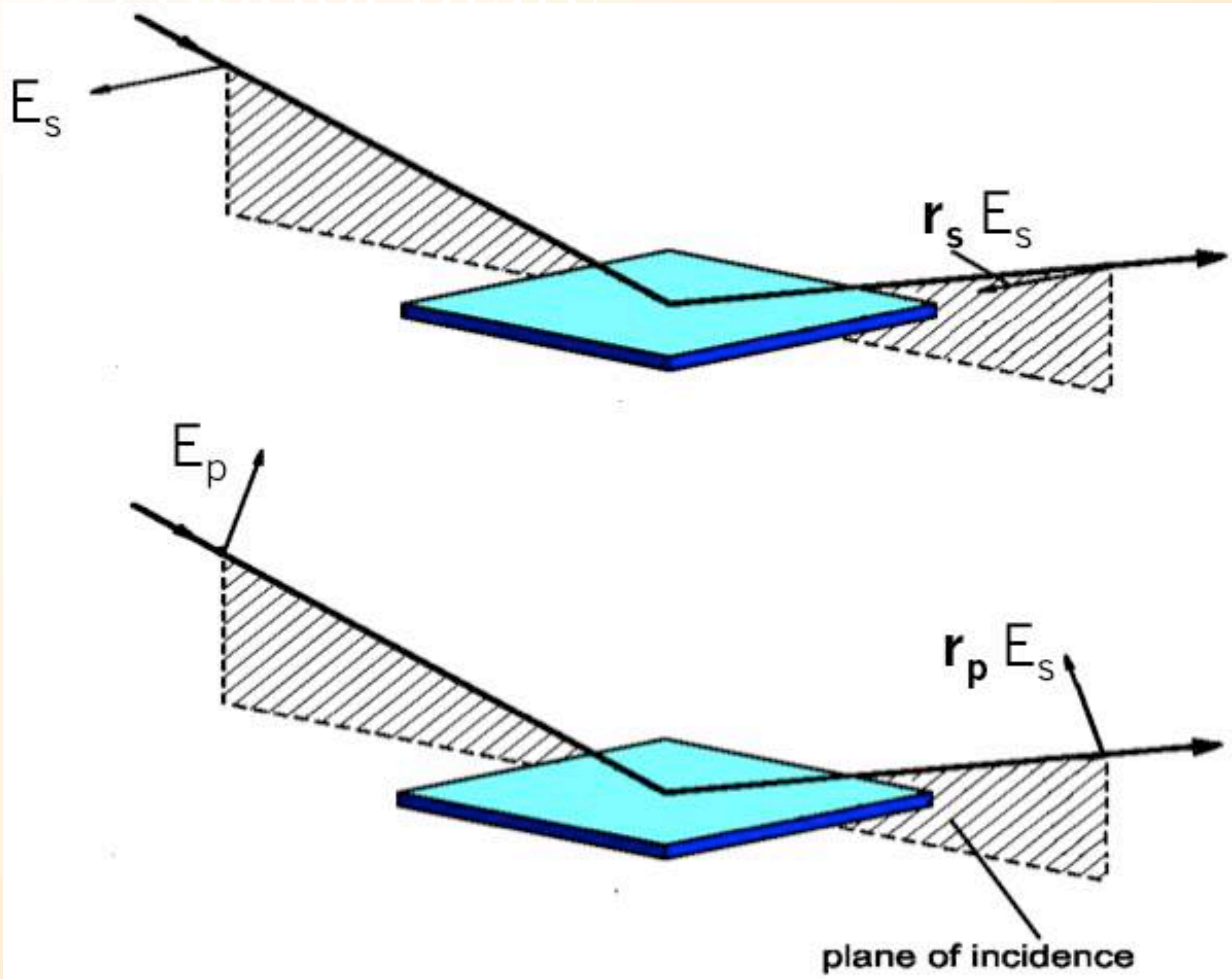
Optical properties of correlated electrons

Introduction
The cuisine of optical spectroscopy

Part I
Insulators – Interactions – Excitons

Part II
The Internal Energy of a Superconductor

The cuisine of optical spectroscopy



$$r_s = \frac{\cos\theta - \sqrt{\epsilon - \sin^2\theta}}{\cos\theta + \sqrt{\epsilon - \sin^2\theta}} \qquad r_p = \frac{\epsilon\cos\theta - \sqrt{\epsilon - \sin^2\theta}}{\epsilon\cos\theta + \sqrt{\epsilon - \sin^2\theta}}$$

Compression and extinction of EM-waves

Index of refraction: $\eta(\omega) = \mathrm{Re}\sqrt{\varepsilon(\omega)}$

Extinction coefficient: $\kappa(\omega) = \mathrm{Im}\sqrt{\varepsilon(\omega)}$

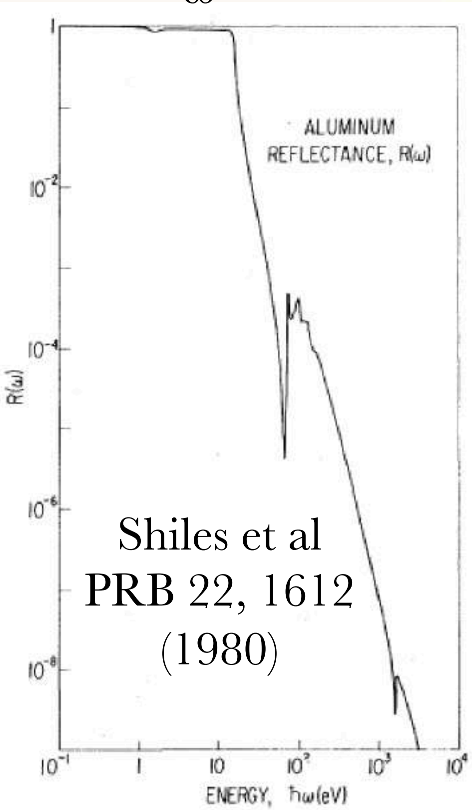
Dissipation of the EM-flux

Optical conductivity: $\mathrm{Re}\,\sigma(\omega) = \dfrac{\omega}{4\pi}\mathrm{Im}\,\varepsilon(\omega)$

The cuisine of optical spectroscopy

Case study: aluminum

1: Measure R(ω) for 0 < ω < ∞

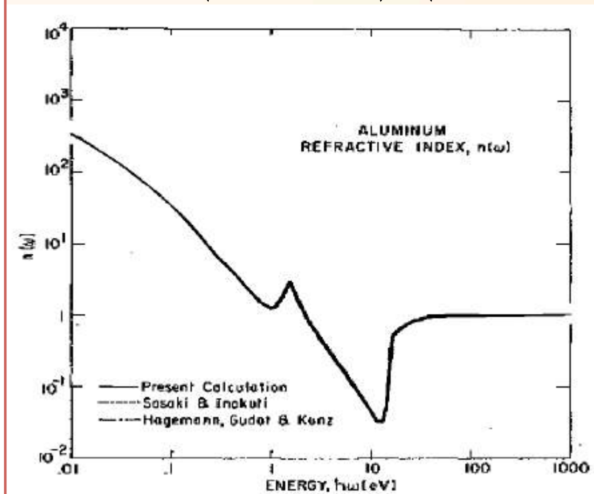


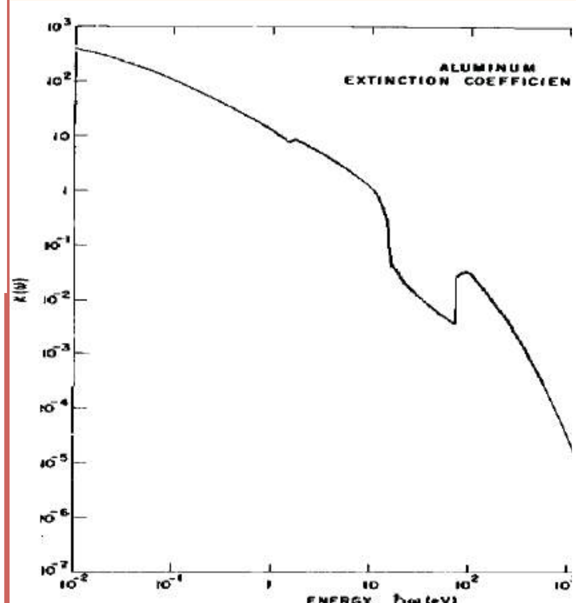
Shiles et al
PRB 22, 1612
(1980)

2: Calculate $\phi(\omega)$ from $\sqrt{R(\omega)}$ using Kramers-Kronig relations

