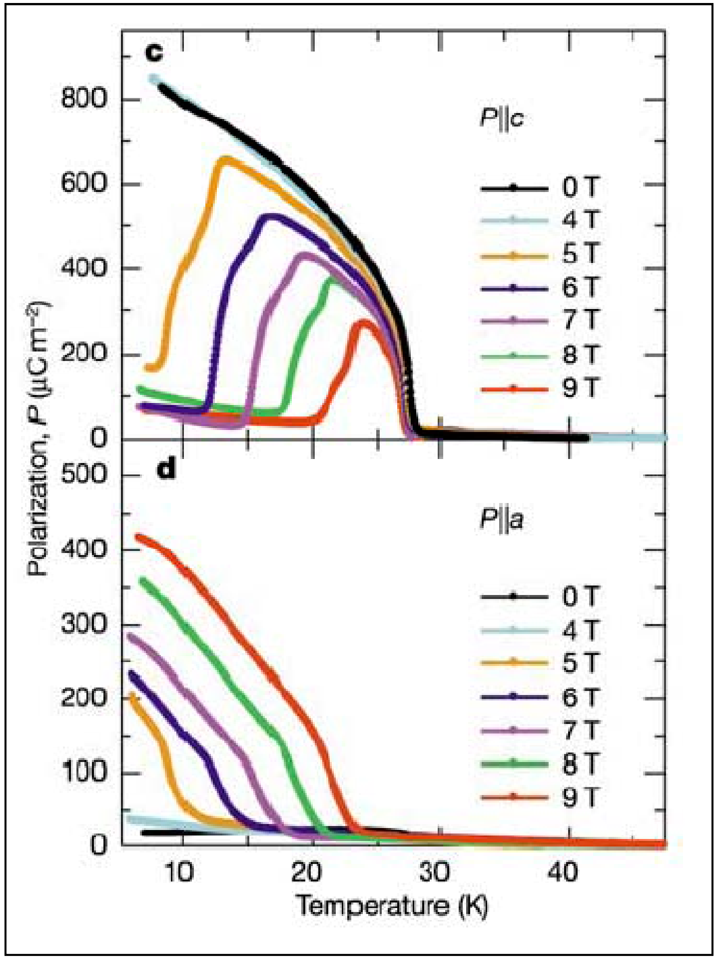
- ✓ $TbMnO_3$
- ✓ Small moments
- ✓ Low temperatures
- ✓ Strong coupling



Kimura, Nature 2003

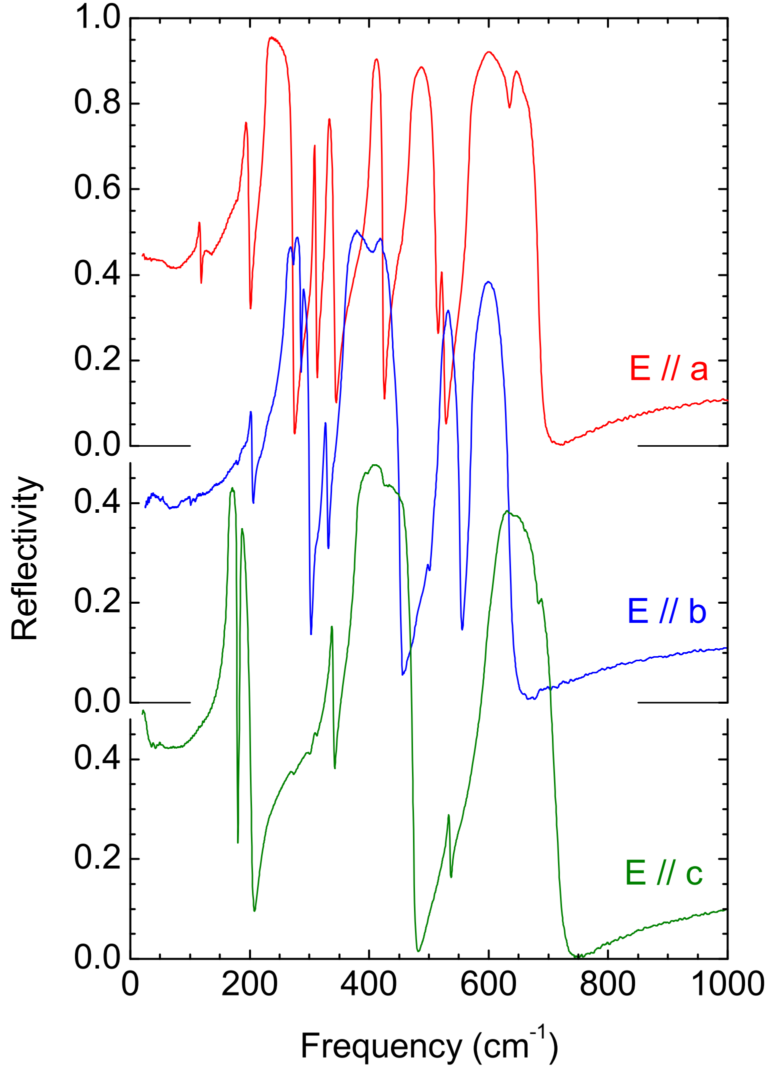
Yang ($TbMnO_3$) is more fun!

