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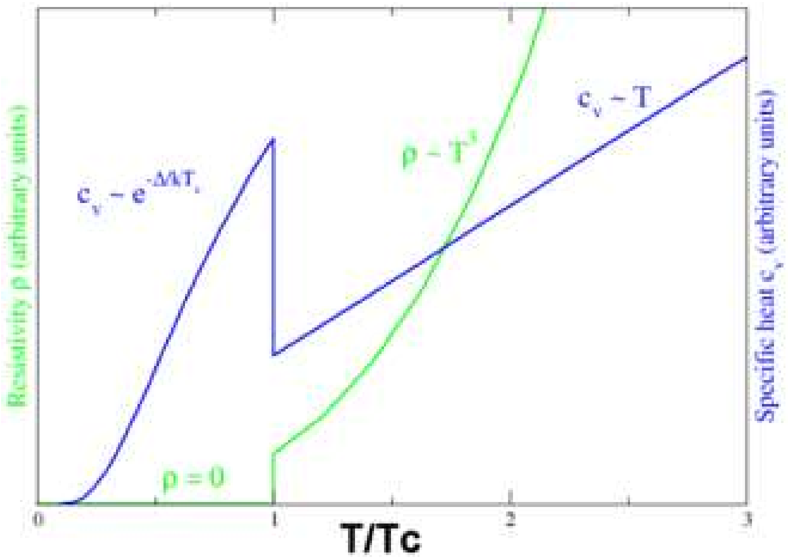
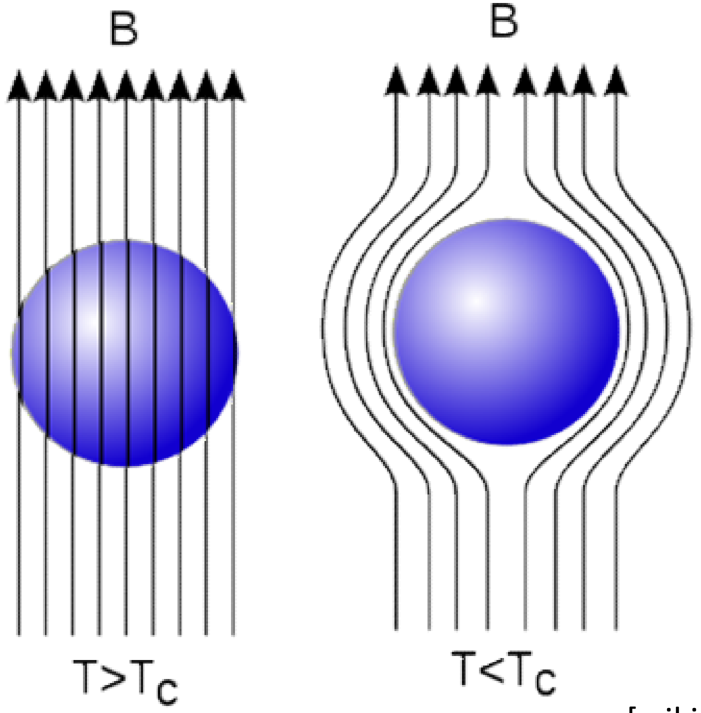
Optical Spectroscopy in Materials Science
Optical properties of interacting systems

Superconductors

Perfect conductivity
(exponentially vanishing specific heat):



Meissner effect:



[wikipedia]

Superconductivity vs. perfect conductors

Perfect conductor: $j = nqv$

$$m\dot{v} = -m\frac{v}{\tau} + qE$$

->0

$$\frac{\partial j}{\partial t} = nq\frac{q}{m}E$$

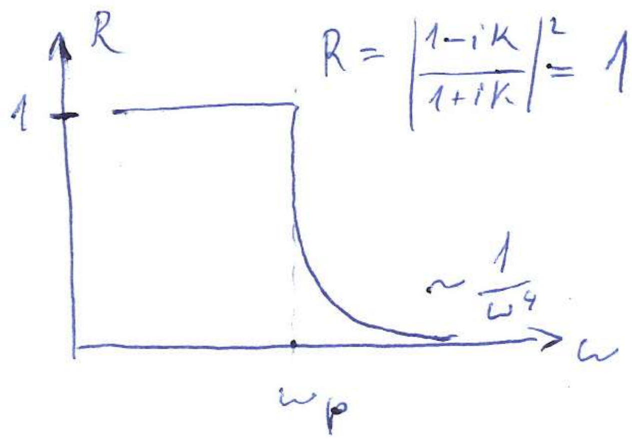
$$\frac{\partial}{\partial t}(\nabla \times j) = -nq\frac{q}{m}\frac{\partial B}{\partial t}$$

$$\frac{\partial}{\partial t}\left(\nabla \times j + \frac{nq^2}{m}B\right) = 0$$

$$\sigma = \frac{nq^2}{m}\tau\left(\frac{1}{1+\omega^2\tau^2} + i\frac{\omega\tau}{1+\omega^2\tau^2}\right)$$

$$\sigma \to \frac{nq^2}{m}\left(\pi\delta(\omega) + i\frac{1}{\omega}\right)$$

- Drude peak narrows to delta peak
- As ε<0 R = 1 (below $\omega<\omega_p$)



London equations:

$$\frac{\partial j}{\partial t} = \frac{nq^2}{m}E$$

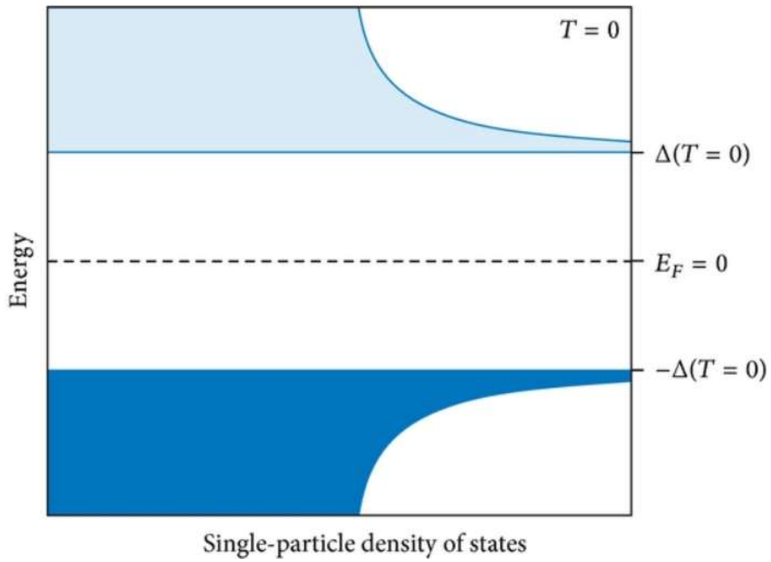
$$\nabla \times j + \frac{nq^2}{m}B = 0$$

$$\frac{\partial j}{\partial t} = -\frac{nq^2}{m}\frac{\partial A}{\partial t}$$

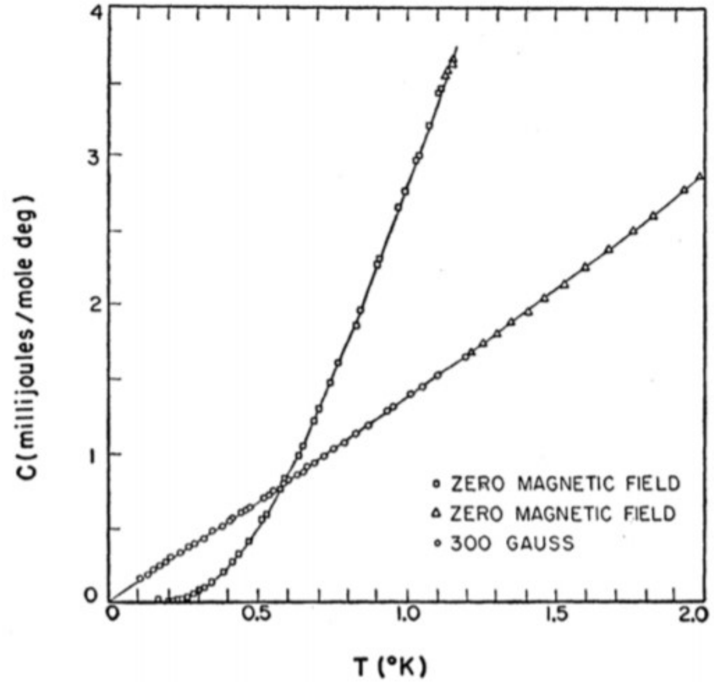
$$j = -\frac{nq^2}{m}A$$

BCS theory: gapped electronic excitations

BCS theory:



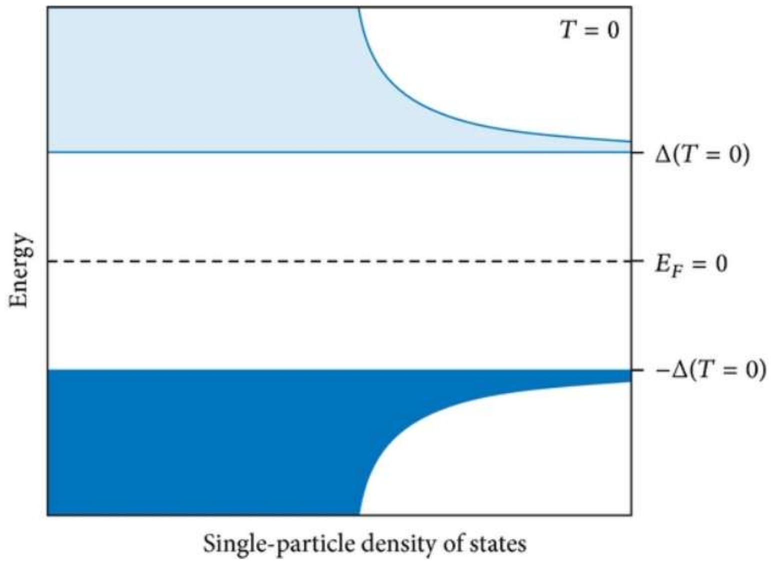
Norman E. Phillips. Heat Capacity of Aluminum between 0.1 K and 4.0 K. *Phys. Rev.*, 114(3):676–685, May 1959.