3: Calculate

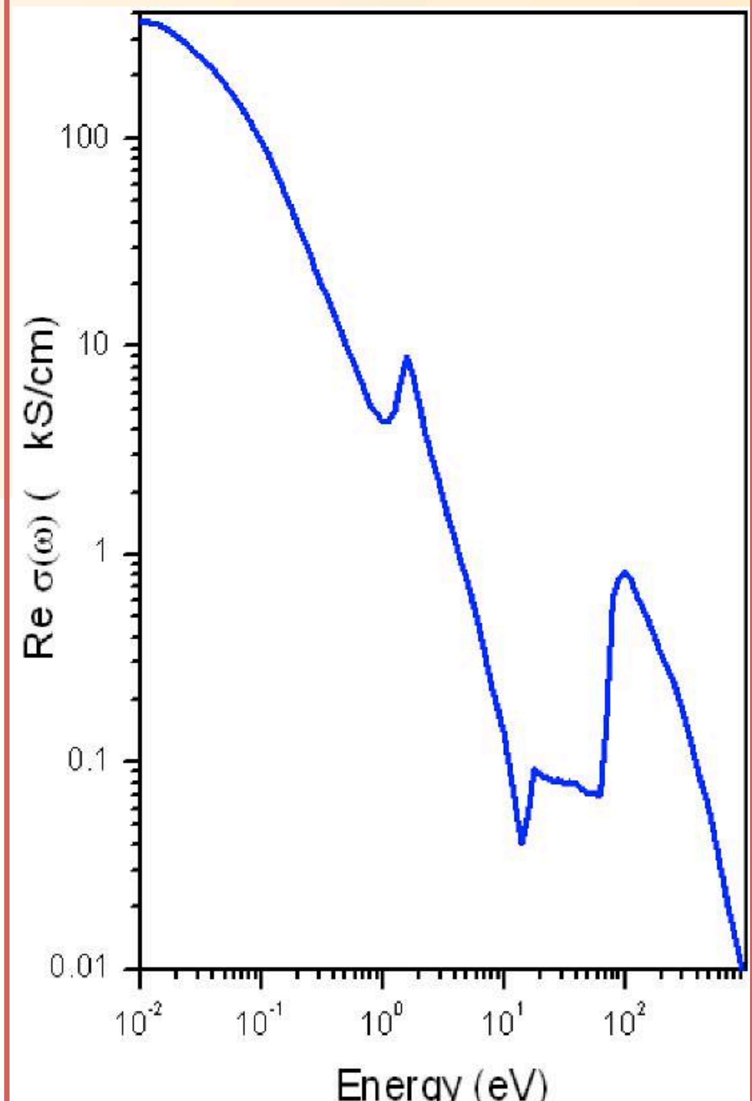
$$n + i\kappa = \left(1 - e^{i\varphi}\sqrt{R}\right) / \left(1 + e^{i\varphi}\sqrt{R}\right)$$





4: Calculate

$$\mathrm{Re}\,\sigma = \omega\, n\, \kappa / (2\pi)$$



Optical conductivity

EM field couples to every charged particle

Charge $Q_\mu \rightarrow$ current $j_\mu = v_\mu Q_\mu$

Coupling to EM field: $H = -A \bullet j_\mu$

Linear response: $\sigma(\omega) = \frac{e^2}{\omega V}\left\{\frac{iN}{m} + \int_0^\infty dt e^{i\omega t} \sum_\nu Z_\nu \left\langle \nu \left| \left[\hat{j}(t), \hat{j}(0)\right] \right| \nu \right\rangle \right\}$

$$\Rightarrow \mathrm{Re}\,\sigma(\omega) = \frac{\pi e^2}{V} \sum_{\mu\nu} \left(Z_\mu - Z_\nu\right) \frac{\left\langle \nu \middle| \hat{j} \middle| \mu \right\rangle \left\langle \mu \middle| \hat{j} \middle| \nu \right\rangle}{E_\mu - E_\nu} \delta\left(\hbar\omega + E_\nu - E_\mu\right)$$

F-sum rule for the optical conductivity

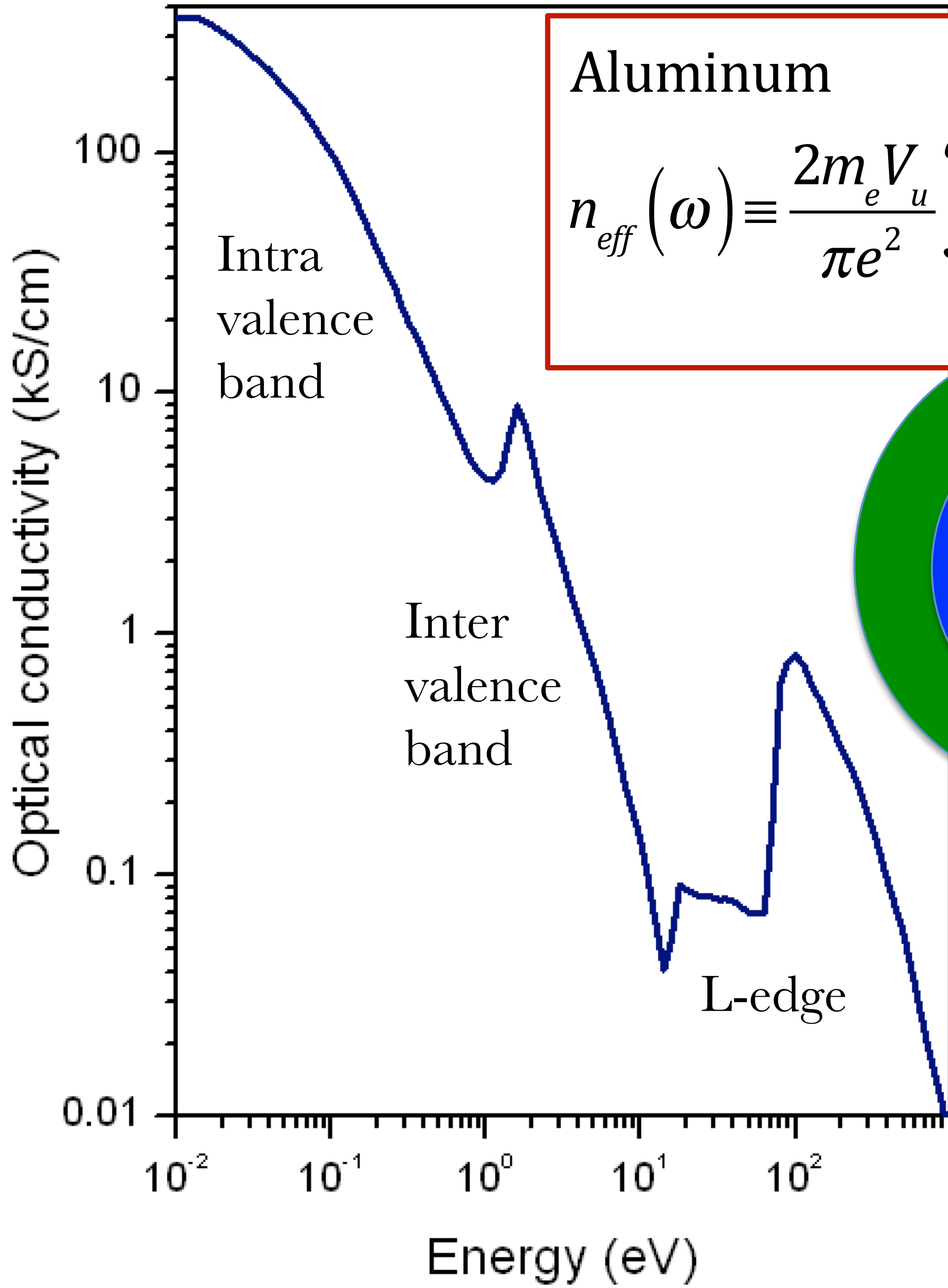
$$\mathrm{Re}\,\sigma(\omega)=\frac{\pi e^2}{V}\sum_{\mu\nu}\left(Z_\mu - Z_\nu\right)\frac{\langle\nu|\hat{j}|\mu\rangle\langle\mu|\hat{j}|\nu\rangle}{E_\mu - E_\nu}\delta\left(\hbar\omega + E_\nu - E_\mu\right)$$

$$\Rightarrow \int_{-\infty}^{\infty}\mathrm{Re}\,\sigma(\omega)\,d\omega=\frac{2\pi e^2}{V}\sum_{\mu\nu}Z_\mu\frac{\langle\nu|\hat{j}|\mu\rangle\langle\mu|\hat{j}|\nu\rangle}{E_\mu - E_\nu}$$

$$\left.\begin{array}{l}\hat{H}|\nu\rangle = E_\nu|\nu\rangle \\ \hat{j}=i\hbar^{-1}\left[\hat{H},\hat{x}\right]\end{array}\right\} \Rightarrow \int_{-\infty}^{\infty}\mathrm{Re}\,\sigma(\omega)\,d\omega=\frac{\pi e^2}{i\hbar V}\left\langle\left[\hat{j},\hat{x}\right]\right\rangle$$