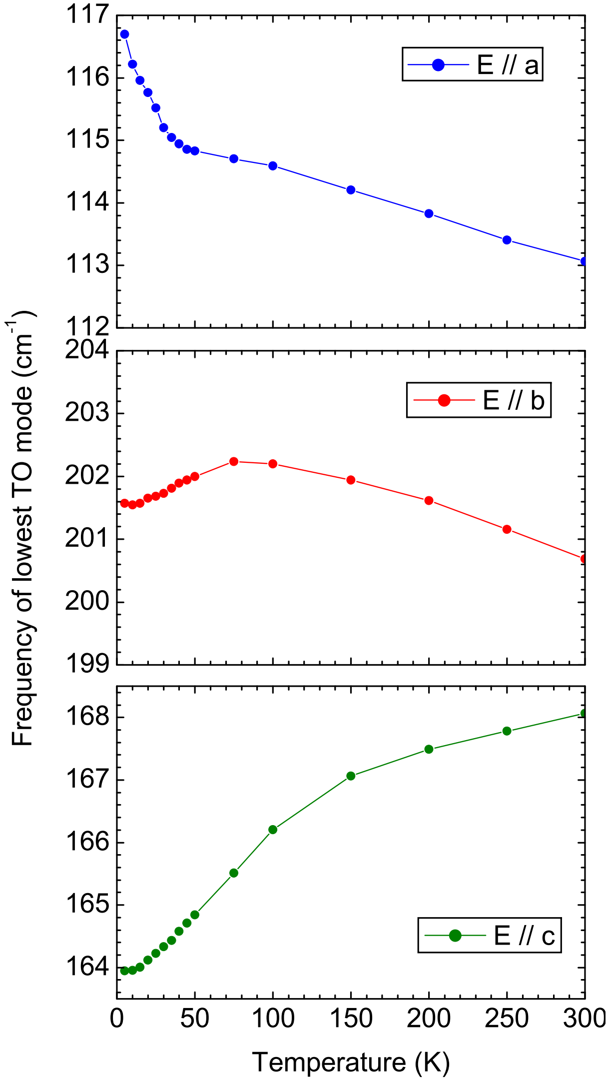




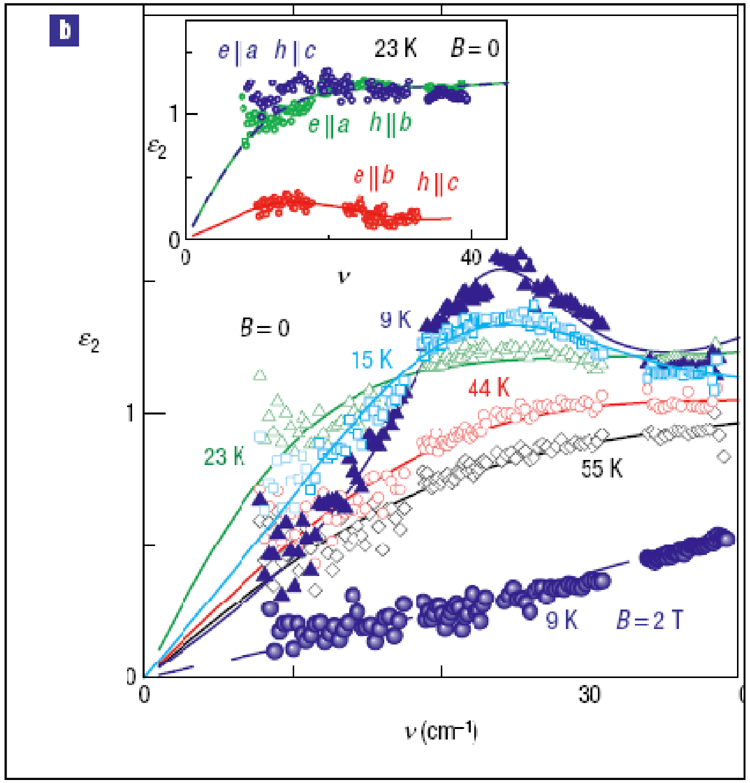
Kimura, Nature 2003.

$TbMnO_3$ Phonon Spectra (T = 5 K)

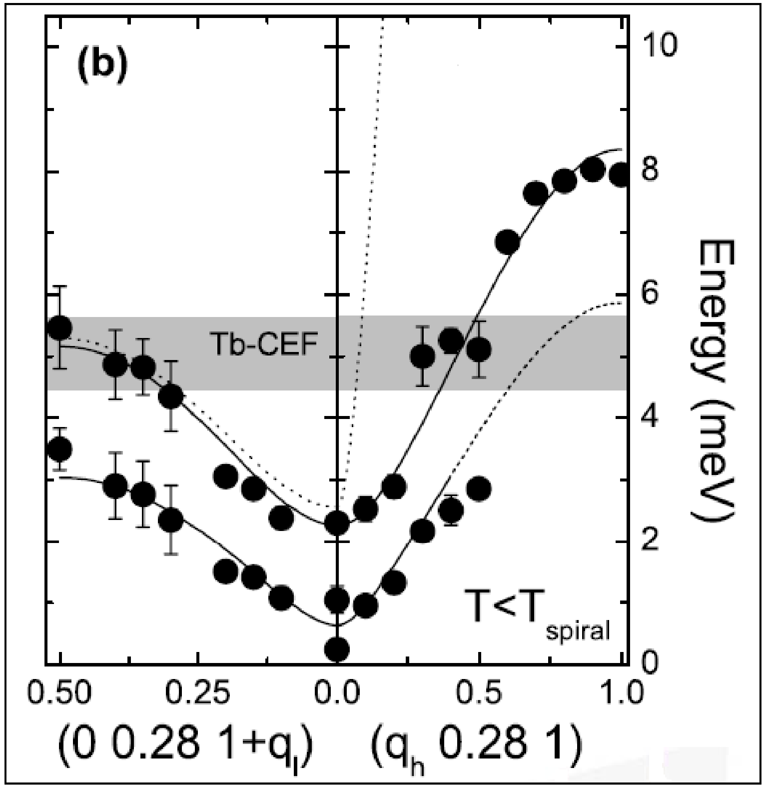




So, where is the action?



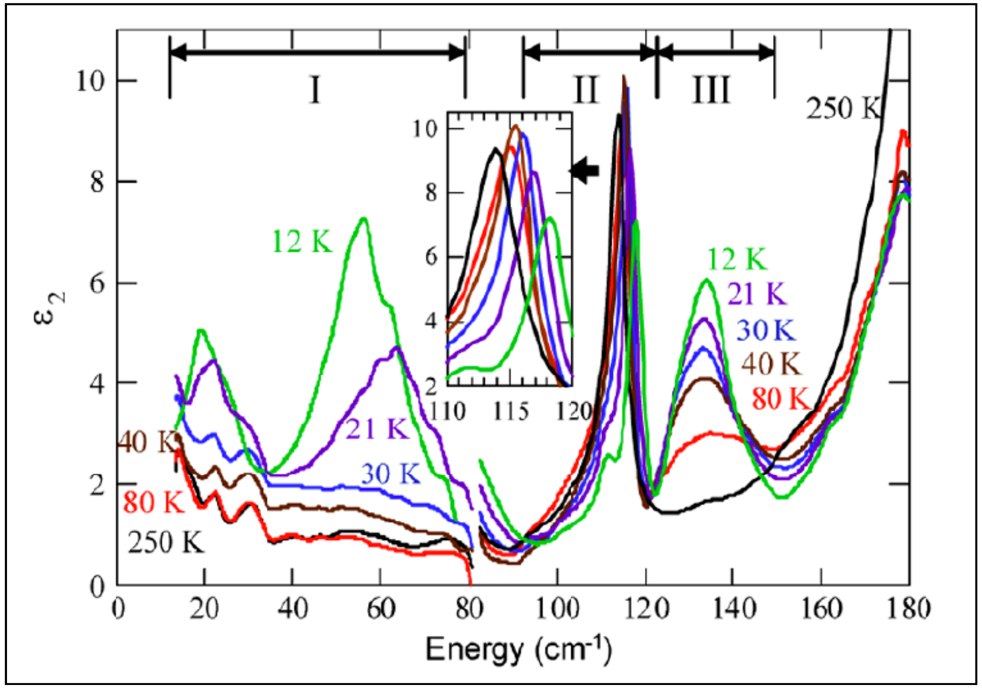
Pimenov et al., Nat. Phys, 2006.