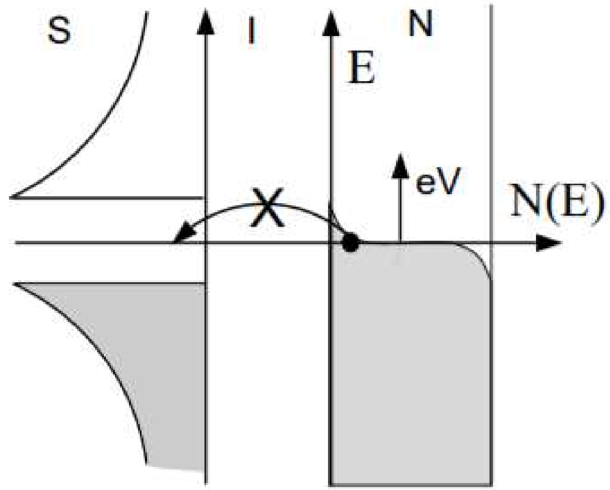


M. Dressel, Advances in Condensed Matter Physics 2013, 104379

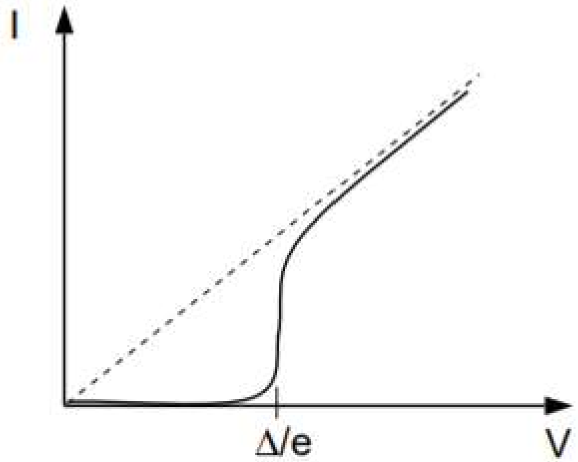
BCS theory: gapped electronic excitations

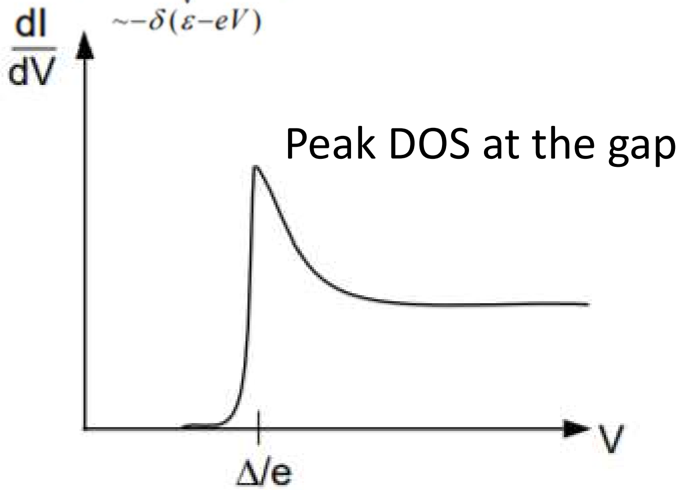
BCS theory:





Applied solid state physics: $\frac{\mathrm{d}I}{\mathrm{d}V} \sim T \cdot g_{\mathrm{N}}(\varepsilon_{\mathrm{F}}) \int \mathrm{d}\varepsilon\, g_{\mathrm{S}}(\varepsilon) \underbrace{f_{\mathrm{N}}^{'}(\varepsilon - eV)}_{\sim -\delta(\varepsilon - eV)} \sim T \cdot g_{\mathrm{N}}(\varepsilon_{\mathrm{F}}) g_{\mathrm{S}}(\varepsilon - eV)$