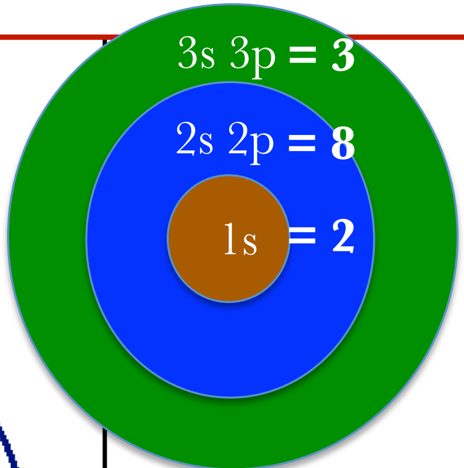
$$\left[\hat{j},\hat{x}\right]=i\frac{\hbar N}{m}\Rightarrow \int_{-\infty}^{\infty}\mathrm{Re}\,\sigma(\omega)\,d\omega=\frac{\pi e^2 n}{2m}$$

The cuisine of optical spectroscopy



Aluminum

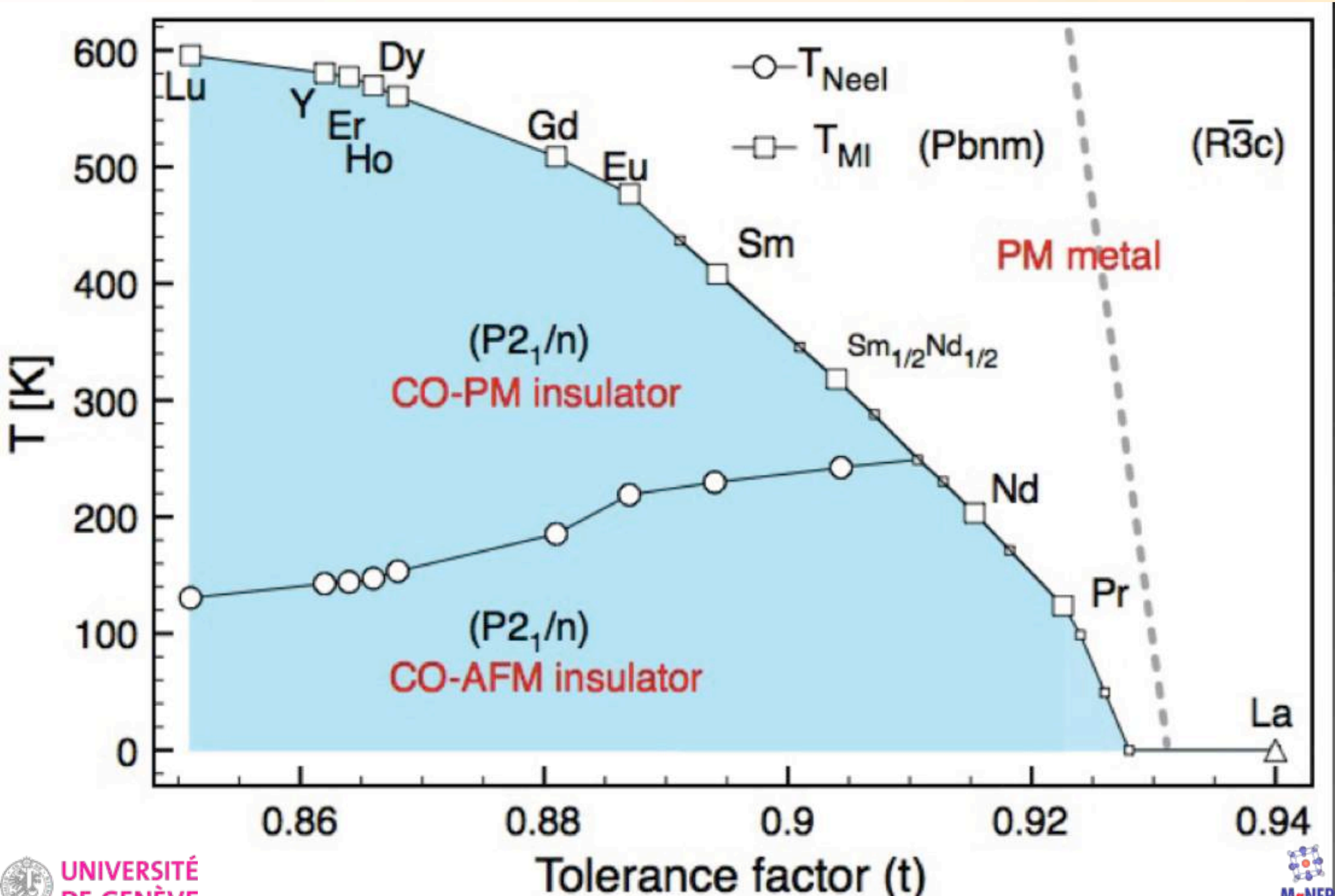
$$n_{eff}(\omega) \equiv \frac{2m_e V_u}{\pi e^2} \int_0^{\omega} \mathrm{Re}\,\sigma(\omega')\,d\omega'$$



2+8+3=13

After Shiles et al, PRB 22, 1612 (1980)

The cuisine of optical spectroscopy: Metal-Insulator transition in $RNiO_3$



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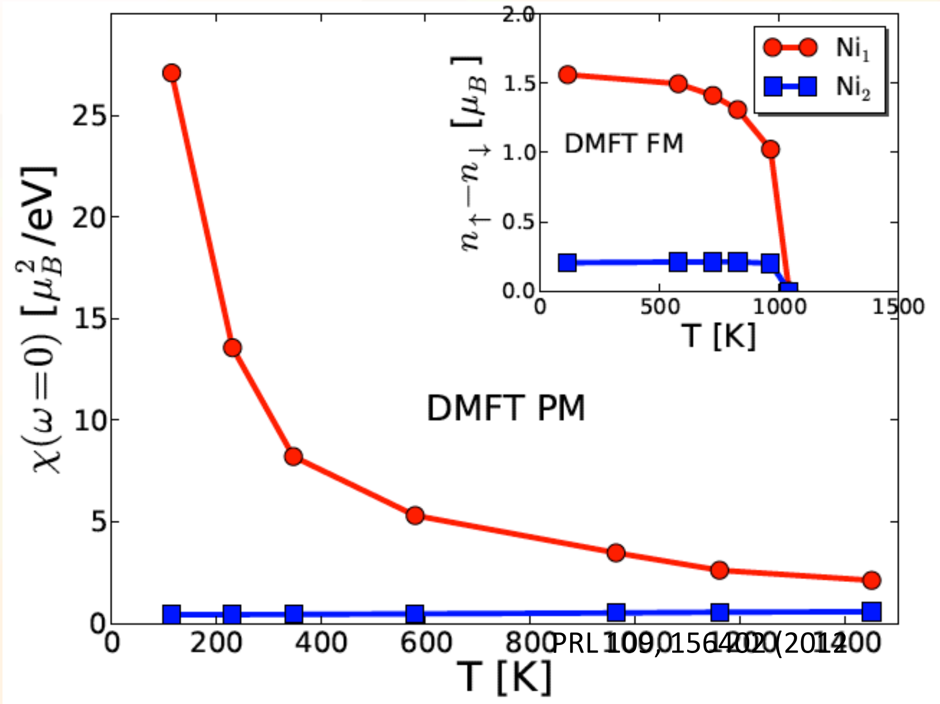
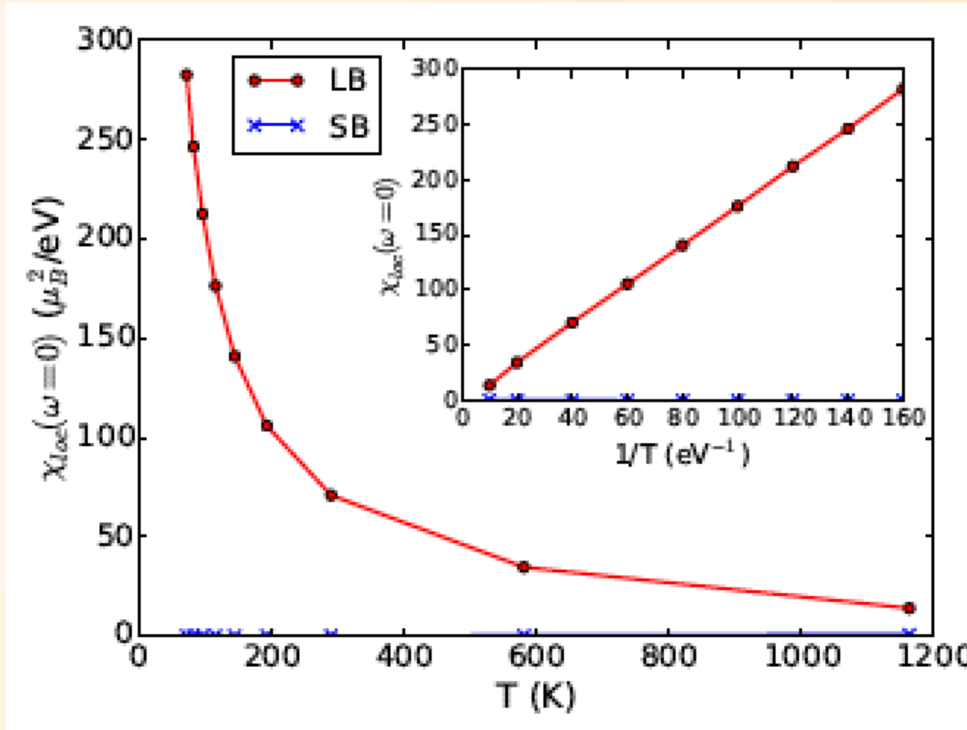
The cuisine of optical spectroscopy: Metal-Insulator transition in $RNiO_3$