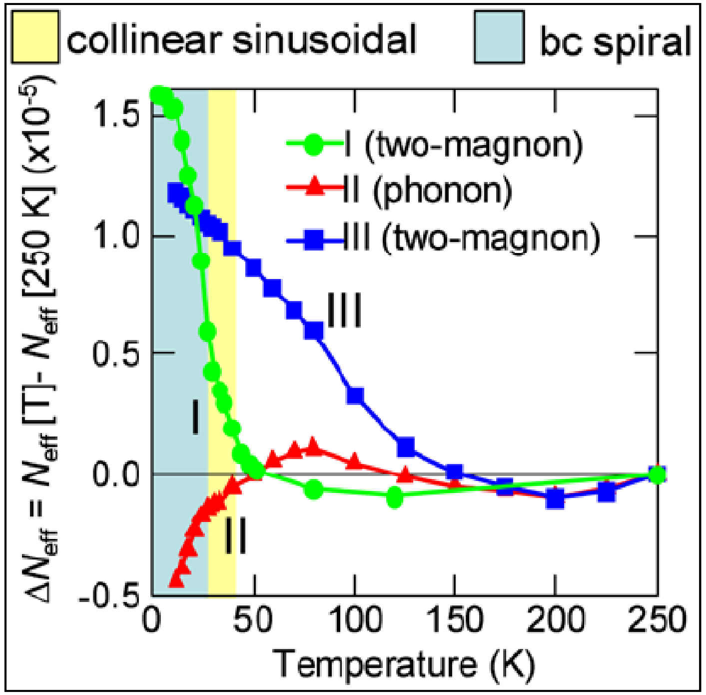


Senff et al. PRL 2007

- ✓ Magnetic excitation
- ✓ Activated by electric field of light only
- ✓ Suppressed by an external magnetic field