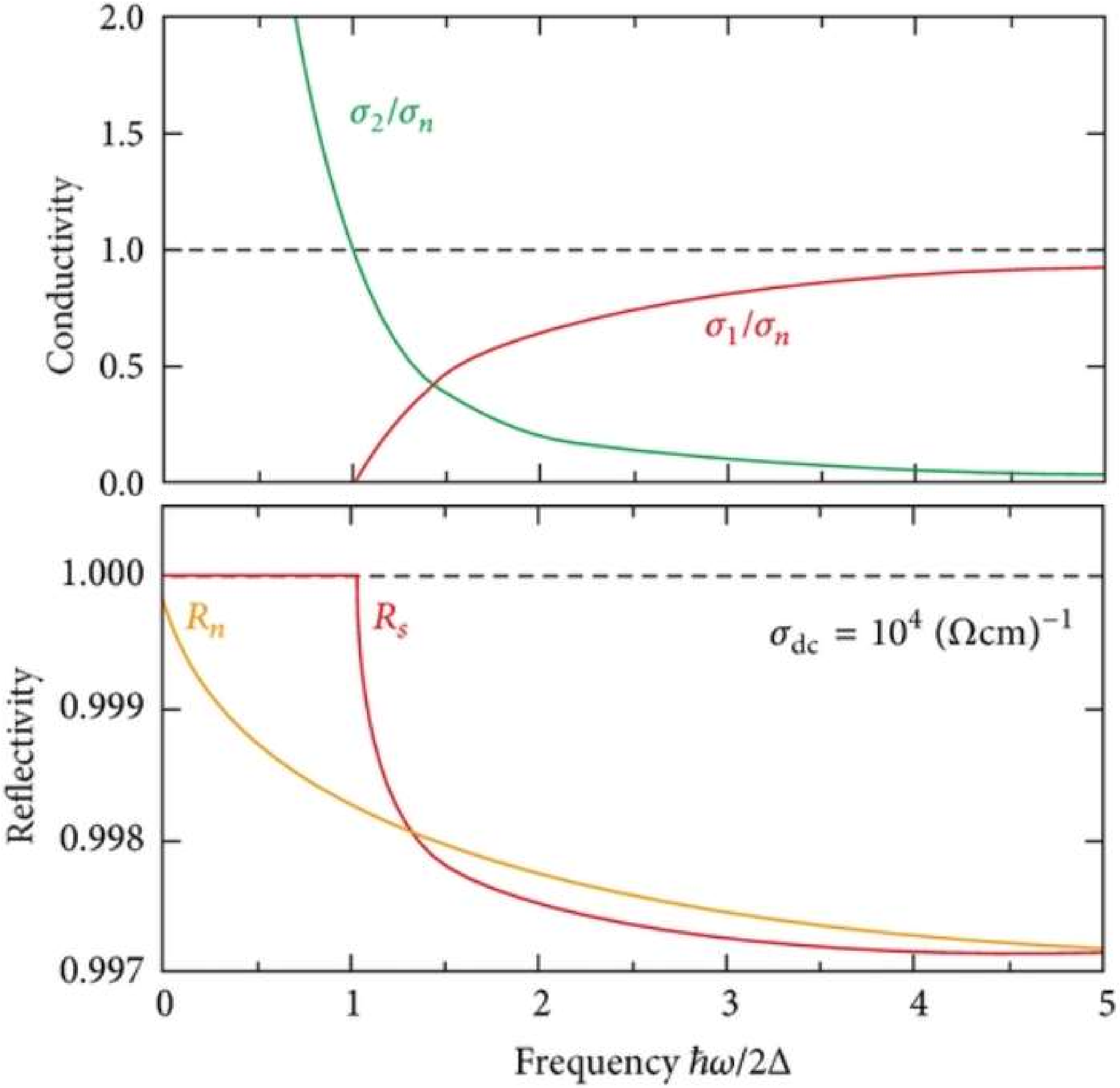




M. Dressel, Advances in Condensed Matter Physics 2013, 104379

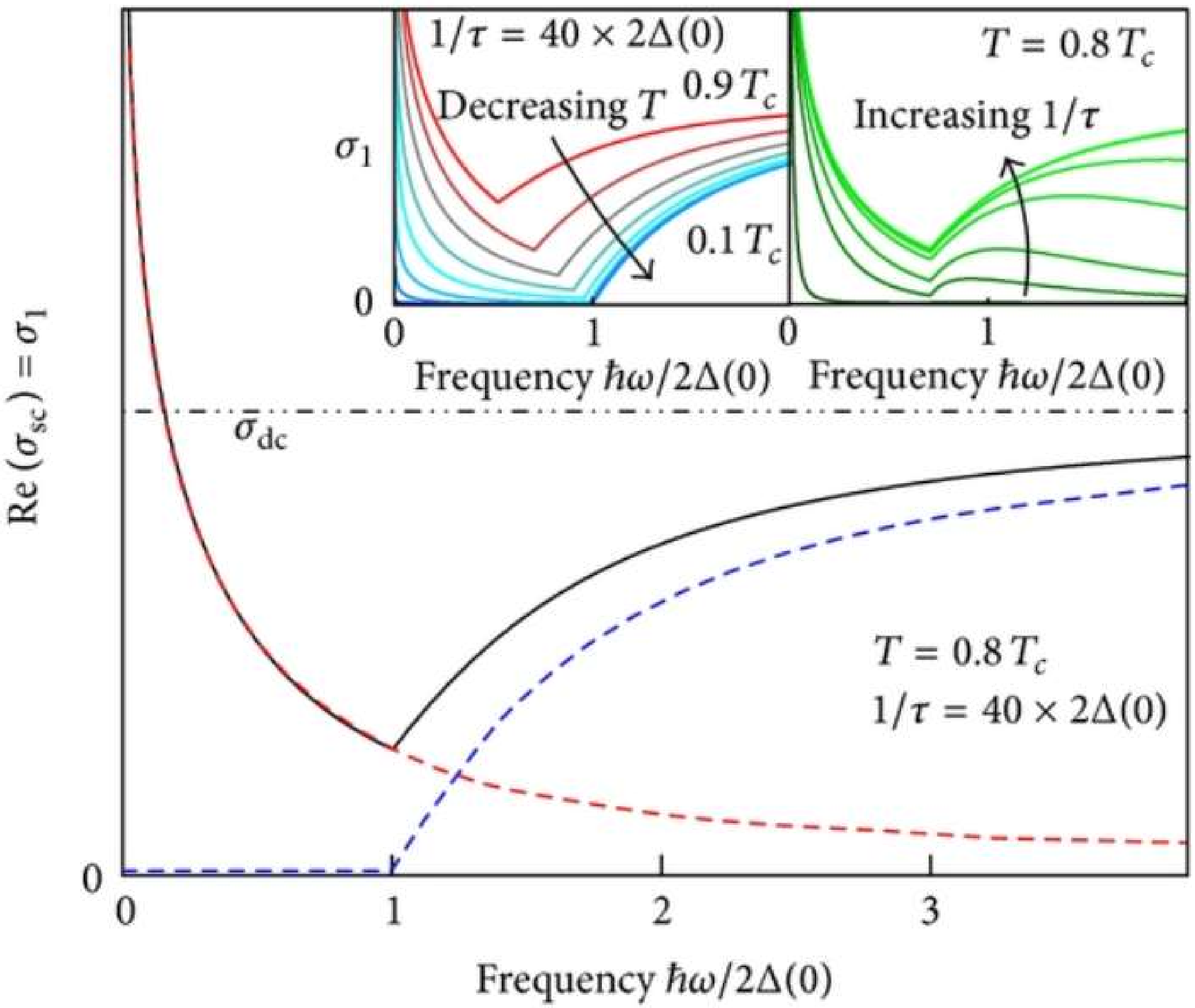
BCS theory: optical excitations

- Matrix element: coherence effect
- Conservation of spectral weight



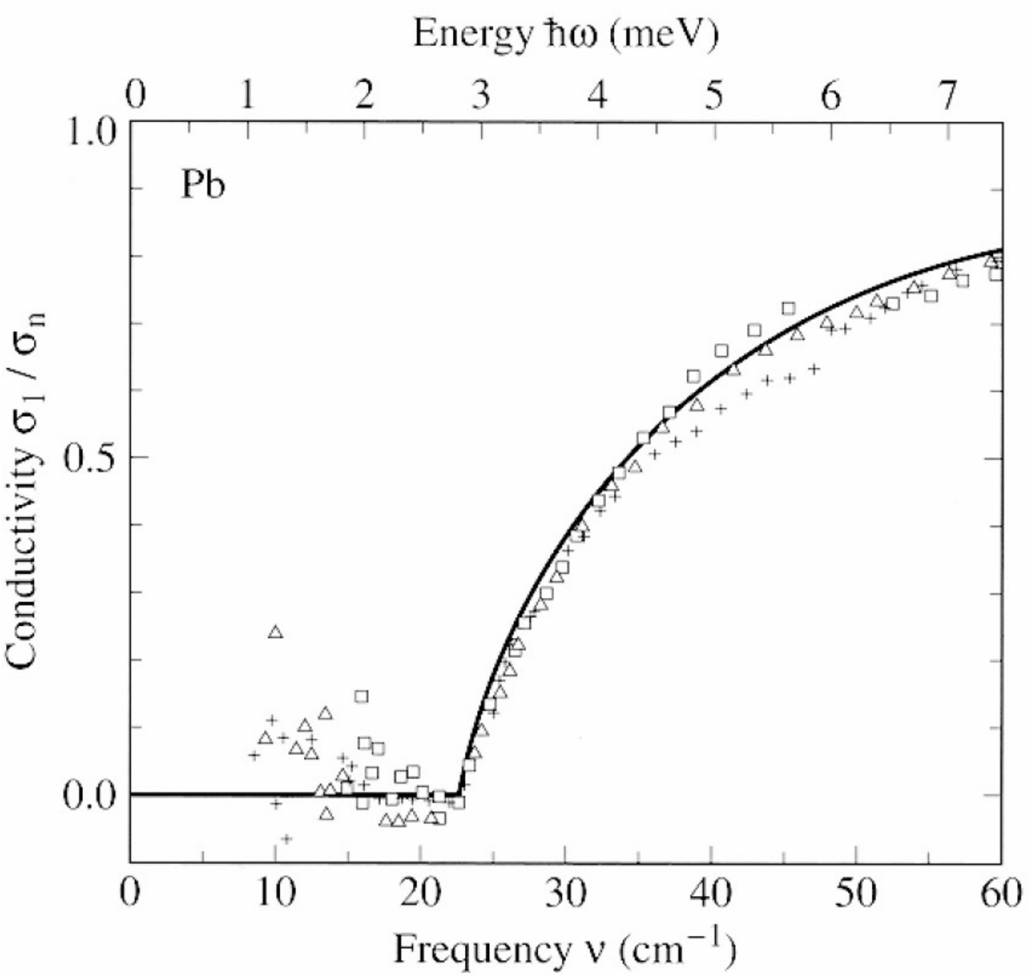
Mattis-Bardeen equation: $$\frac{\sigma_1(\omega,T)}{\sigma_n} = \frac{2}{\hbar\omega}\int_{\Delta}^{\infty}\frac{\left[f(\mathscr{E}) - f(\mathscr{E}+\hbar\omega)\right]\left(\mathscr{E}^2+\Delta^2+\hbar\omega\mathscr{E}\right)}{\left(\mathscr{E}^2-\Delta^2\right)^{1/2}\left[(\mathscr{E}+\hbar\omega)^2-\Delta^2\right]^{1/2}}d\mathscr{E}$$

$$+\frac{1}{\hbar\omega}\int_{\Delta-\hbar\omega}^{-\Delta}\frac{\left[1-2f(\mathscr{E}+\hbar\omega)\right]\left(\mathscr{E}^2+\Delta^2+\hbar\omega\mathscr{E}\right)}{\left(\mathscr{E}^2-\Delta^2\right)^{1/2}\left[(\mathscr{E}+\hbar\omega)^2-\Delta^2\right]^{1/2}}d\mathscr{E}$$

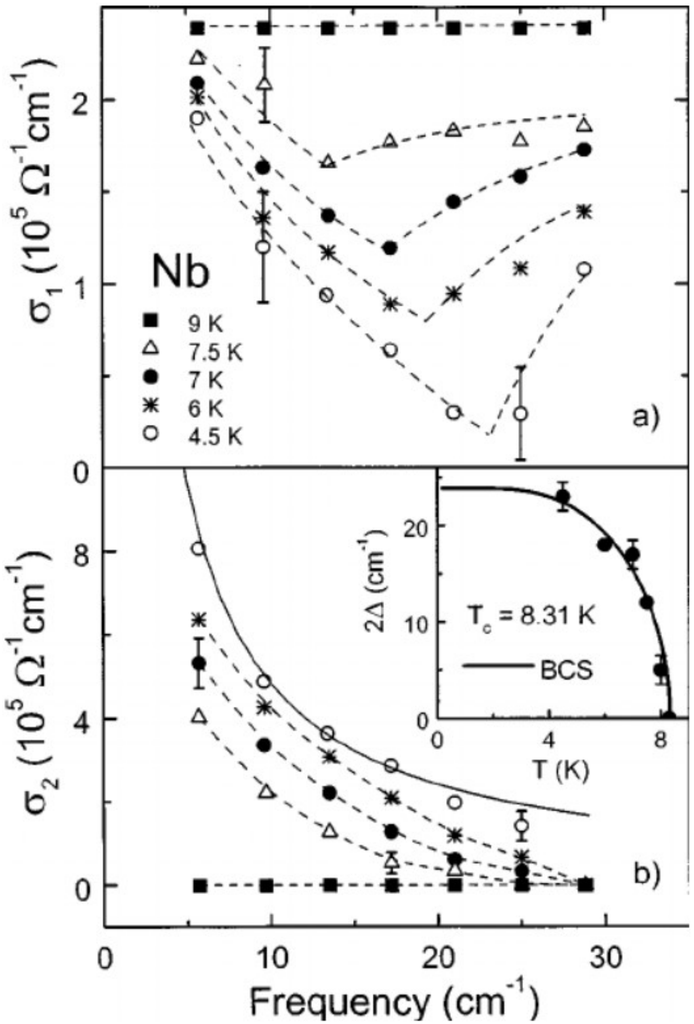


Optical conductivity in experiments

Pb at 2K
Phys. Rev. **165** 588 (1968).

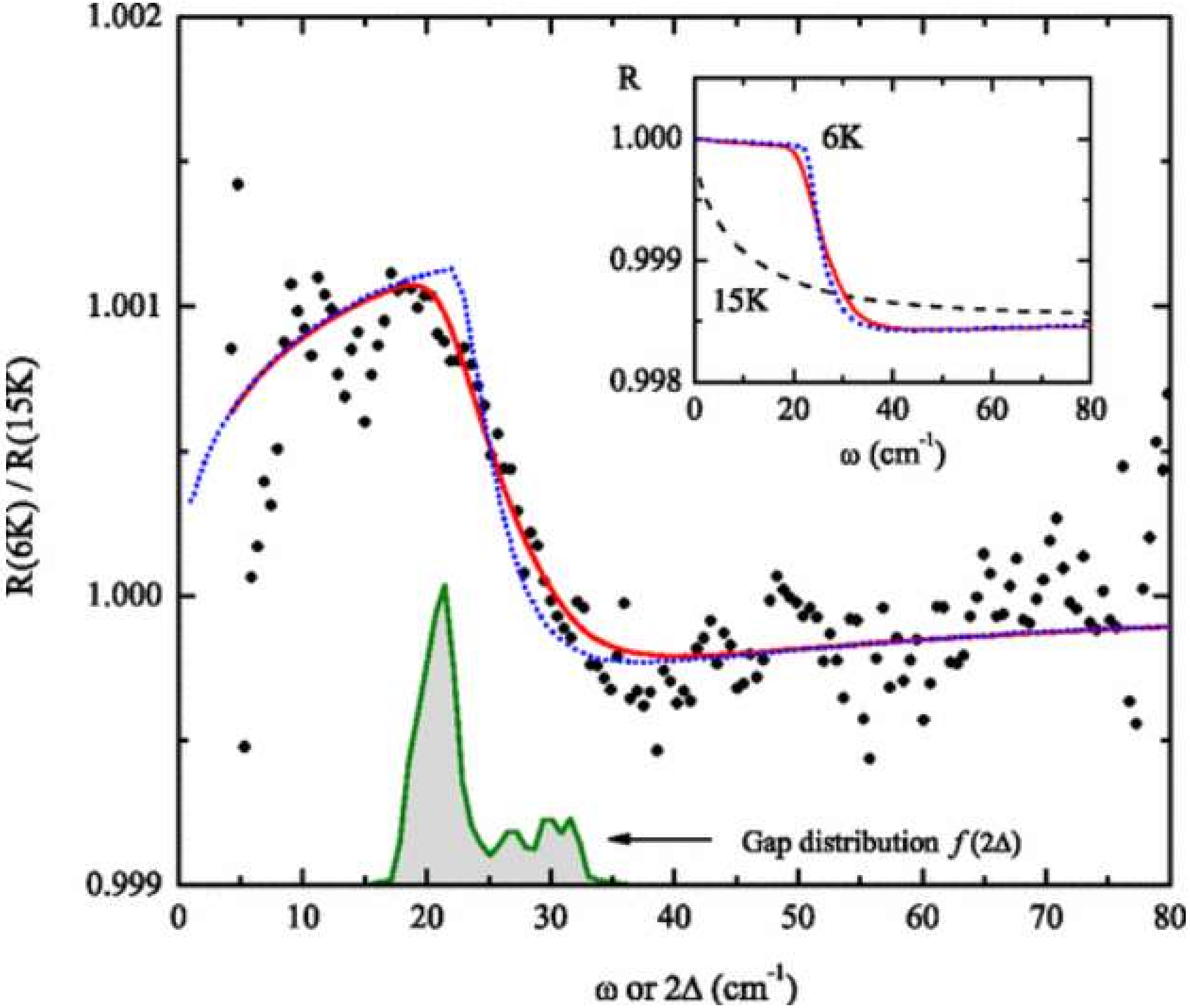


Nb
Phys. Rev. B **57** 14416 (1998).