Short Ni-O bond: $\left|d^8\underline{L}^2\right\rangle$ (4 holes, low spin, S=0)

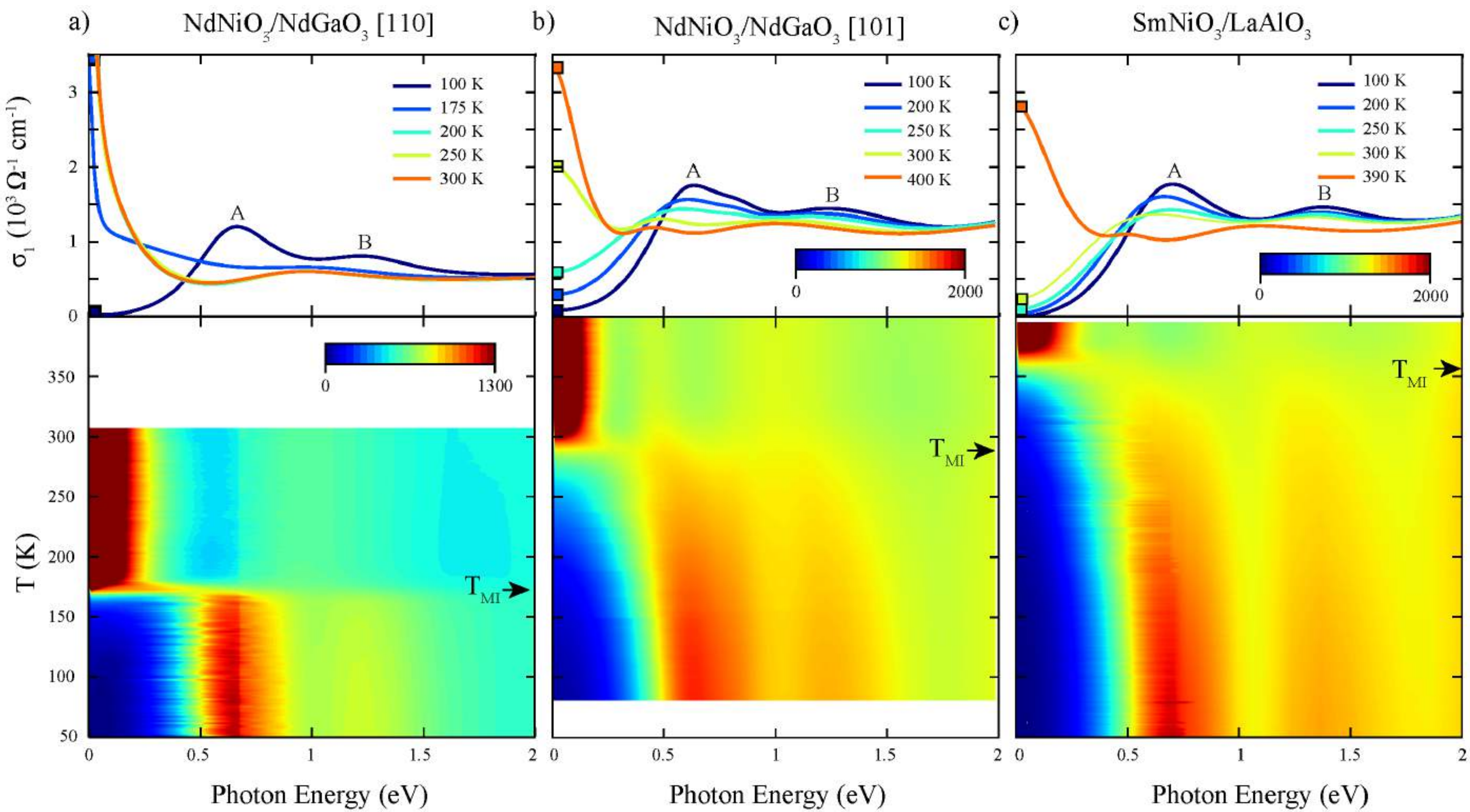
Long Ni-O bond: $\left|d^8\underline{L}^0\right\rangle$ (2 holes, high spin, S=1)