Magnons and Phonons

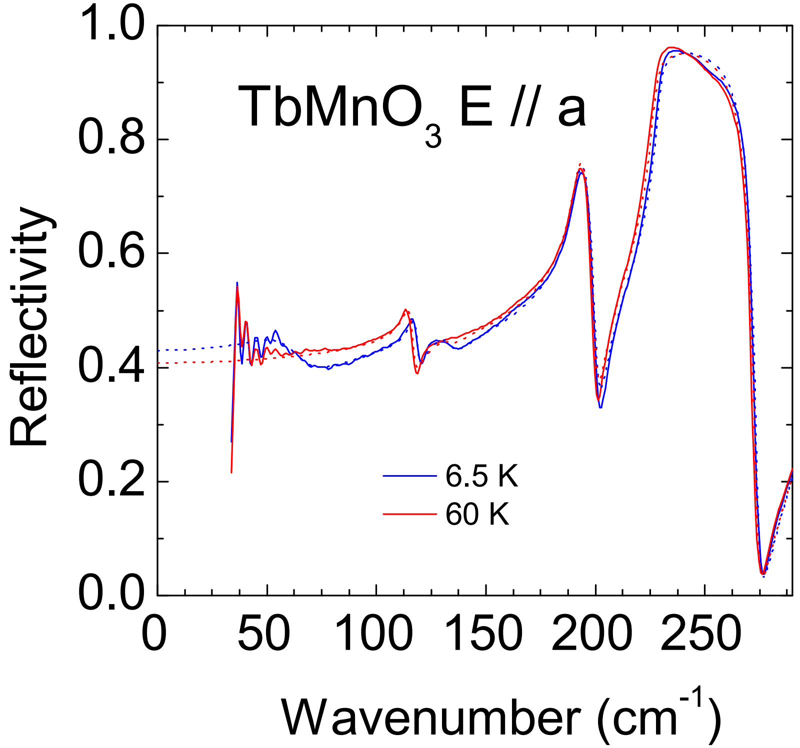


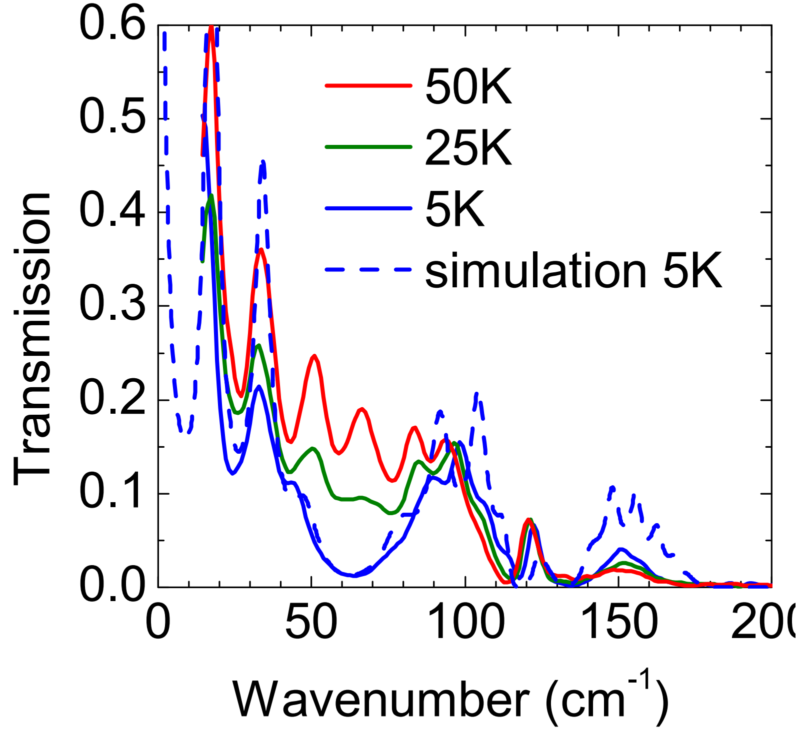


Stronger electromagnon at 60 cm^{-1} coupled to phonon at 110 cm^{-1}

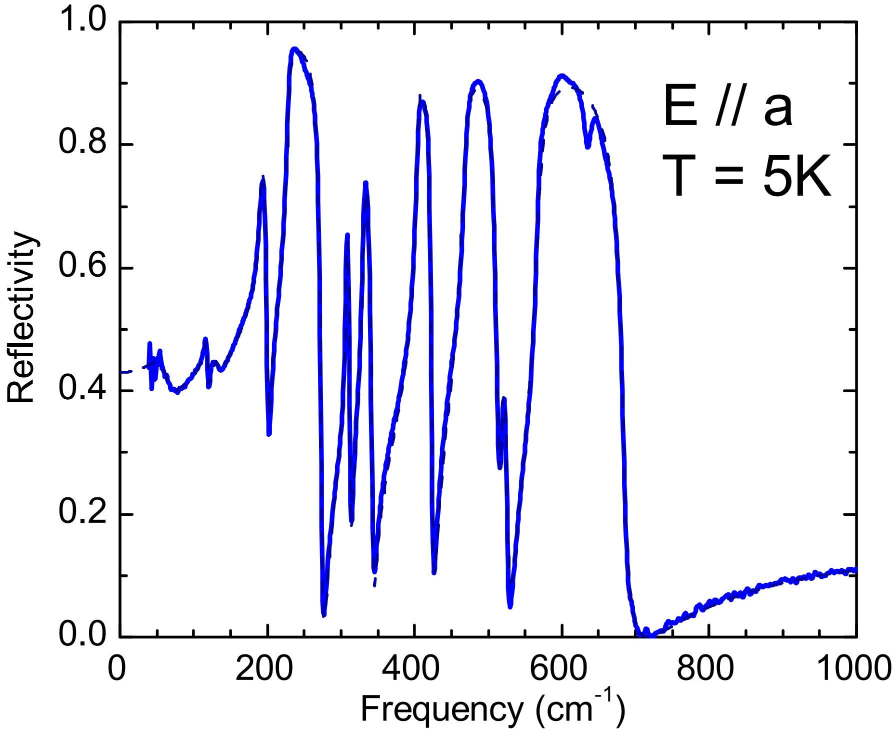
Takahashi *et al.* PRL **101**, 187201 (2008)