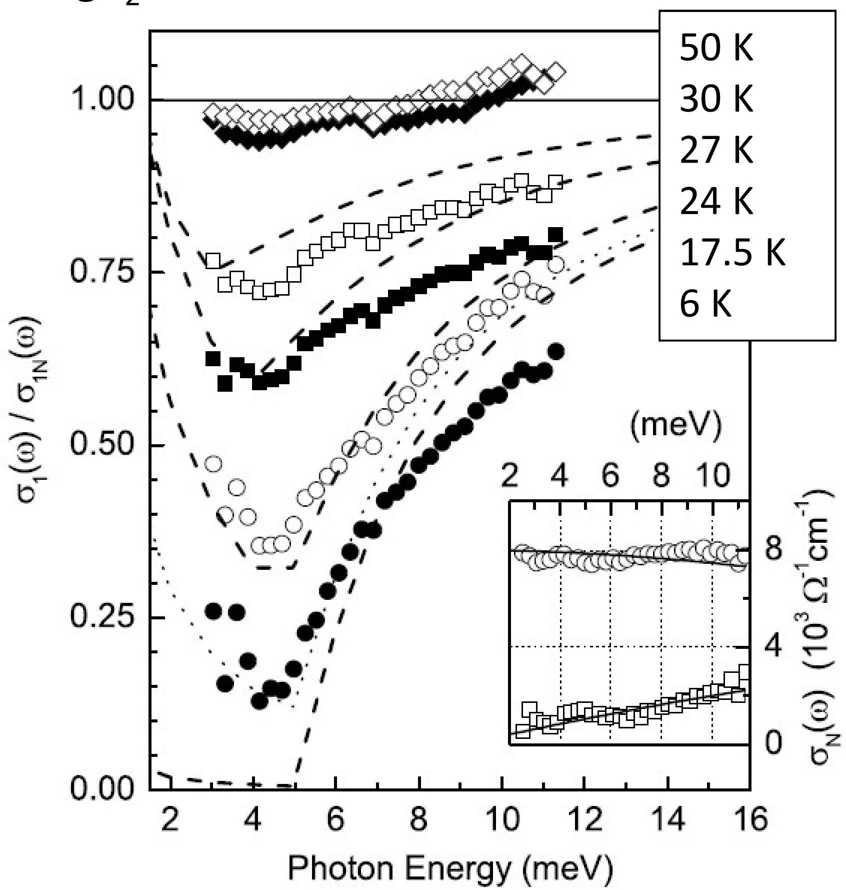
Optical conductivity in experiments

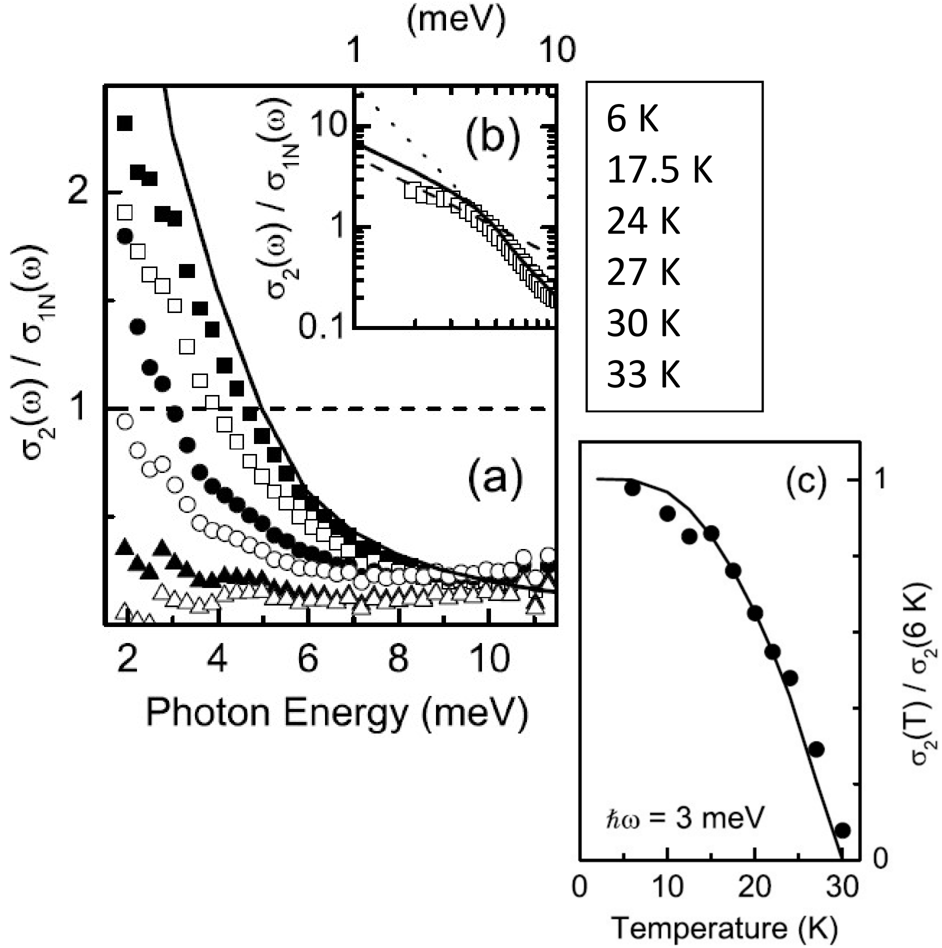
CaC_6
Phys. Rev. B **78**, 041404(R) (2008).



Optical conductivity in experiments

MgB_2





BCS fit: $2\Delta_o=5$ meV

Weak coupling limit: $2\Delta_o=3.5k_BT_C=9$ meV

Gap alatt kvázirészecske gerjesztés

R. A. Kaindl et al., *Phys. Rev. Lett.* **88** 027003 (2002)

Length scales in superconductors

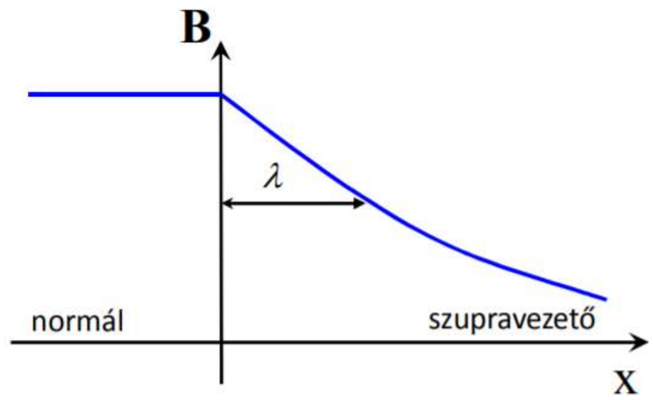
London equations:

$$\nabla \times j + \frac{n_S q^2}{m} B = 0$$

$$\nabla \times \nabla \times j + \frac{n_S q^2}{m} \mu_0 j = 0$$

London length:

$$\lambda = \sqrt{\frac{m^*}{\mu_0 n_s e^{*2}}}$$



Skin depth:

$$\delta \approx \sqrt{\frac{m}{\mu_0 n q^2} \frac{2}{\omega \tau \mu'}} >> \lambda_L \qquad \omega\tau << 1$$

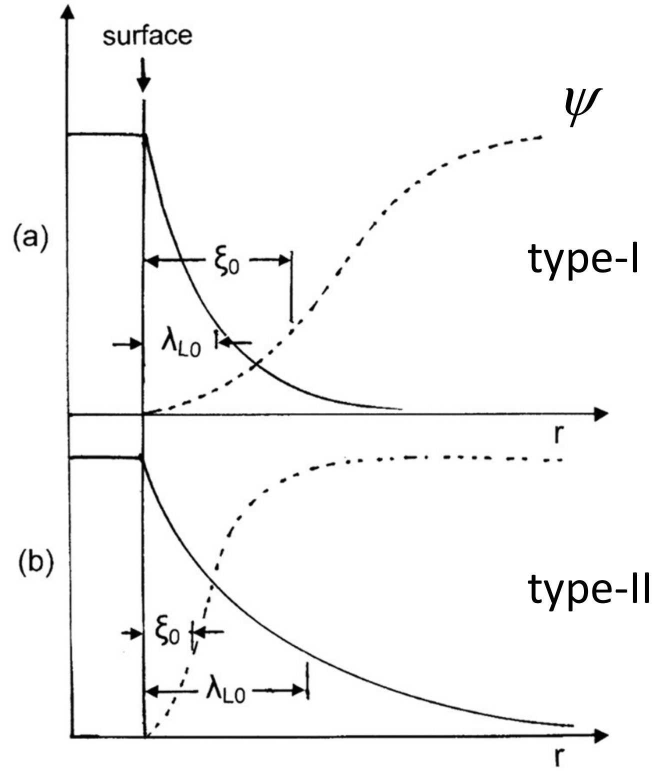
Coherence length:

$$\psi \propto e^{-r/\xi_0}$$

$$\xi_0 \propto \frac{\hbar v_F}{2\Delta}$$

Mean free path:

$$\ell = v_F \tau$$



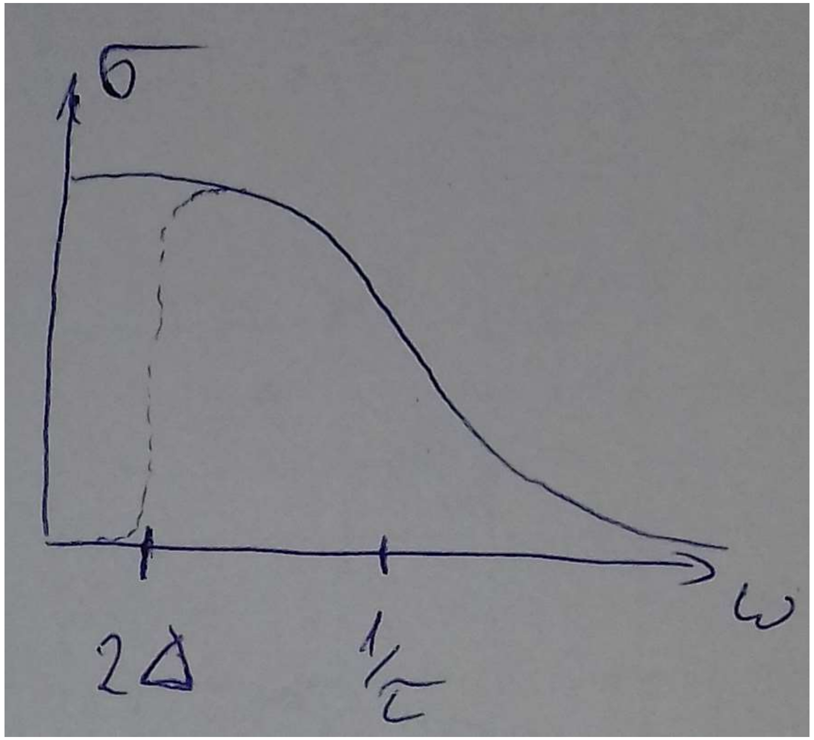
Superconductors in the dirty limit

Only a fraction of the Drude peak contribute to the condenstate:

$$\ell < \xi_0$$

$$v_F \tau < \frac{\hbar v_F}{2\Delta}$$

$$2\Delta < \frac{\hbar}{\tau}$$



$$\int_0^\infty \sigma(\omega) d\omega = \frac{nq^2}{m}\frac{\pi}{2}$$

$$\sigma_{DC} 2\Delta \sim \frac{n_S q^2}{m} = \frac{1}{\mu_0 \lambda_L^2}$$

$$\lambda_L = \sqrt{\frac{m}{\mu_0 n q^2 2\Delta\tau}}$$

>>

Clean limit:

$$\lambda_L = \sqrt{\frac{m}{\mu_0 n q^2}}$$

Extended Drude model

Interactions of electrons:

- other electrons (Coulomb)
- phonons

Frequency dependent scattering rate (Kramers-Kronig should be satisfied):

$$\sigma = i\frac{nq^2}{m}\frac{1}{\omega + i\gamma} \rightarrow i\frac{nq^2}{m}\frac{1}{\omega + i\Gamma(\omega)}$$