T. Mizokawa, D. I. Khomskii, and G. A. Sawatzky, PRB 61, 11263 (2000)

H. Park, A. J. Millis, and C. A. Marianetti, PRL 109, 156402 (2012)

A. Subedi, O. E. Peil, and A. Georges, PRB 91, 075128 (2015)

The cuisine of optical spectroscopy: Metal-Insulator transition in $RNiO_3$

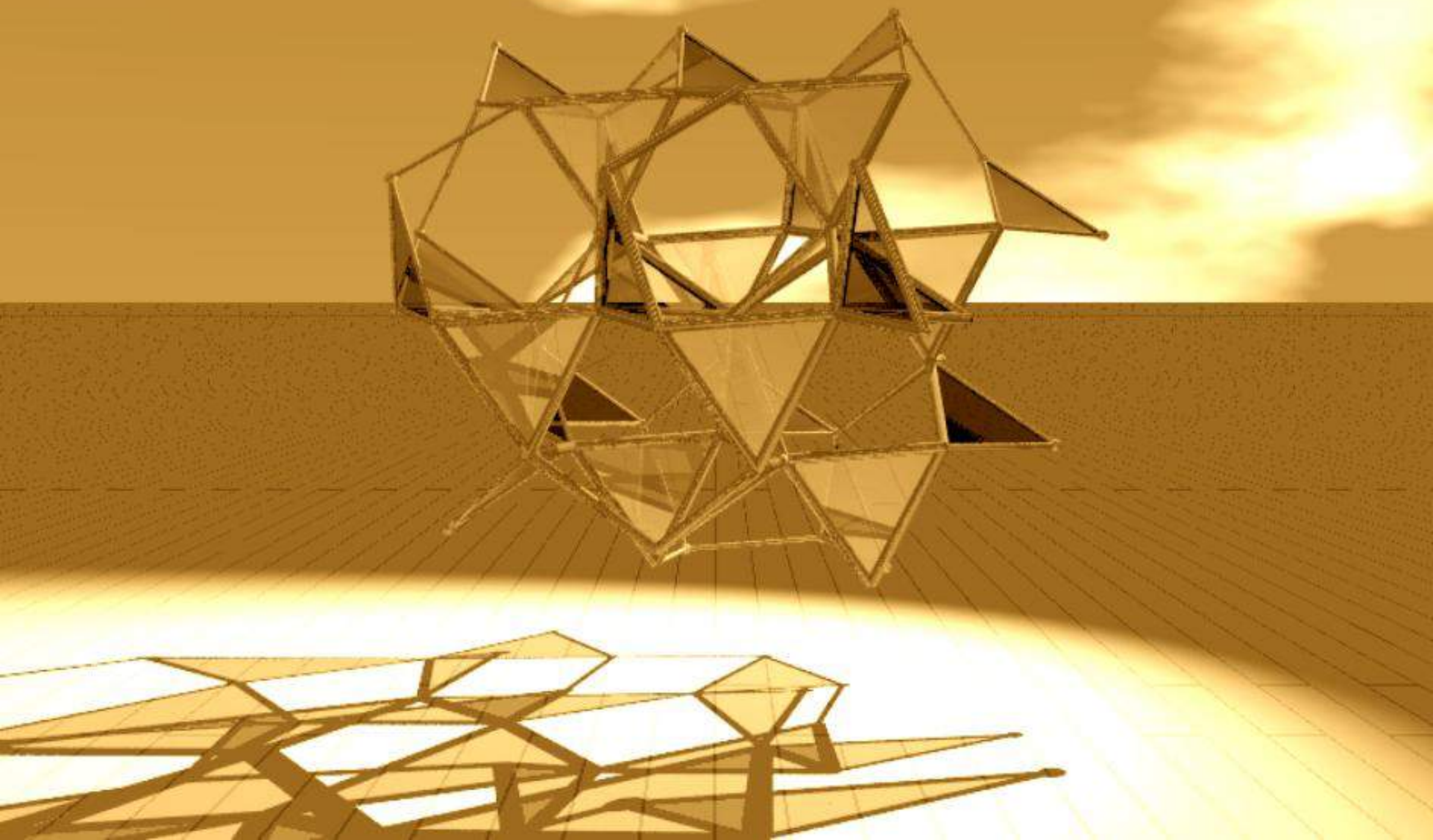


J. Ruppen, J. Teyssier, O.E. Peil, S. Catalano, M. Gibert, J. Mravlje, J.-M. Triscone, A. Georges, and D. van der Marel, Physical Review B 92, 155145 (2015).