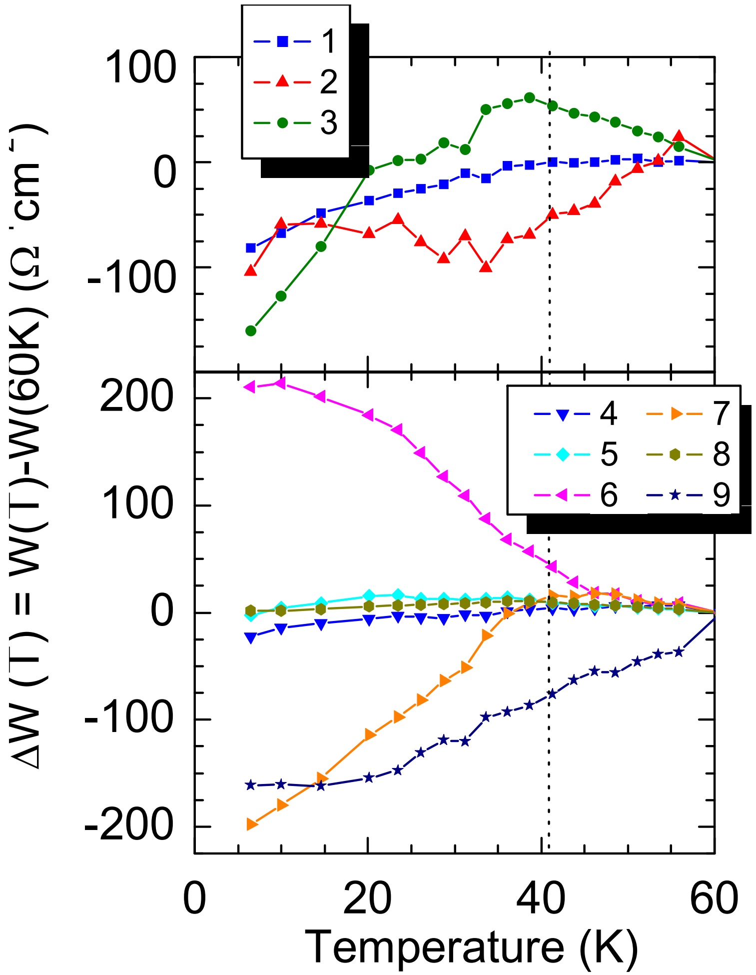
Phonon and magnon optical response



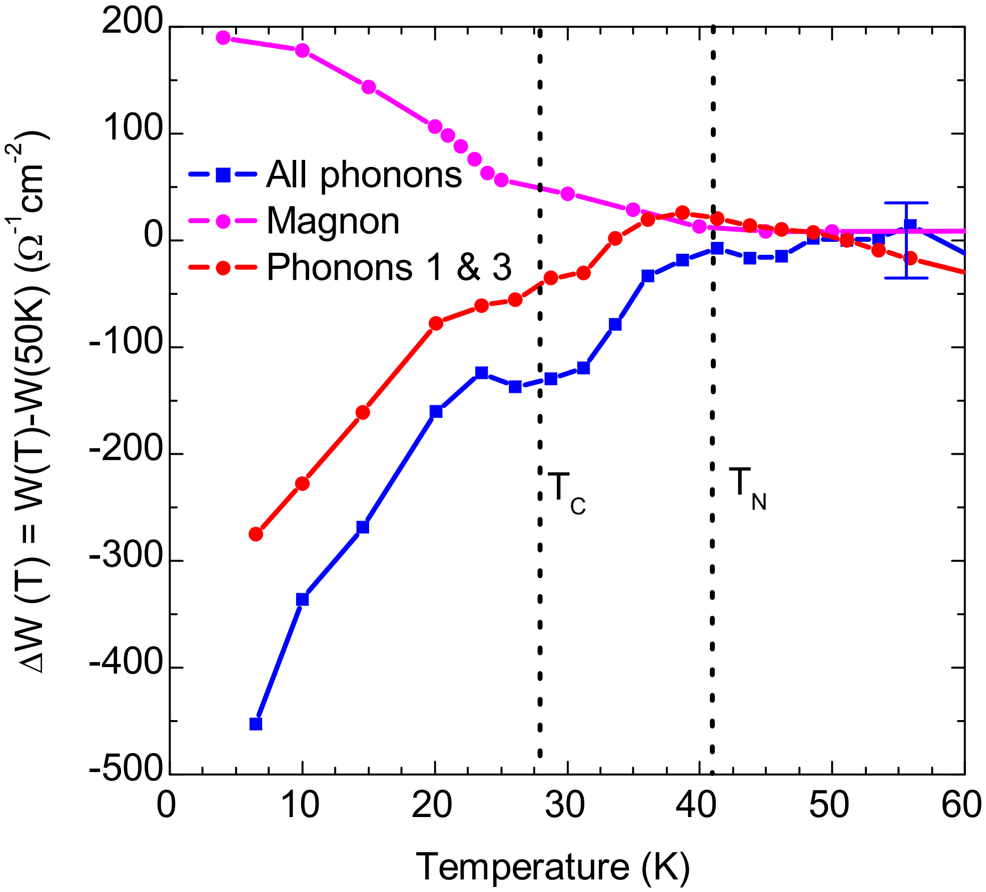


Phonon spectral weigths



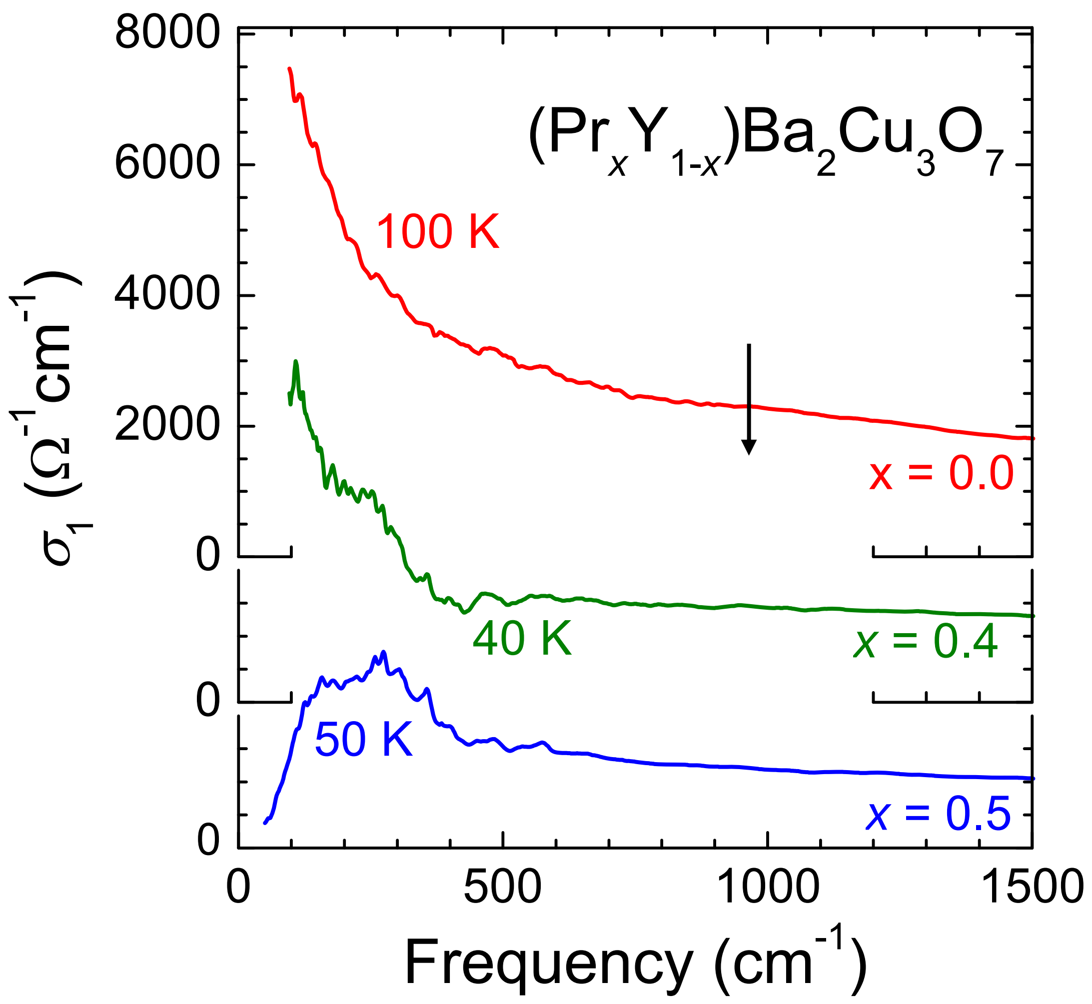


The electromagnon is built from two phonons



DiSordEred MEdiA

Disorder in (Y,Pr)$Ba_2Cu_3O_7$

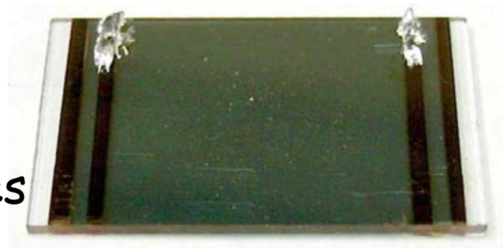


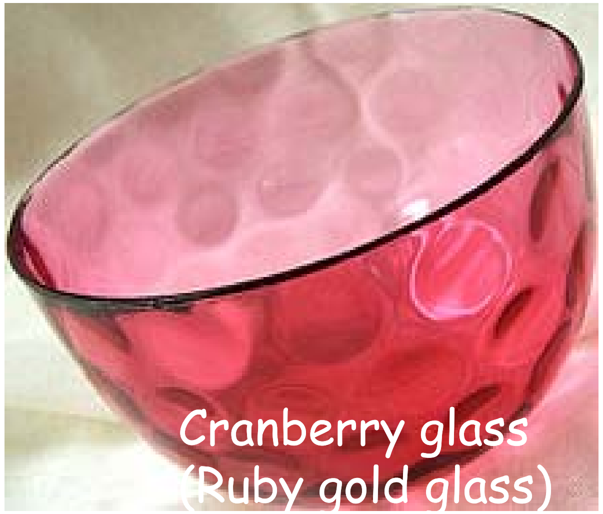
- ✓Pr is an underdoping agent
- ✓Empties CuO2 planes
- ✓Localizes charges along chains

Lobo et al. PRB 65, 104509 (2002).