Frequency dependent scattering rate

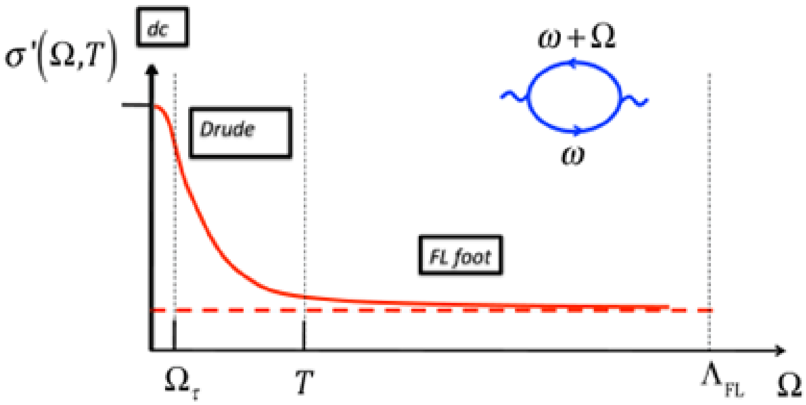
$$\frac{1}{\tau^*} = \Re e\{\Gamma(\omega)\}$$

Frequency dependent mass enhancement

$$\frac{m^*}{m} = 1 - \frac{\Im m\{\Gamma(\omega)\}}{\omega}$$

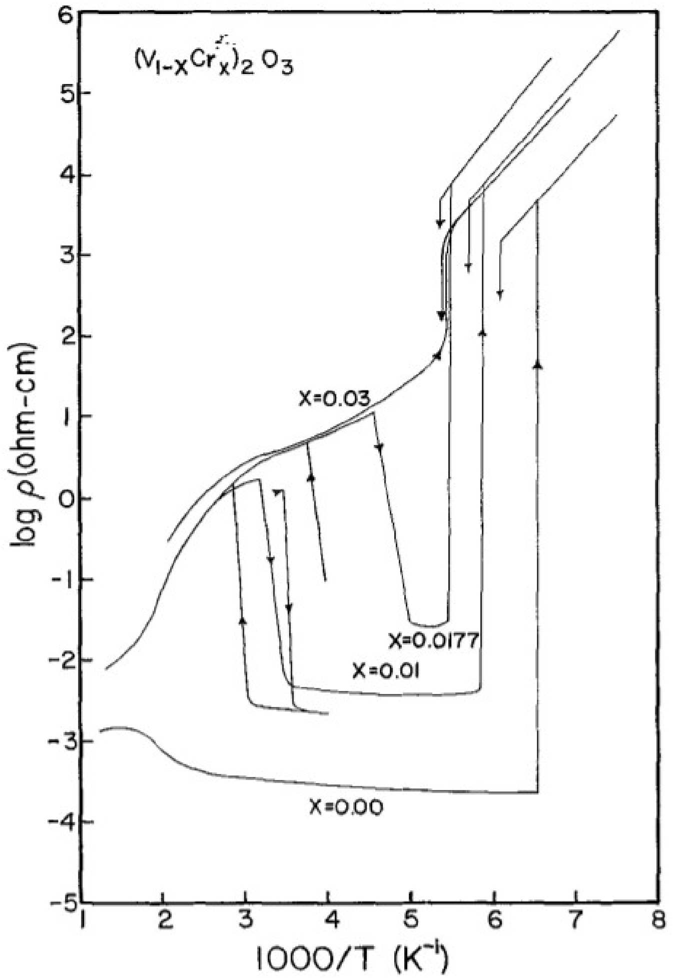
Weakly interacting electron gas (Fermi liquid):

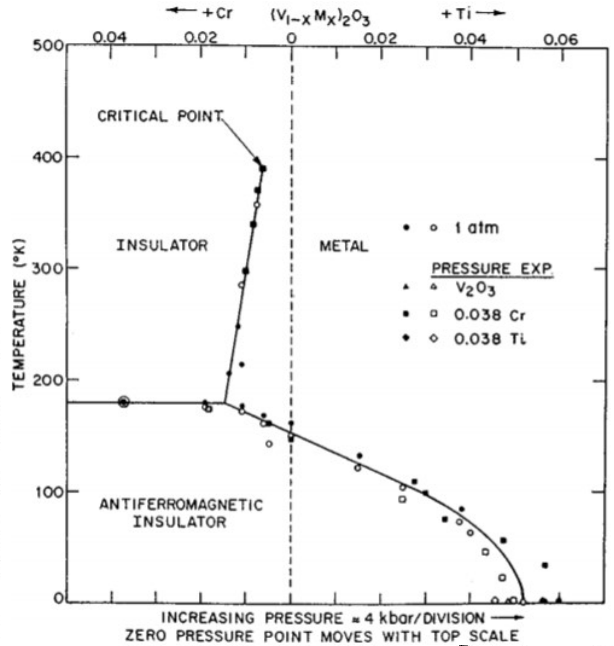
$$\frac{\hbar}{\tau^*} = \frac{2}{3E_F}(\hbar\omega)^2 + (2\pi k_B T)^2$$

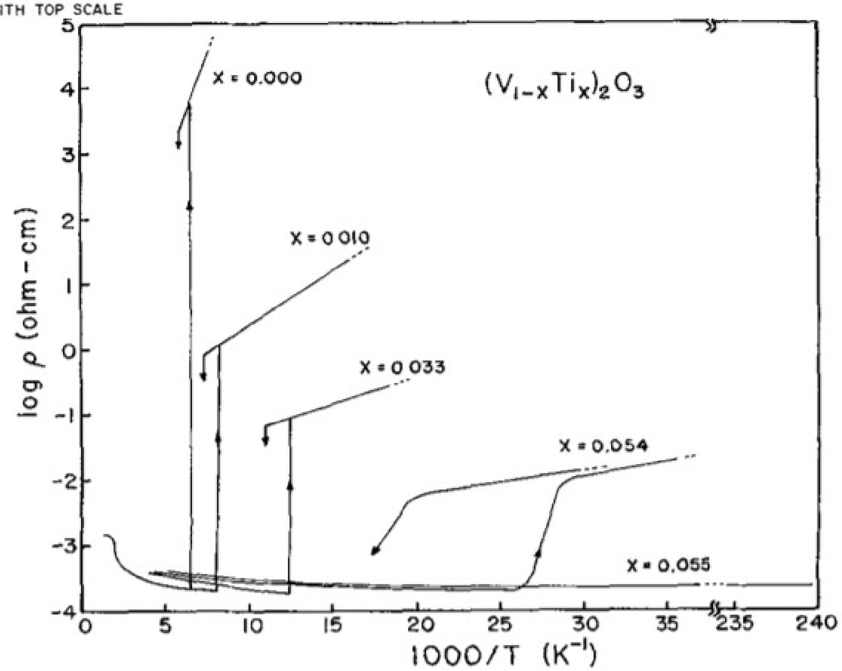


Maslov, D. L., & Chubukov, A. V. *Rep. Prog. Phys., 80(2), 026503* (2016).

Electron-electron interaction: Mott insulators

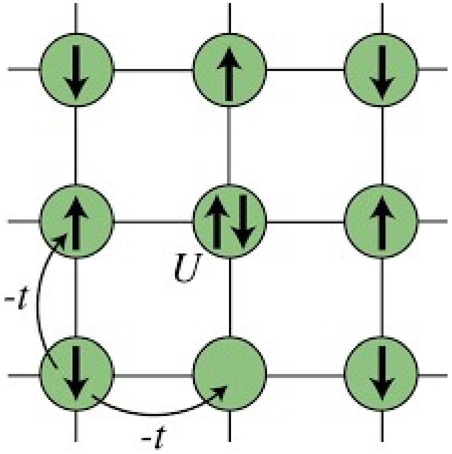


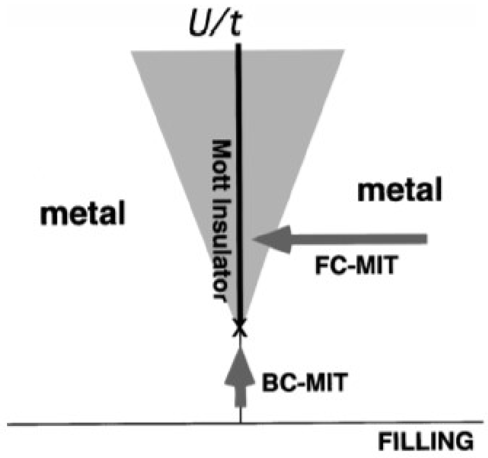




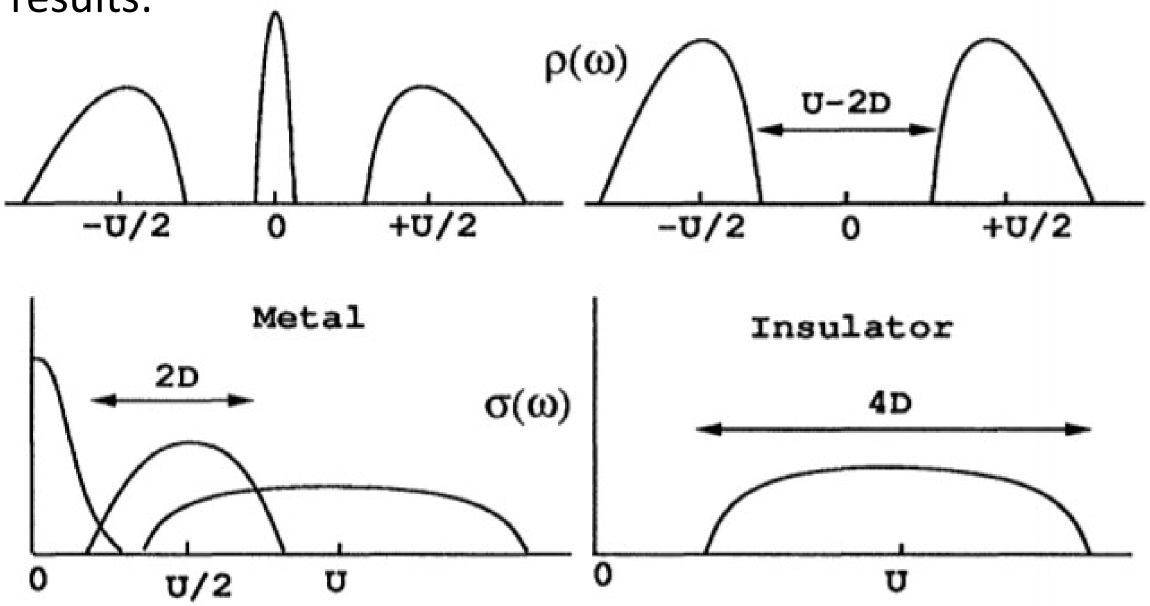
Electron-electron interaction: Mott insulators

Hubbard model: $$\mathcal{H} = \sum_{i,j} t_{i,j}(c^{+}_{i,\sigma}c_{j,\sigma} + c^{+}_{j,\sigma}c_{i,\sigma}) + U\sum_{i} n_{i,\sigma}n_{i,-\sigma}$$