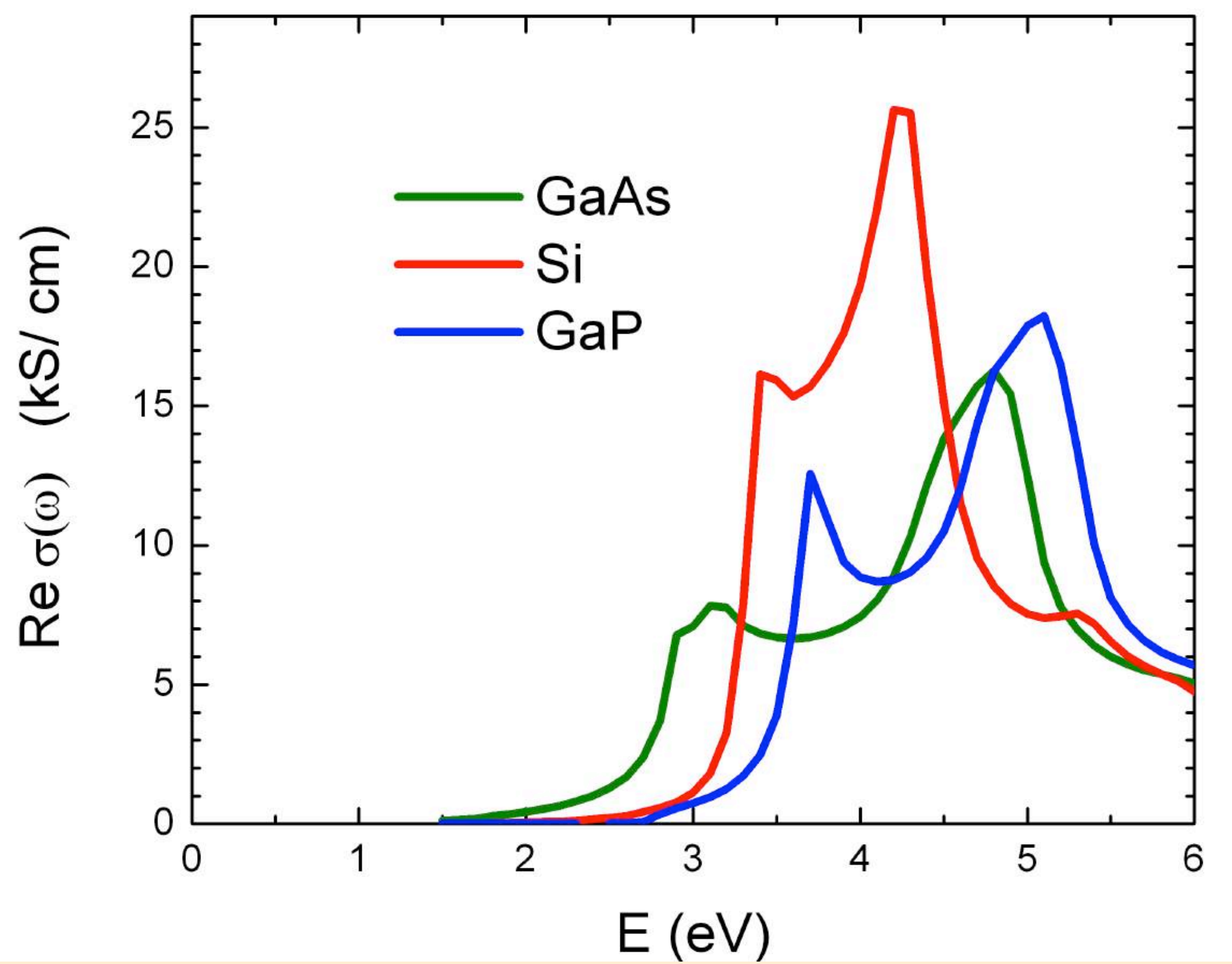
The cuisine of optical spectroscopy

Optical conductivity

Part I
Insulators – Interactions – Excitons

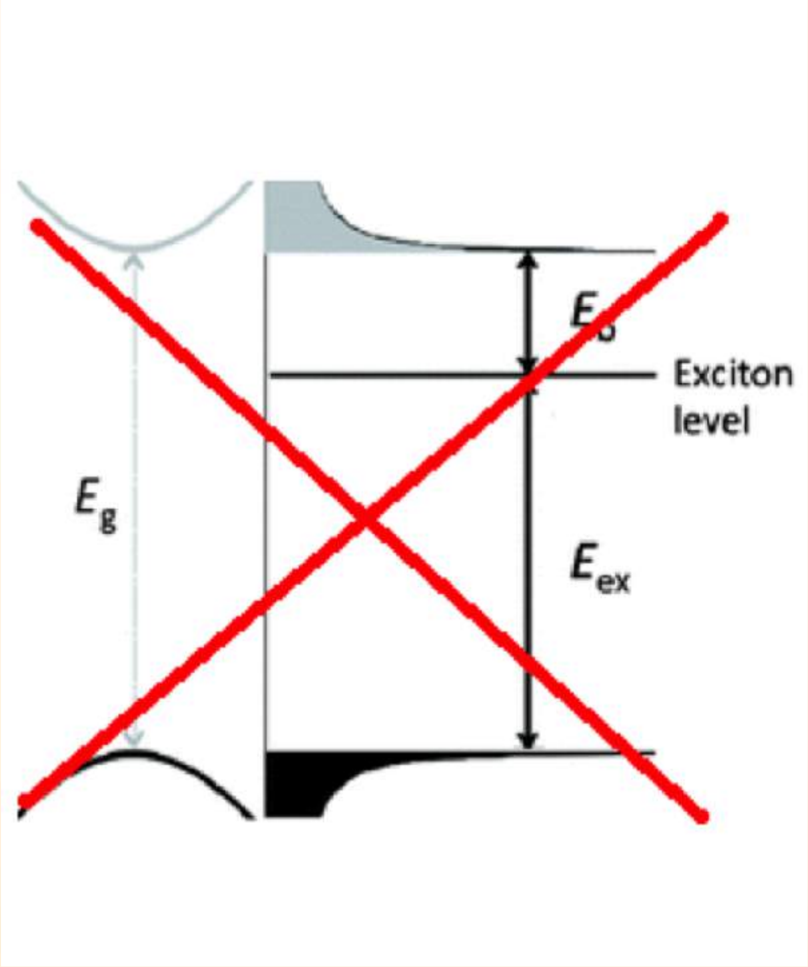


Optical conductivity of common semiconductors

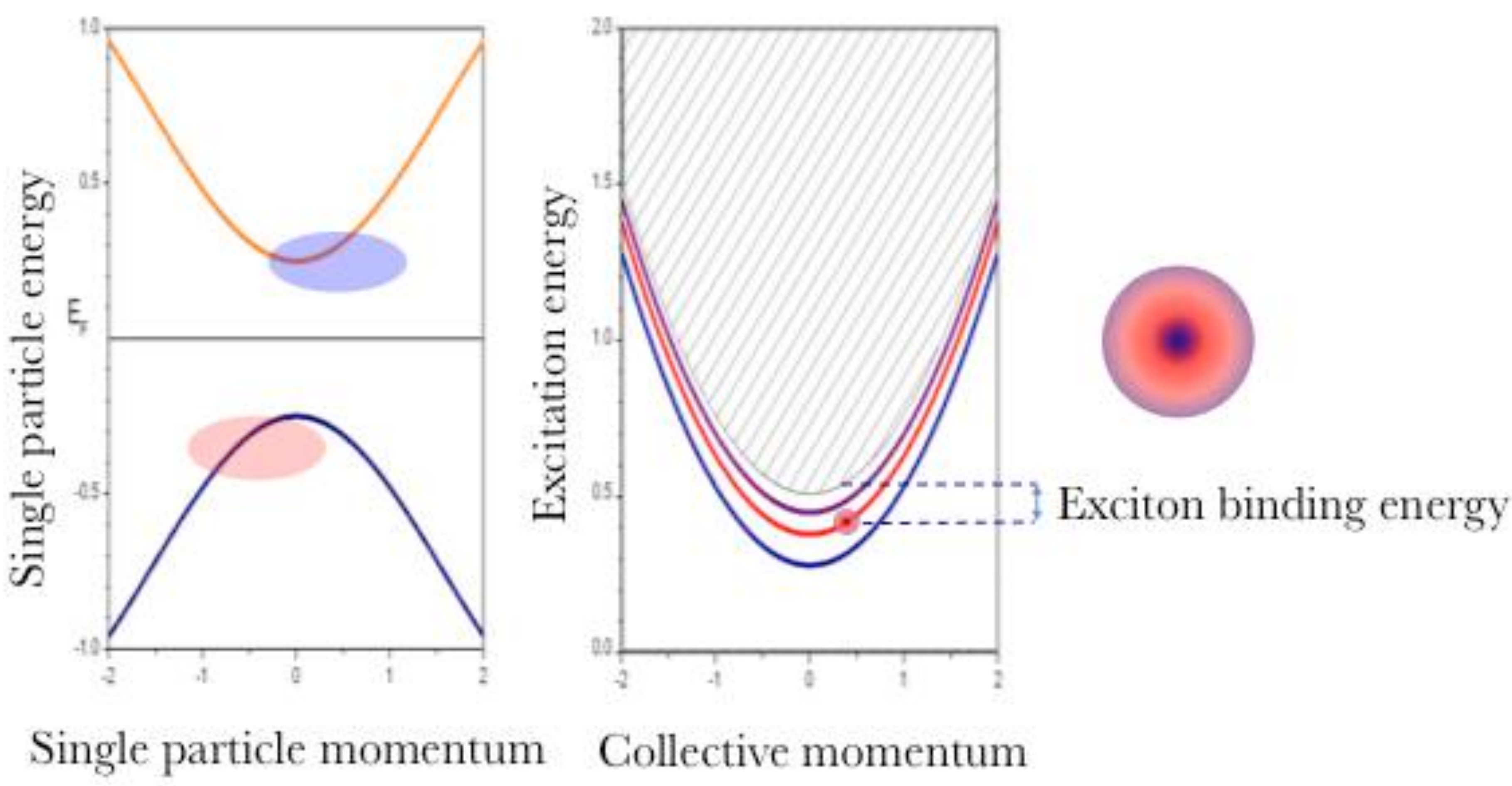


After D. E. Aspnes and A. A. Studna, Phys. Rev. B 27, 985 (1983)

Excitons



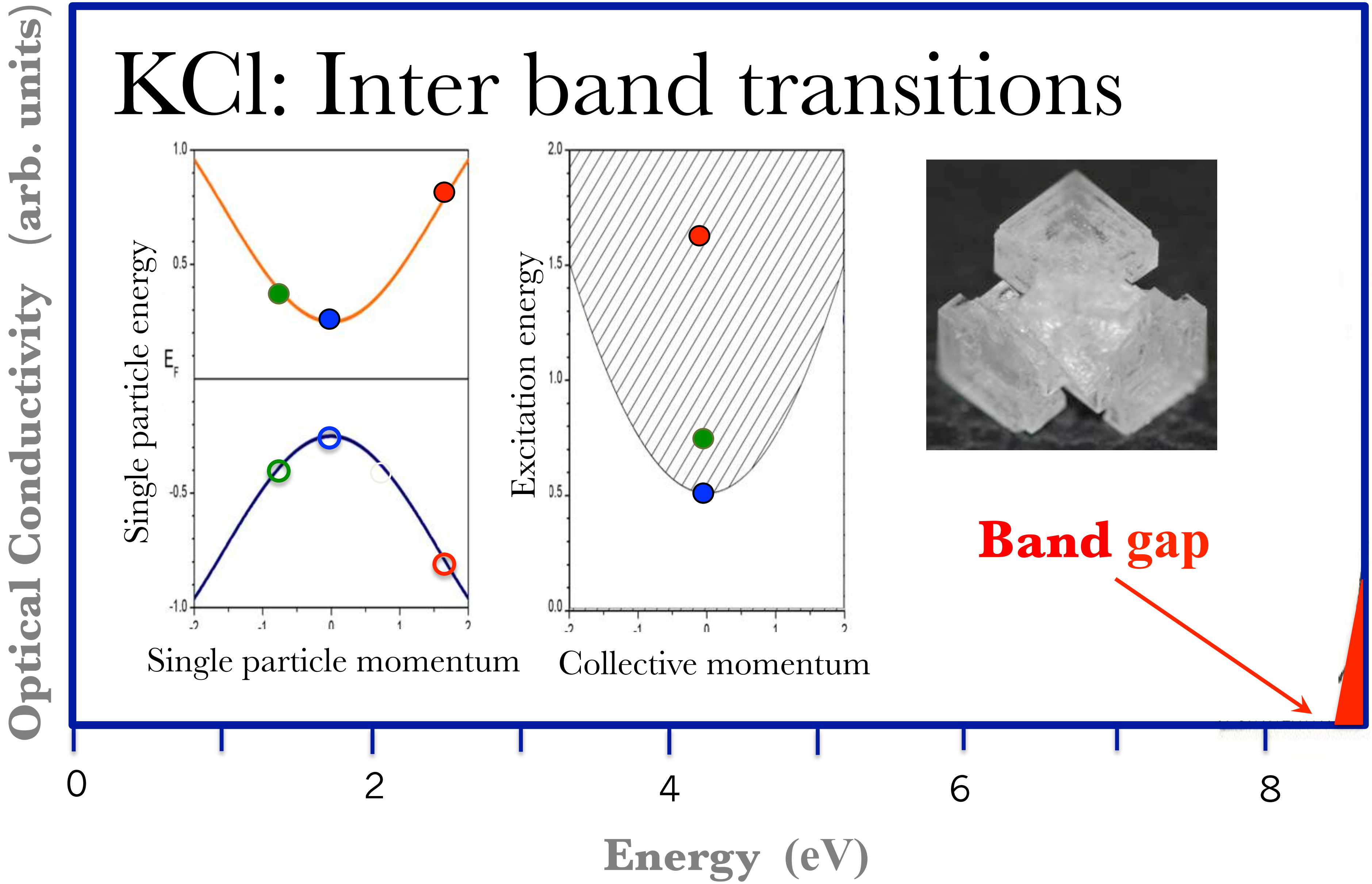
Excitons



Single particle energy
E_F
1.0
0.5
-0.5
-1.0
-2
-1
0
1
2
Single particle momentum
Excitation energy
2.0
1.5
1.0
0.5
0.0
-2
-1
0
1
2
Collective momentum
Exciton binding energy