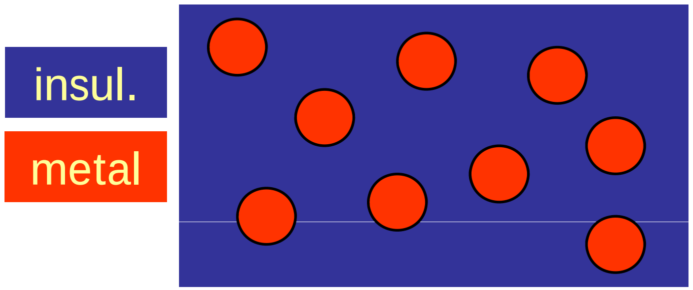
Effective Medium Theories

Gold evaporated
onto hot (600K)
sapphire substrates

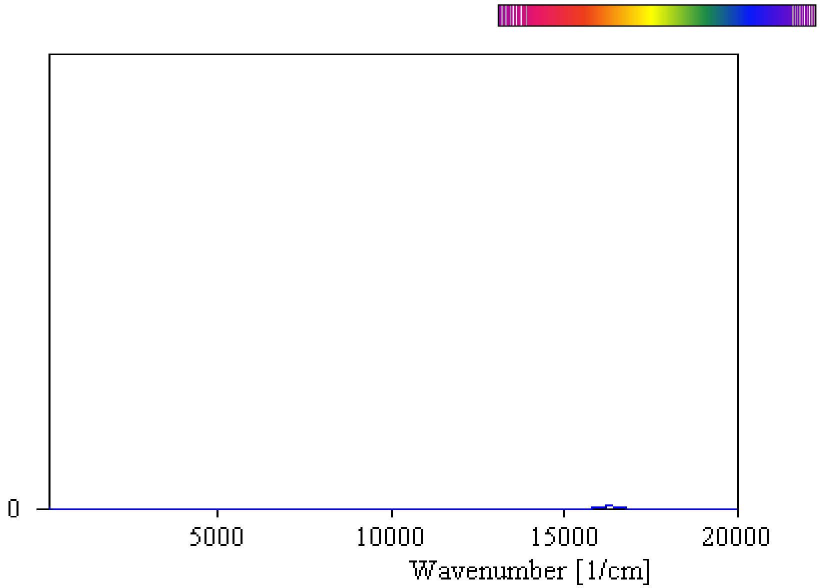




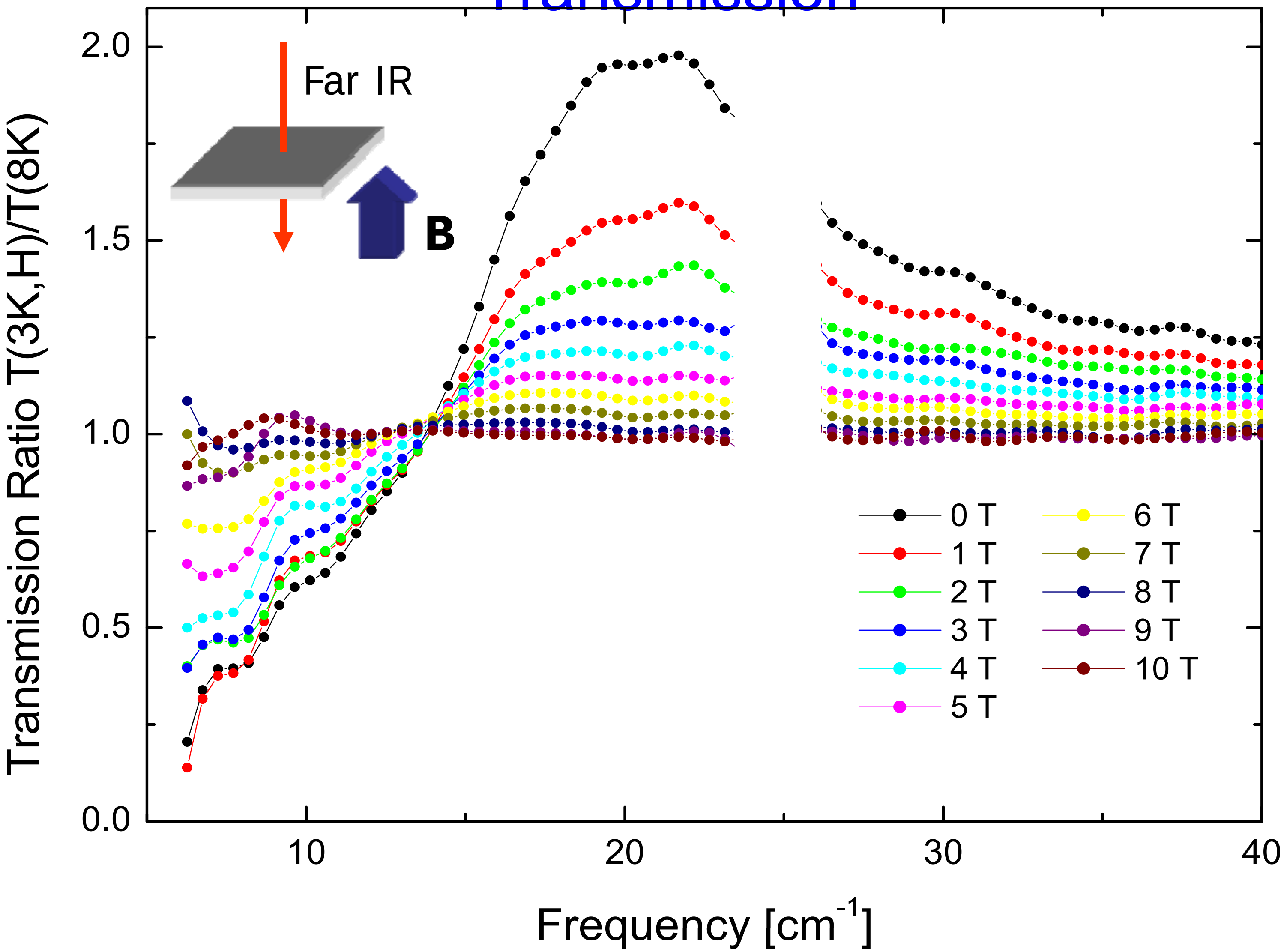
Maxwell-Garnett Theory



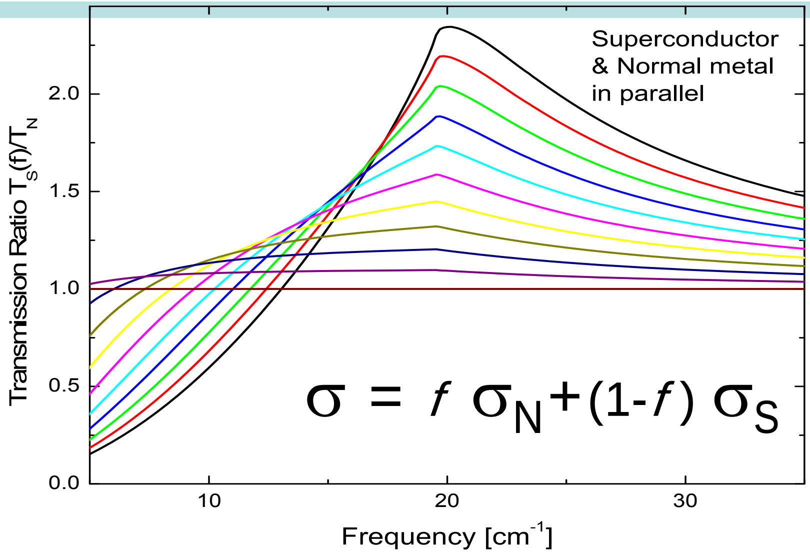