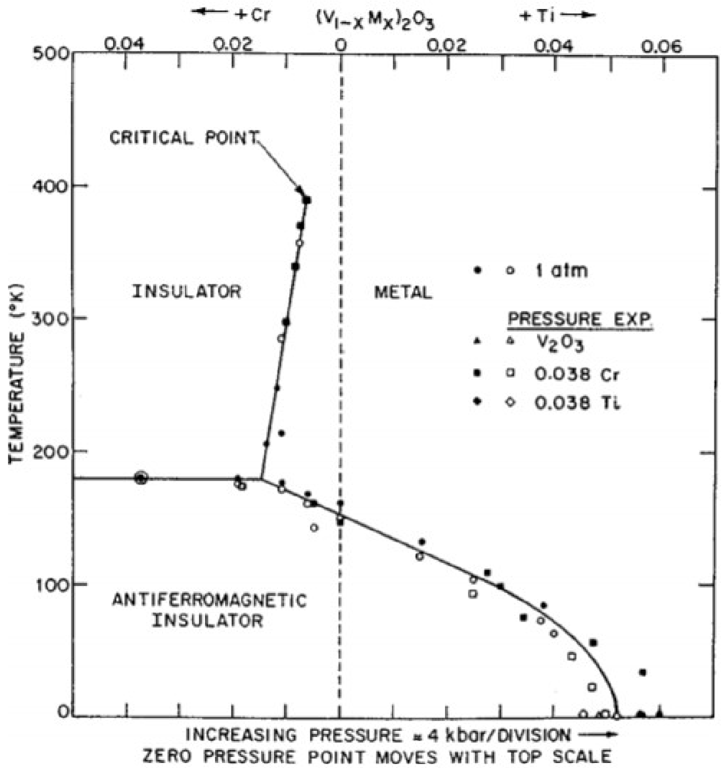


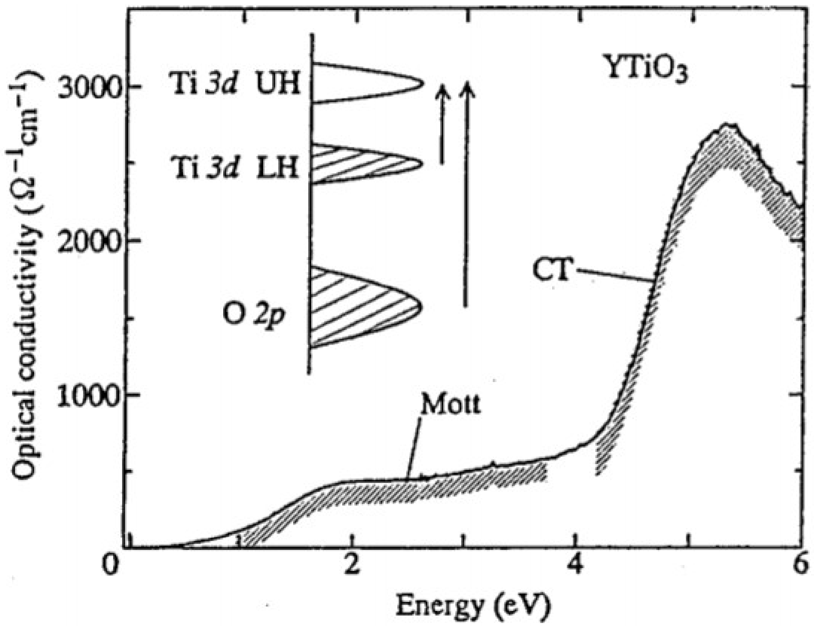


DMFT results:



Phys. Rev. Lett. **75**, 105 (1995).





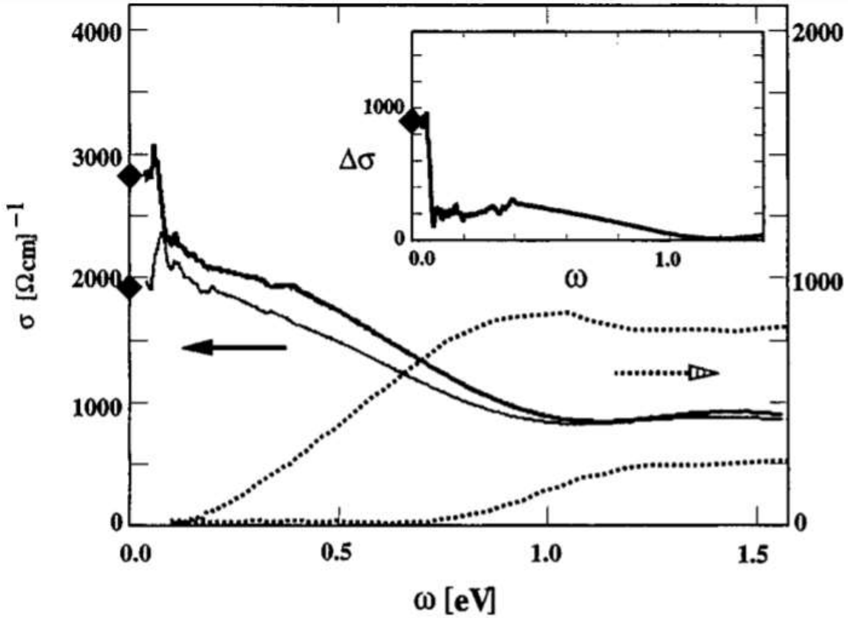
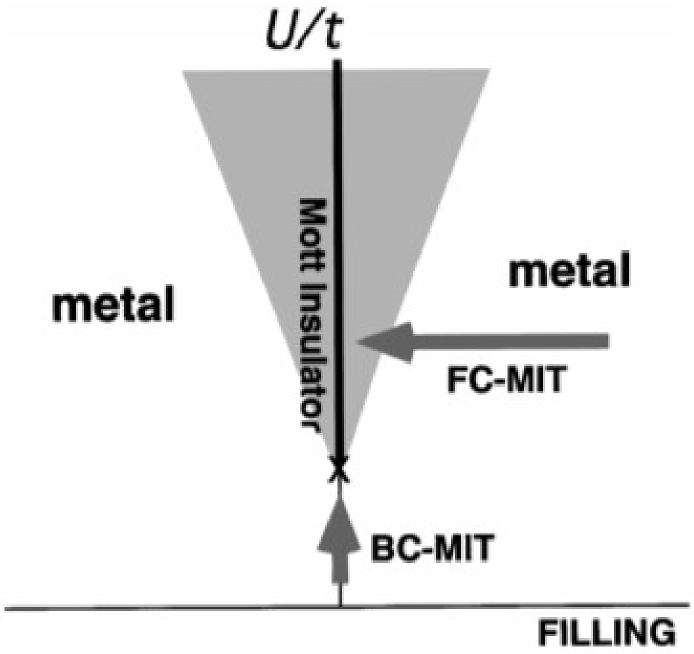
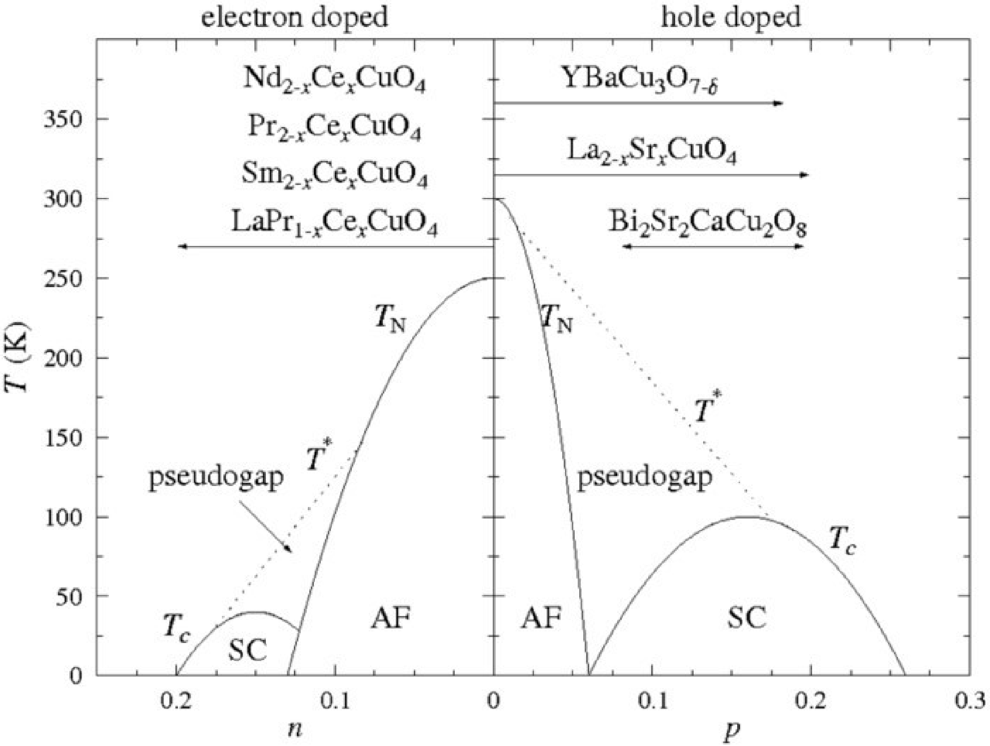
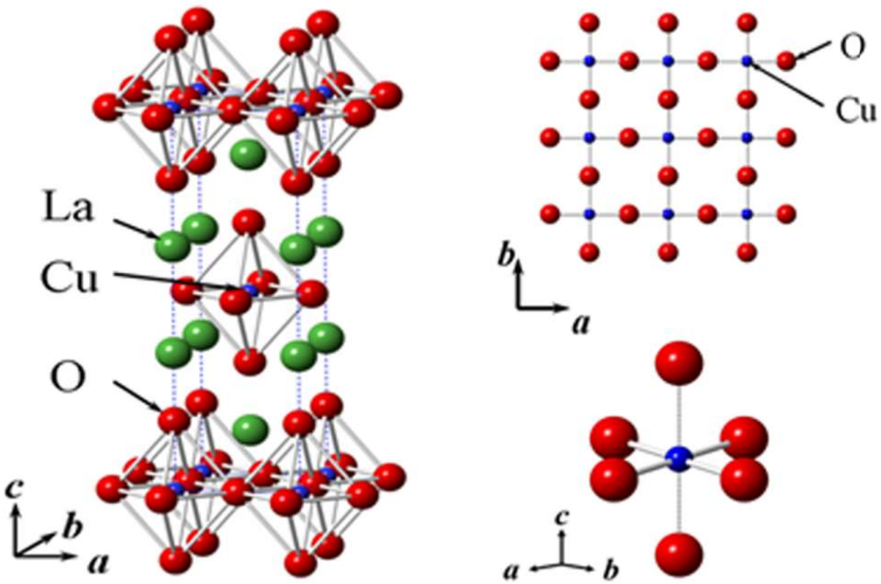
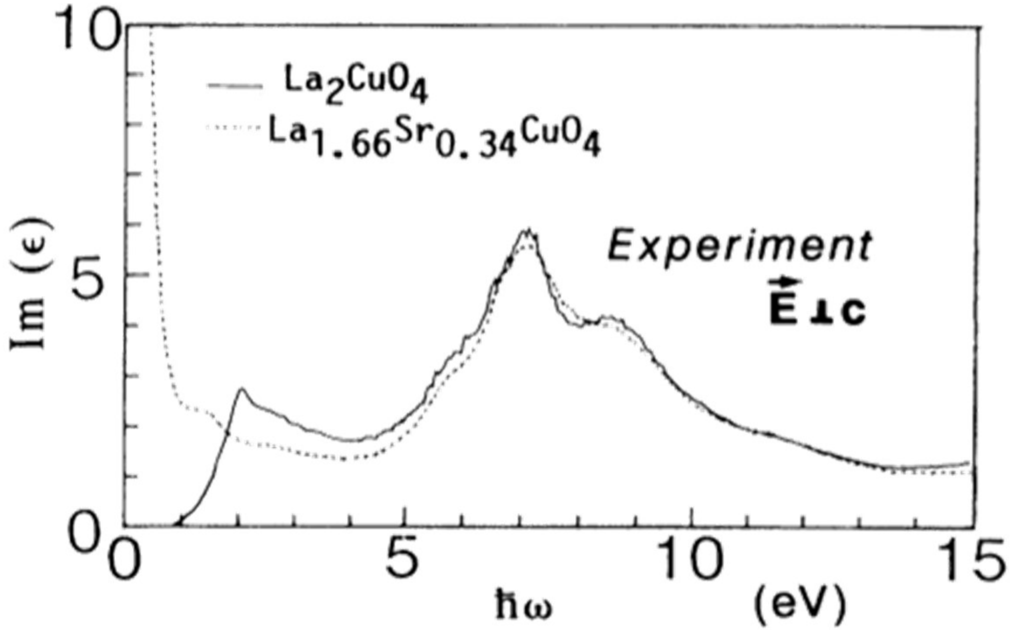