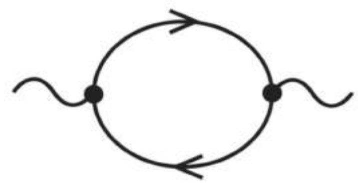
potassium chloride

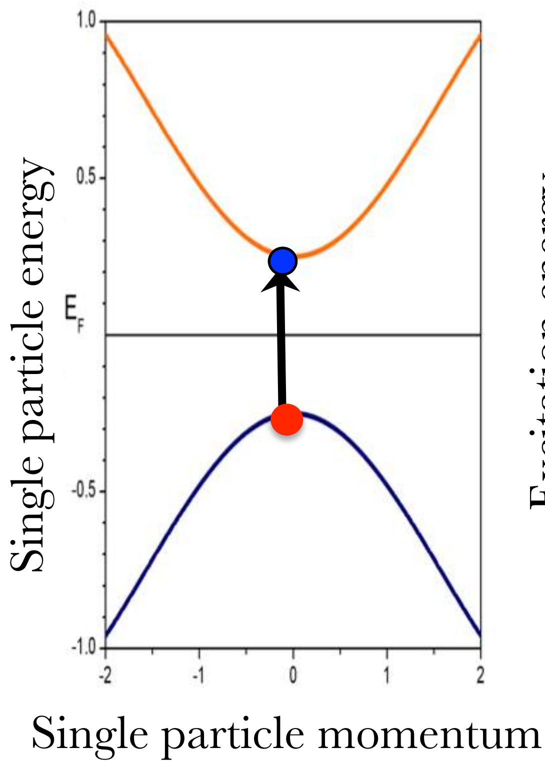
KCl: Inter band transitions

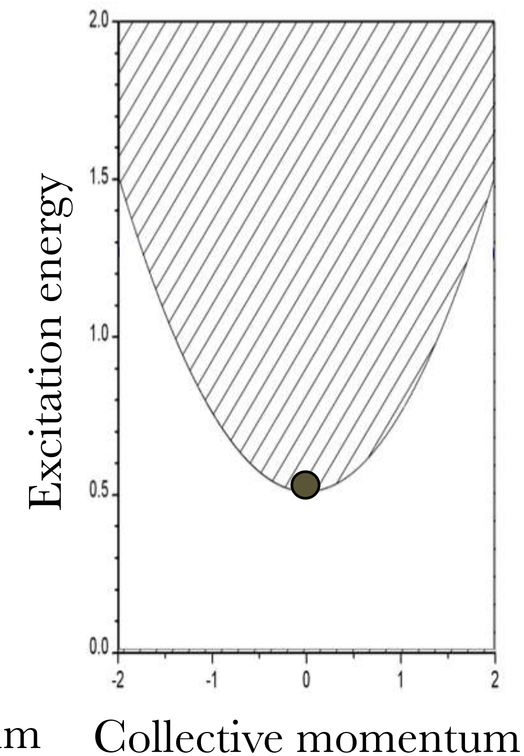


T. Tomiki, J. Phys. Soc. Jpn. 26, 738 (1969)

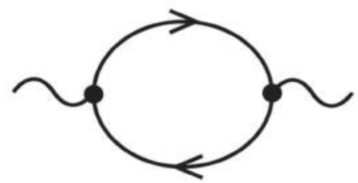
Optical excitation across the gap: electron-hole excitation

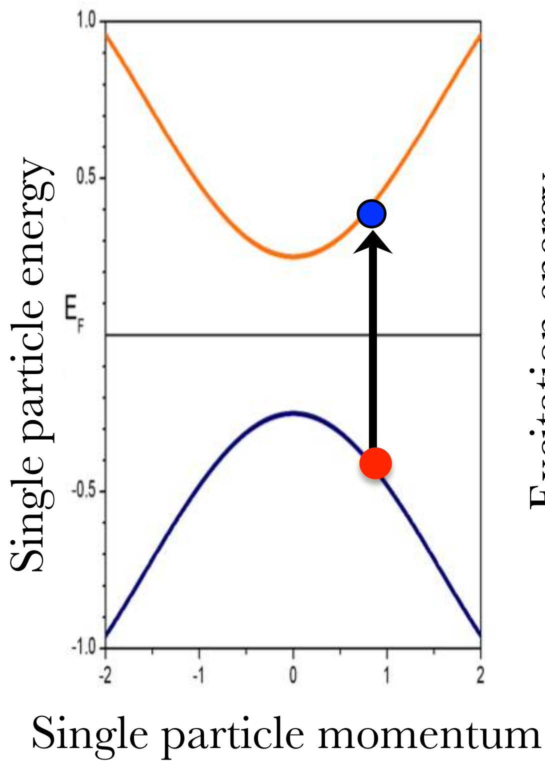


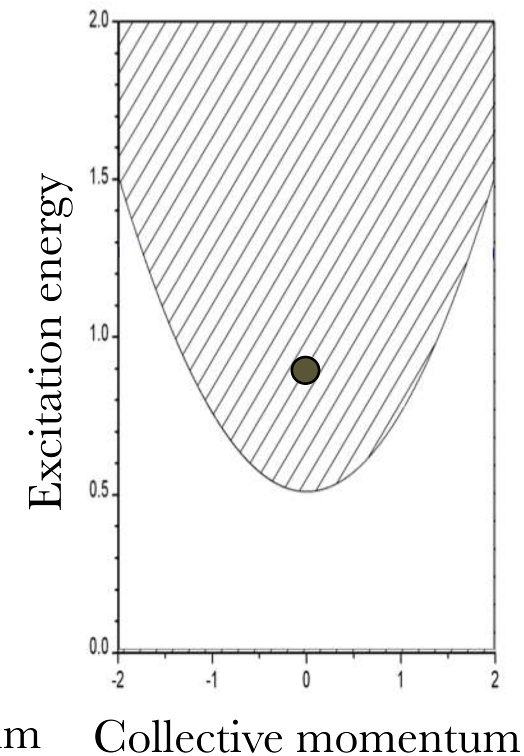




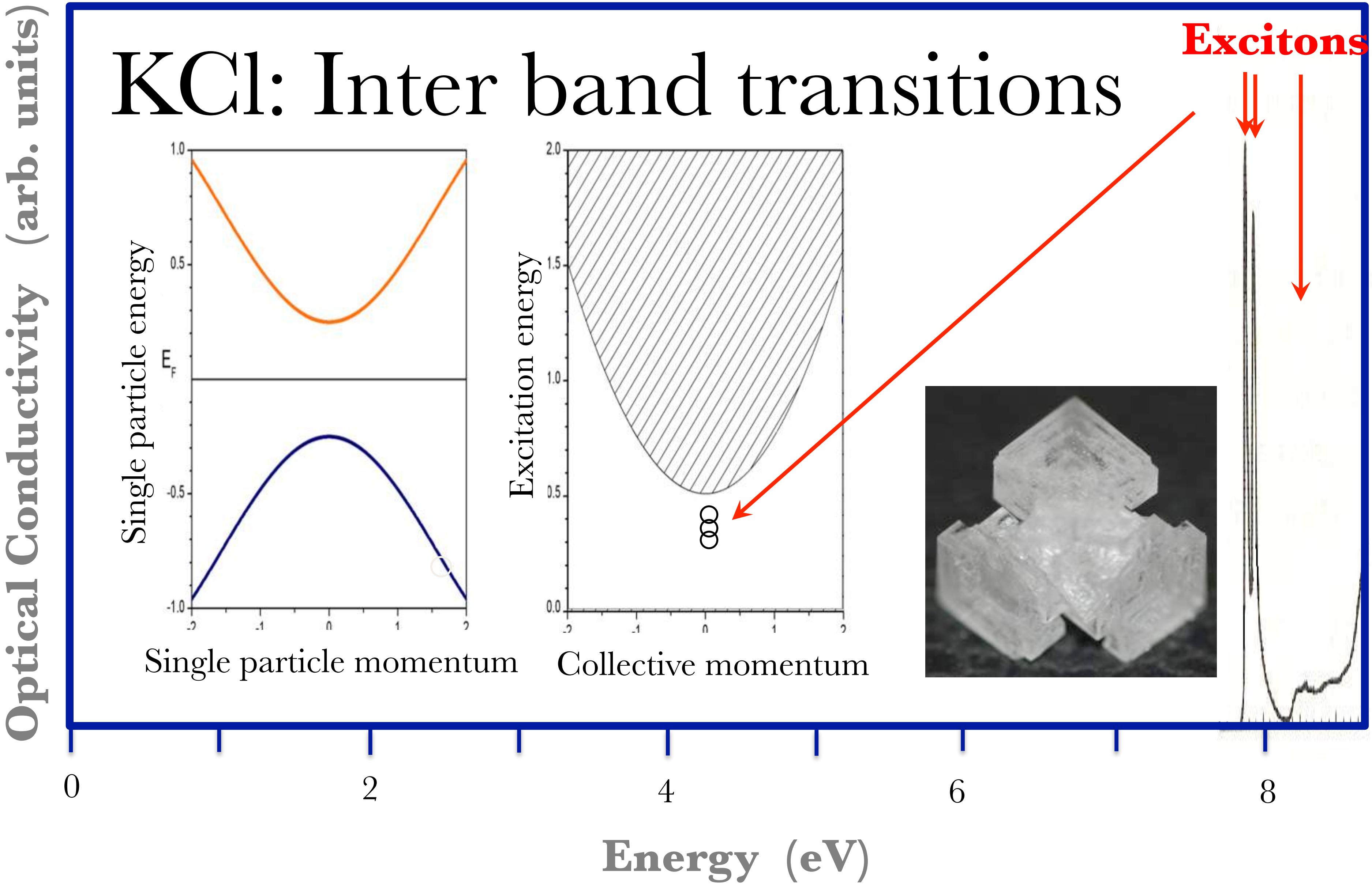
Optical excitation across the gap: electron-hole excitation