J. C. *Maxwell Garnett, Colours in Metal Glasses and in Metallic Films*, Philos. Trans. R. Soc. 203, 385 (1904).

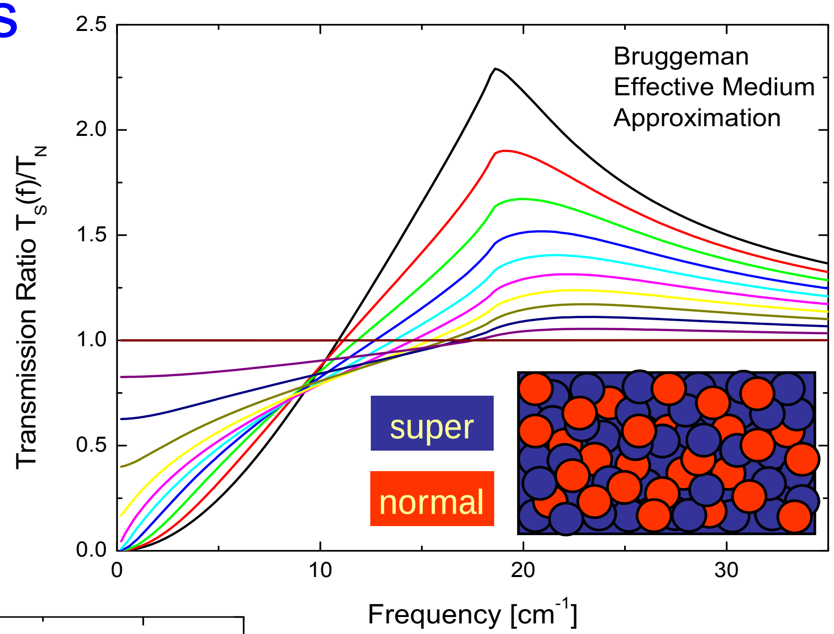


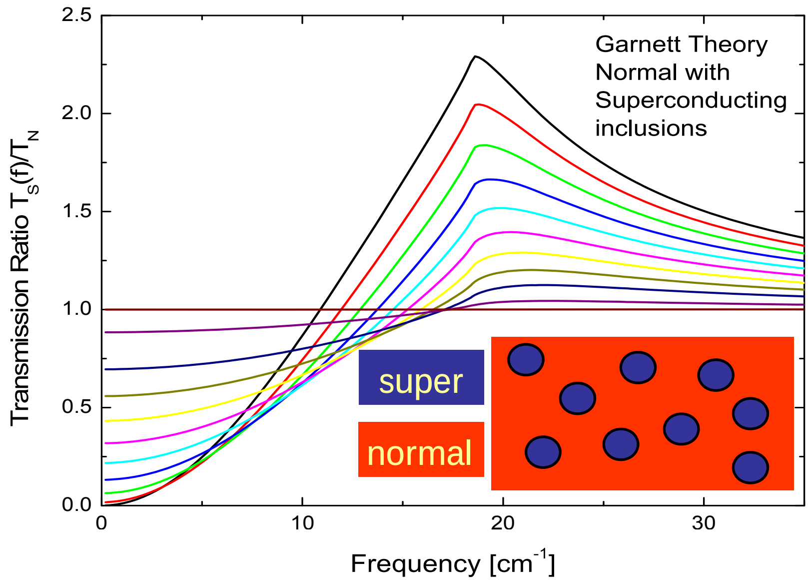
Superconducting MoGe: Field Dependent Transmission



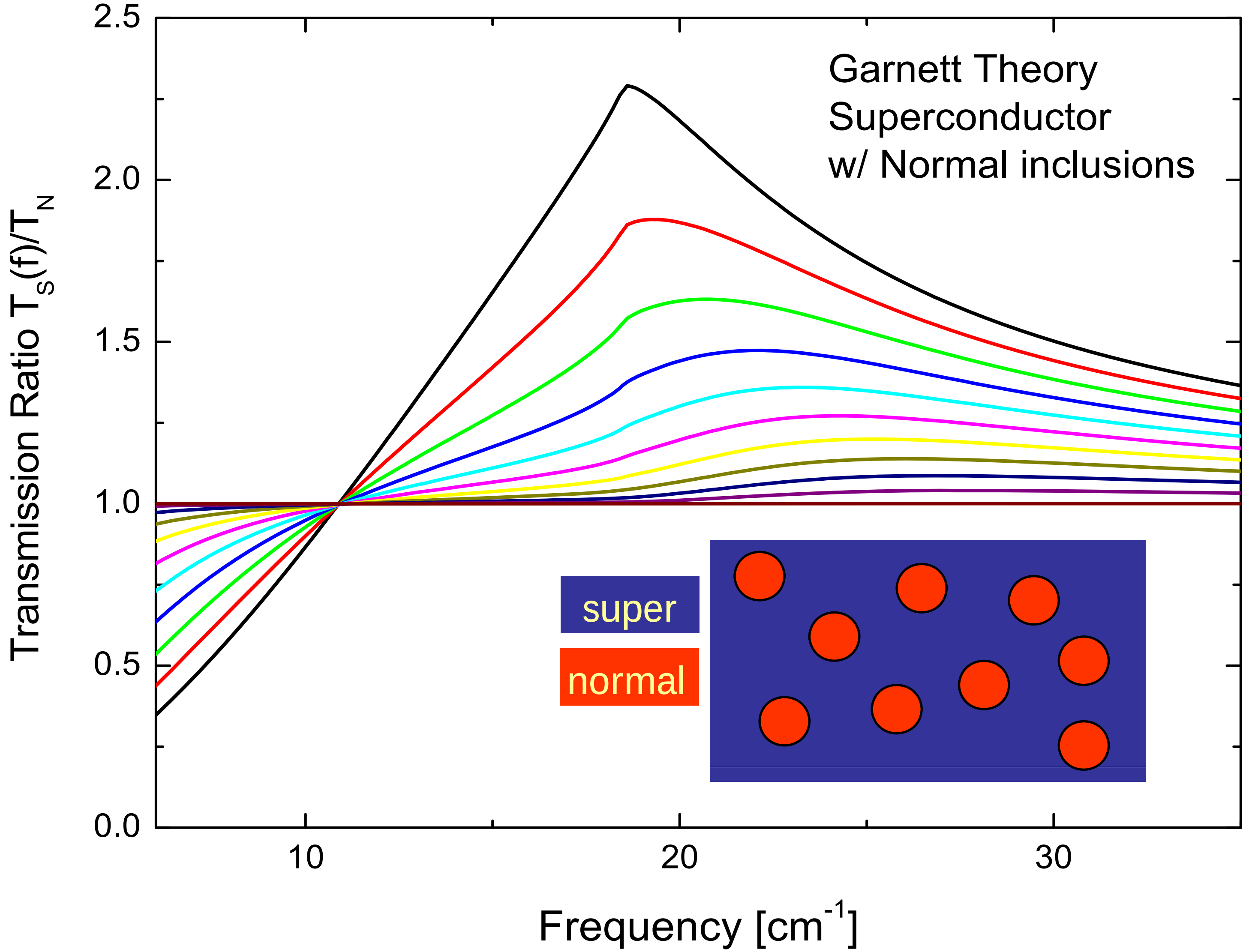
Effective Medium Models for a Superconductor with Vortices