FIG. 75. Optical conductivity spectra of $V_{2-y}O_3$ in the metallic phase (full lines) at $T=170$ K (upper) and $T=300$ K (lower). The inset contains the difference of the two spectra $\Delta\sigma(\omega) = \sigma_{170\,K}(\omega) - \sigma_{300\,K}(\omega)$. Diamonds indicate the measured dc conductivity. Dotted lines indicate $\sigma(\omega)$ of insulating phase with $y=0.013$ at 10 K (upper) and $y=0$ at 70 K (lower). From Rozemberg *et al.*, 1995.

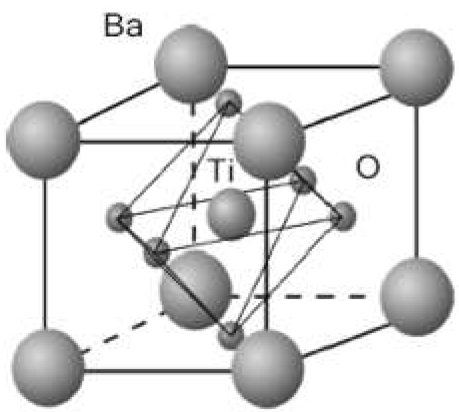

U/t
Mott Insulator
metal
metal
FC-MIT
BC-MIT
FILLING

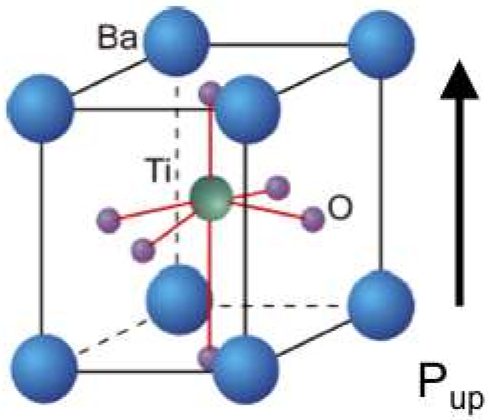

electron doped
hole doped
$Nd_{2-x}Ce_xCuO_4$
$Pr_{2-x}Ce_xCuO_4$
$Sm_{2-x}Ce_xCuO_4$
$LaPr_{1-x}Ce_xCuO_4$
$YBaCu_3O_{7-\delta}$
$La_{2-x}Sr_xCuO_4$
$Bi_2Sr_2CaCu_2O_8$
T (K)
T_N
T_N
T^*
T^*
pseudogap
pseudogap
T_c
T_c
SC
SC
AF
AF
n
p

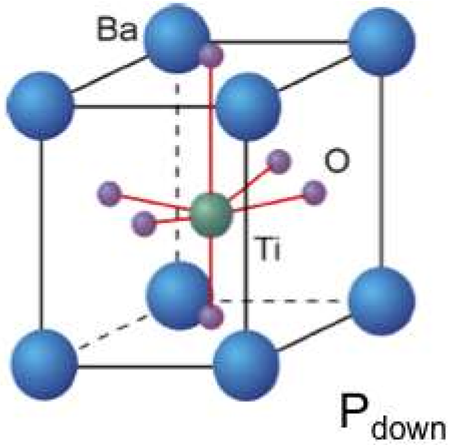

La
Cu
O
O
Cu
c
b
a
b
a
c
a
b

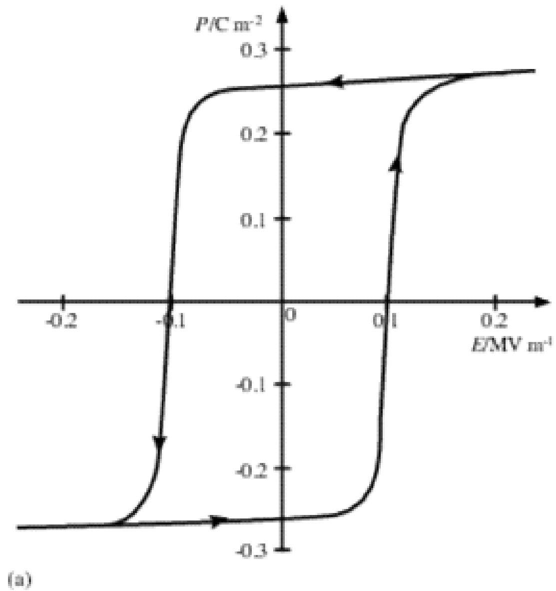

La_2CuO_4
$La_{1.66}Sr_{0.34}CuO_4$
Im (ε)
Experiment
$\vec{E} \perp c$
ħω
(eV)

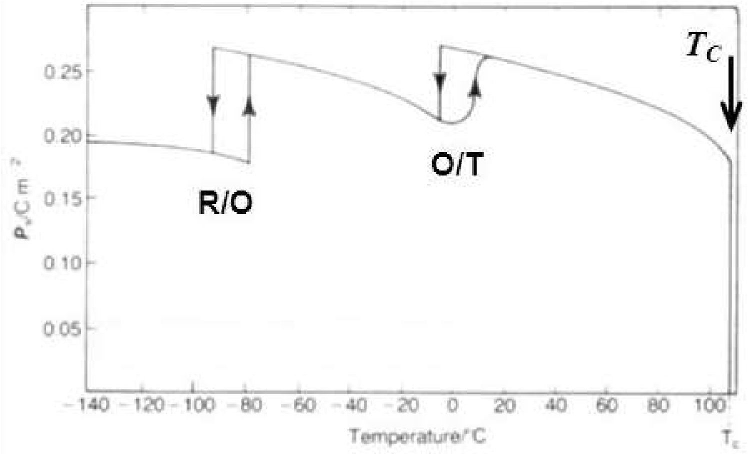
Ferroelectrics

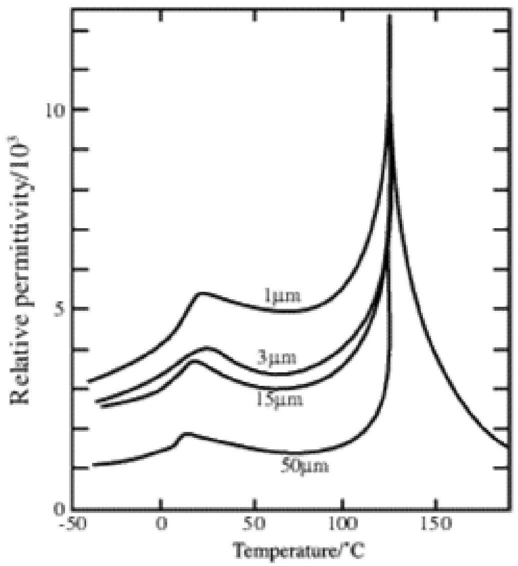








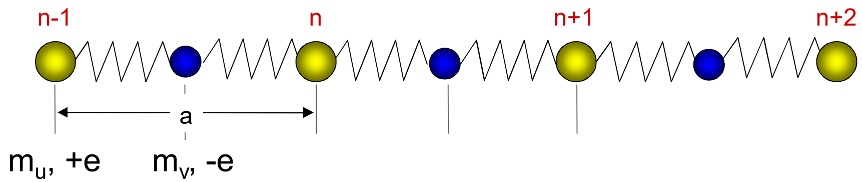




Vibrational spectroscopy

$$m_u \frac{d^2 u_n}{dt^2} = D(v_n + v_{n-1} - 2u_n) - \gamma m_u \frac{du_n}{dt} + eE(t)$$

$$m_v \frac{d^2 v_n}{dt^2} = D(u_n + u_{n-1} - 2v_n) - \gamma m_v \frac{dv_n}{dt} - eE(t)$$



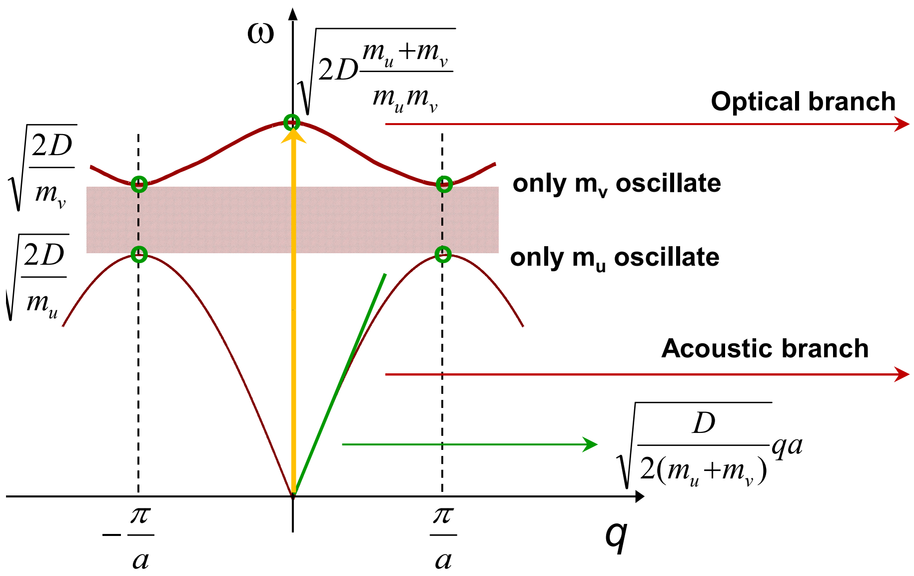
$$\lambda >> a \Rightarrow q << \frac{\pi}{a} \Rightarrow \begin{cases} E(r,t) \approx E_\omega e^{i\omega t} \\ u_n(t) \approx u e^{-i\omega t} \\ v_n(t) \approx v e^{-i\omega t} \end{cases}$$