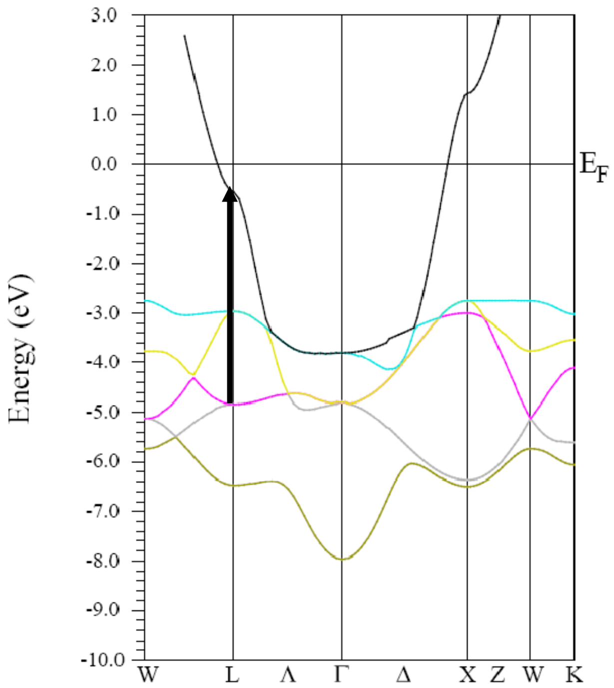
Effective Medium Models for a Superconductor with Vortices

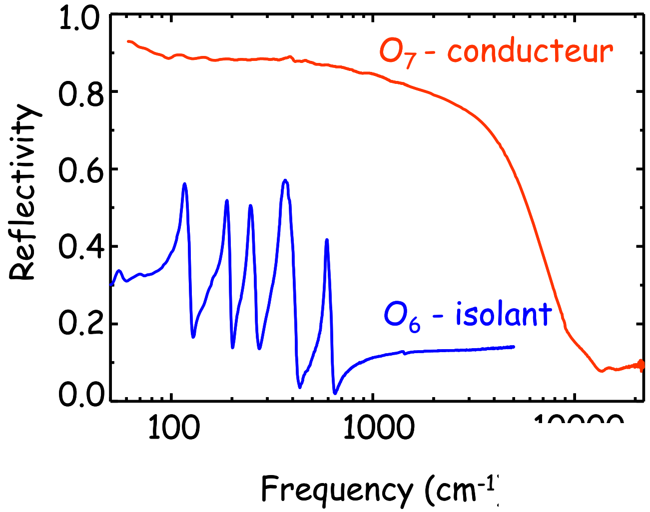


Summary

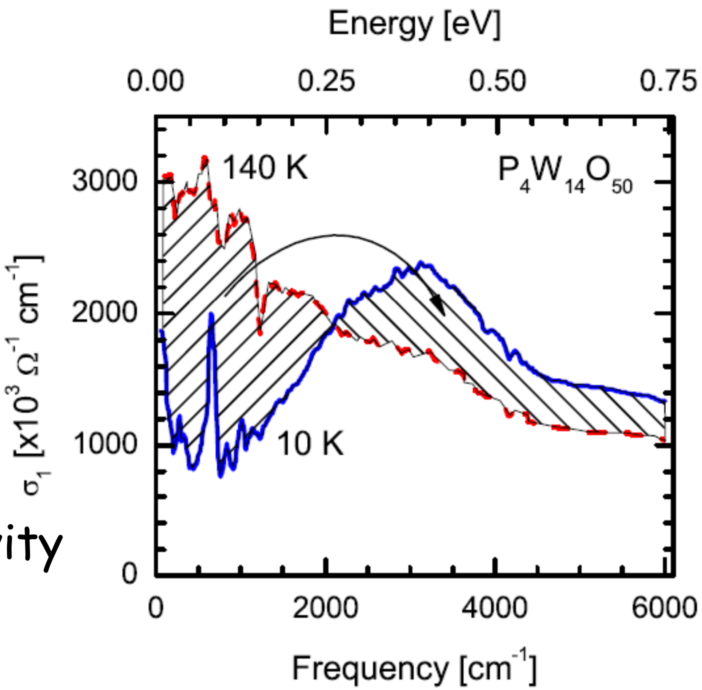
$$\Delta\vec{k} \approx 0$$

Momentum averaged technique





Optics is mostly an electrical measurement



The optical conductivity specgtral weight is always conserved

Rappels

- ✓ Technique moyennée en impulsion
- ✓ Mesure de la conductivité électrique aux hautes fréquences
- ✓ Pic à fréquence nulle = charge mobile
- ✓ Pic à fréquence finie = charge localisée
- ✓ Accès aux états électroniques et à leur distribution en énergie
- ✓ Règle de somme de la conductivité