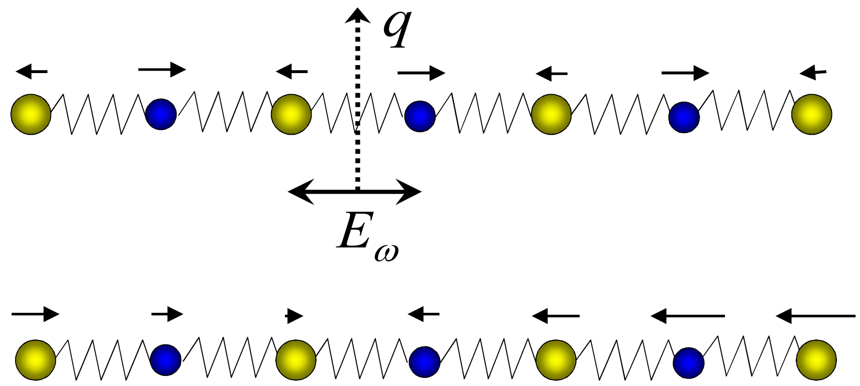
$$\omega_{TO} = \sqrt{2D \frac{m_u + m_v}{m_u m_v}}$$

$$P_\omega = en(u_\omega - v_\omega) = \frac{ne^2}{\mu} \frac{1}{\omega_{TO}^2 - \omega^2 - i\gamma\omega} E_\omega$$

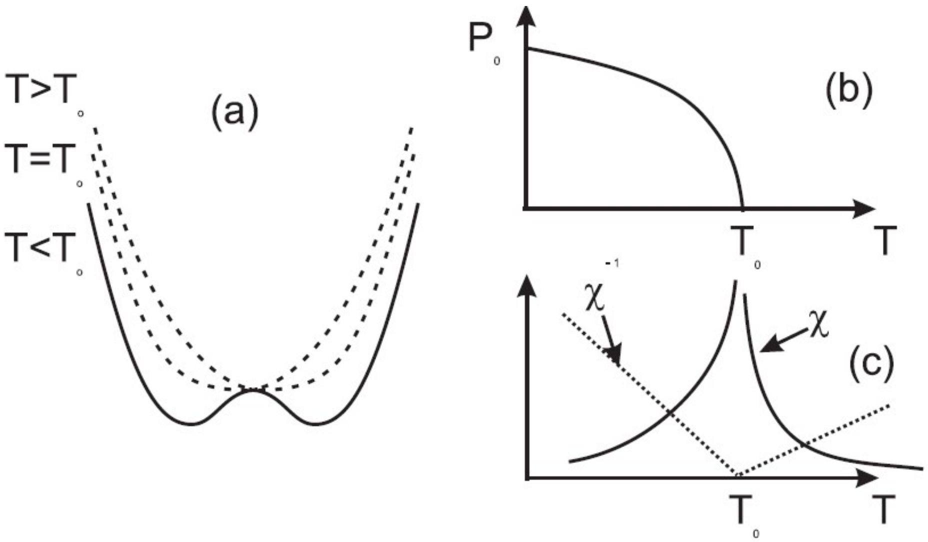
$$\varepsilon(\omega) = 1 + \frac{\Omega_{pl}^2}{\omega_{TO}^2 - \omega^2 - i\gamma\omega}$$

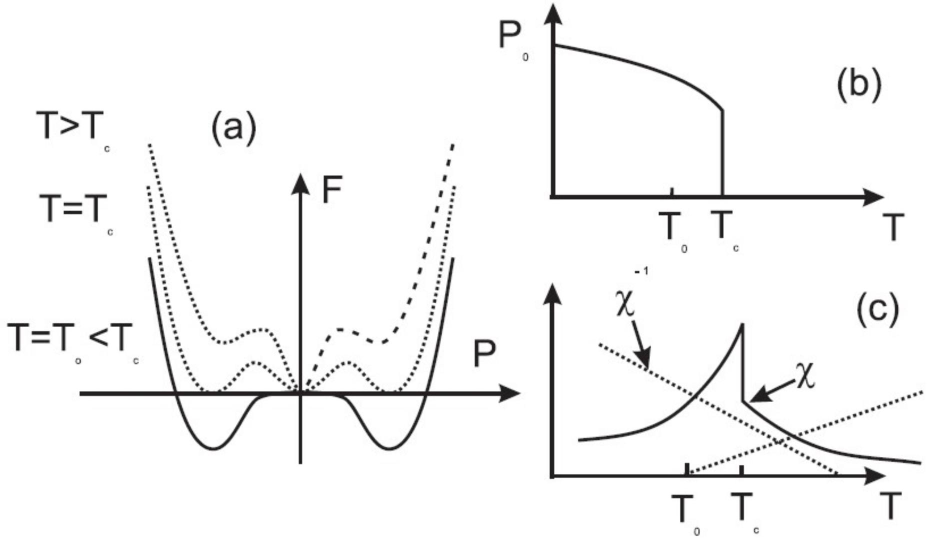
The q=0 case is equivalent to a diatomic molecule, atoms move respect to the center of mass





$$\mathcal{F}_P = \frac{1}{2}aP^2 + \frac{1}{4}bP^4 + \frac{1}{6}cP^6 + ... - EP$$





$$m\frac{\partial^2 u_{opt}}{\partial t^2} = -\frac{\partial \mathcal{F}}{\partial u_{opt}} \propto -a(T)u_{opt}$$

$$\omega(q=0)^2 \propto \frac{1}{\chi}$$

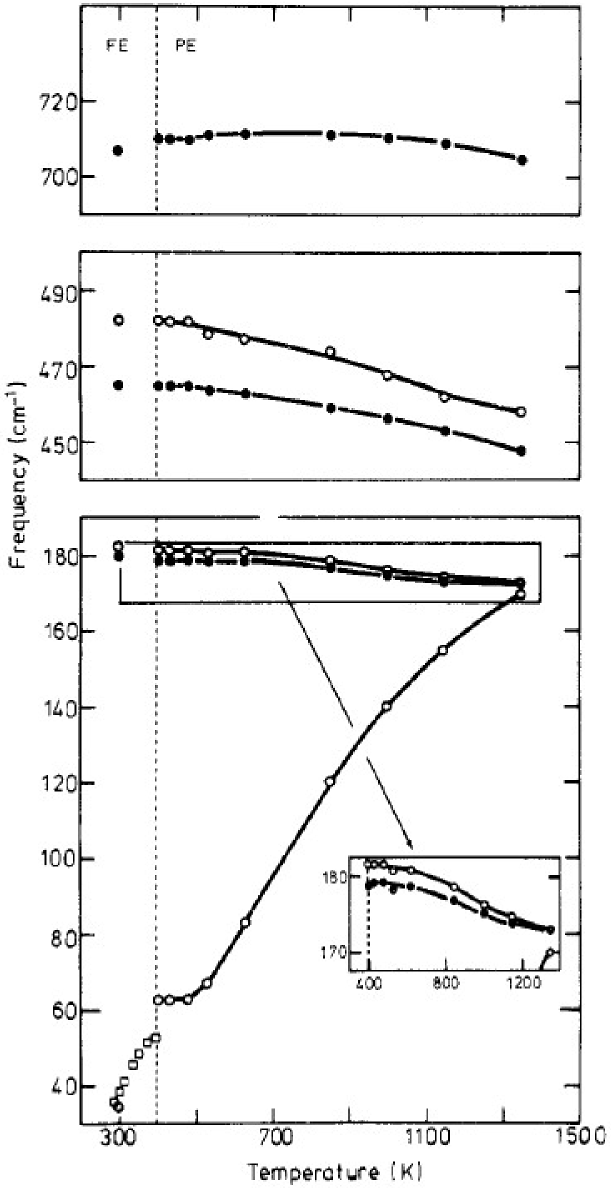


Figure 4. Frequencies of the E modes at room temperature and temperature dependence of the frequencies of F_{1u} modes in the cubic phase for $BaTiO_3$. ○, transverse modes; ●, longitudinal modes. Raman data (□) are taken from Scalabrin *et al* (1977).

$$\chi'(0) = \frac{2}{\pi} \wp \int_0^\infty \frac{\chi''(\omega')}{\omega'} d\omega'$$

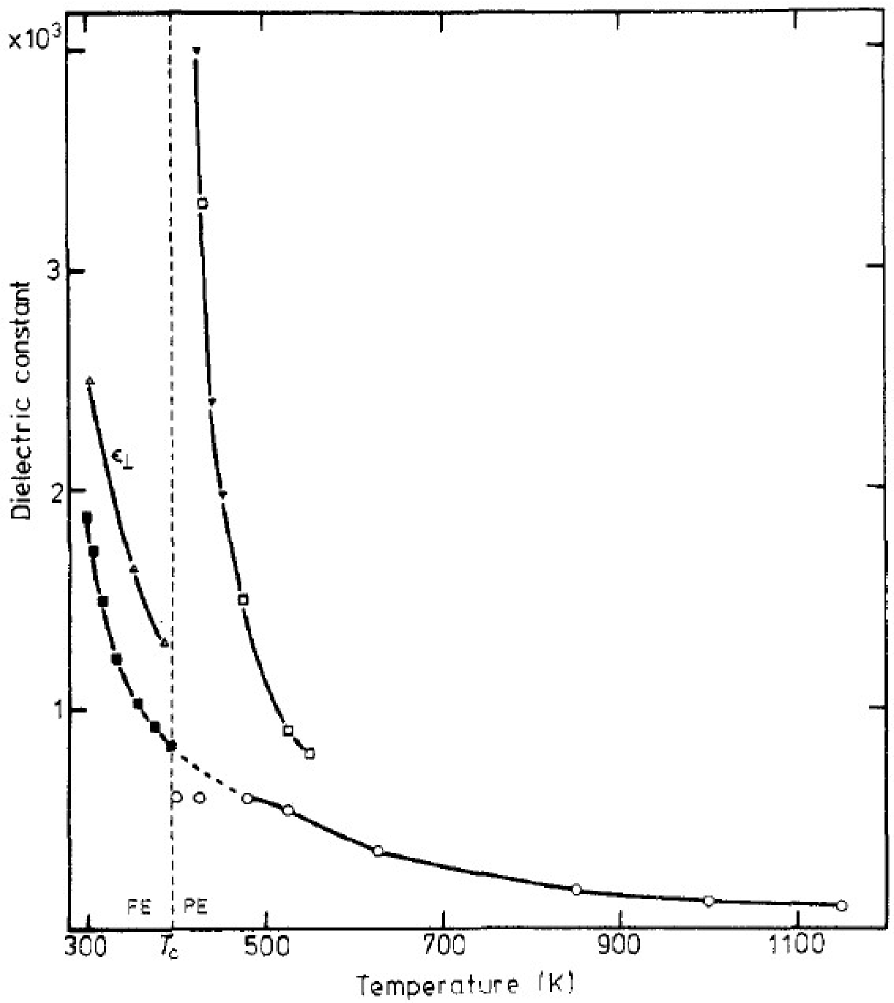


Figure 10. Temperature dependence of the dielectric constant of $BaTiO_3$: ○ present IR study; ■ Raman data of Scalabrin *et al* (1977); direct dielectric measurements ▼ (24 GHz), □ (37 GHz), △ (250 MHz) are those of Benedict and Durand (1958), Poplavko (1966) and Wemple *et al* (1968) respectively.