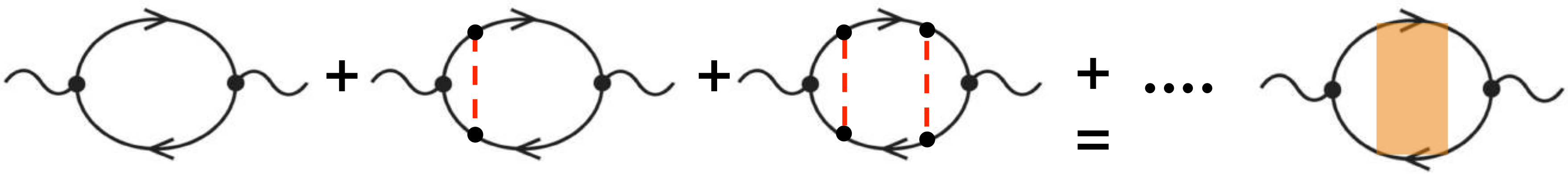


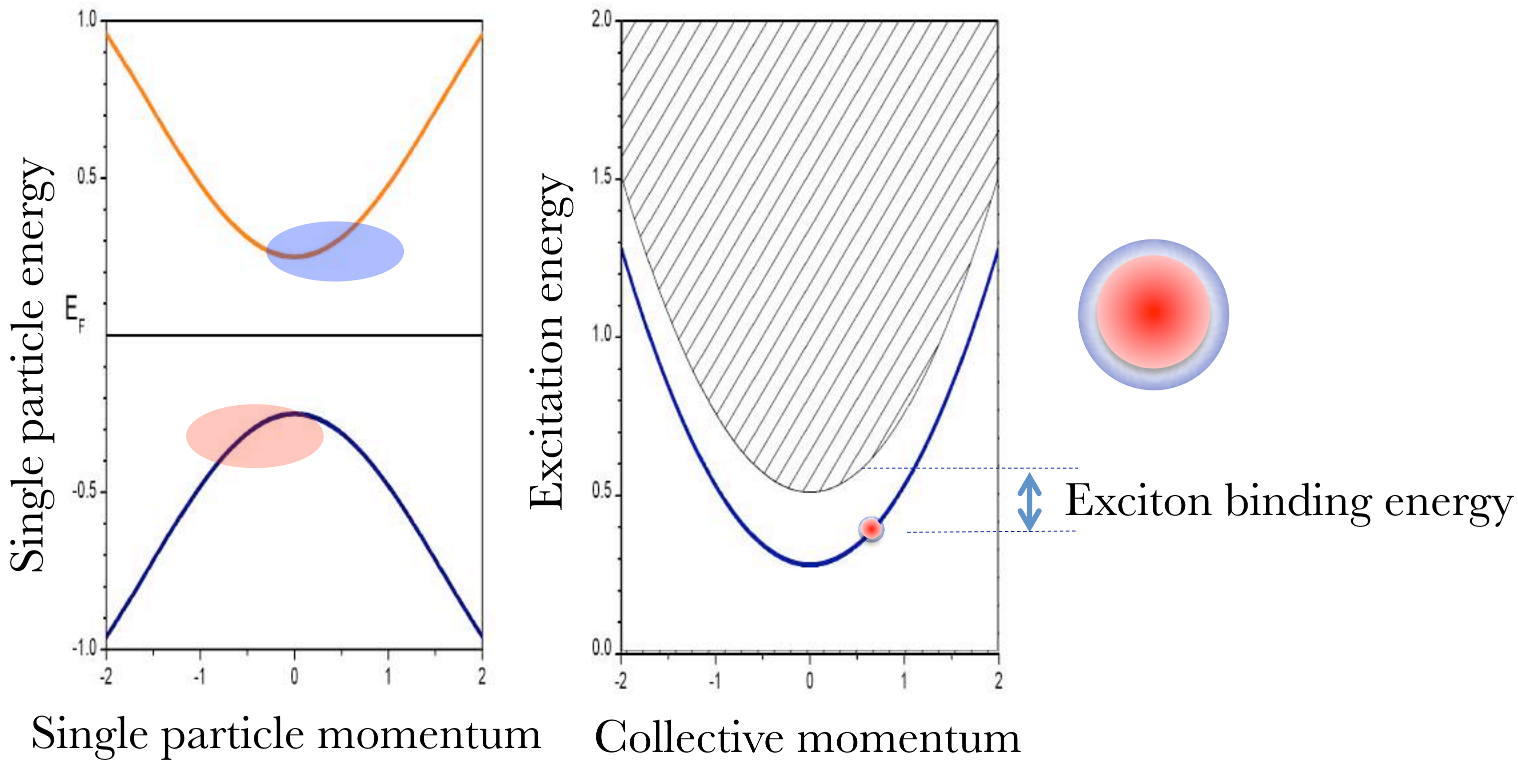
KCl: Inter band transitions



T. Tomiki, J. Phys. Soc. Jpn. 26, 738 (1969)

Electron + hole + Coulom interaction: Exciton = bound electron-hole state





Model: Free electron, free hole, and screened Coulomb interaction

$$H = \frac{P_{coll}^2}{2M} + \frac{p_{rel}^2}{2\mu} - \frac{e^2}{\varepsilon r_{rel}}$$

$$M = m_e + m_h$$

$$\mu^{-1} = m_e^{-1} + m_h^{-1}$$

Continuum state energy: $E_{gap} + \frac{\hbar^2 q^2}{2M} + \frac{\hbar^2 k^2}{2\mu}$

Bound state energy: $E_{gap} + \frac{\hbar^2 q^2}{2M} - E_{B,n}$

Exciton binding energy: $E_{B,n} = \frac{Ry^*}{n^2}$

Effective Rydberg: $Ry^* \equiv \frac{\mu e^4}{2\varepsilon^2 \hbar^2}$

$$\Rightarrow \mathrm{Re}\,\sigma(\omega) = \sum_n f_n \ \delta\left(\omega - E_{gap} + \frac{Ry^*}{n^2}\right) + \int_0^\infty dk \ f(k) \ \delta\left(\omega - E_{gap} - \frac{\hbar^2 k^2}{2\mu}\right)$$

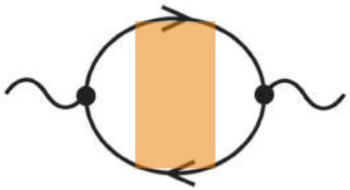
Exciton Binding energy

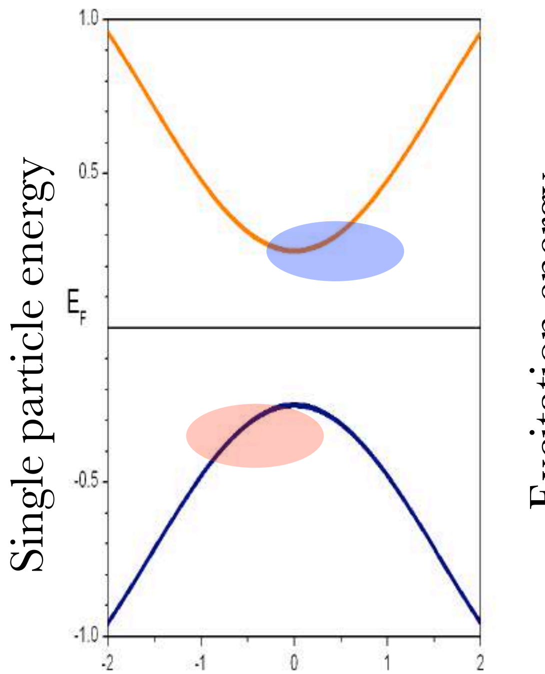
$$E_B = -\frac{Ry^*}{n^2} \qquad \text{where}: \quad Ry^* \equiv \frac{\mu e^4}{2\varepsilon^2\hbar^2}$$

Some crude estimates of exciton binding energies

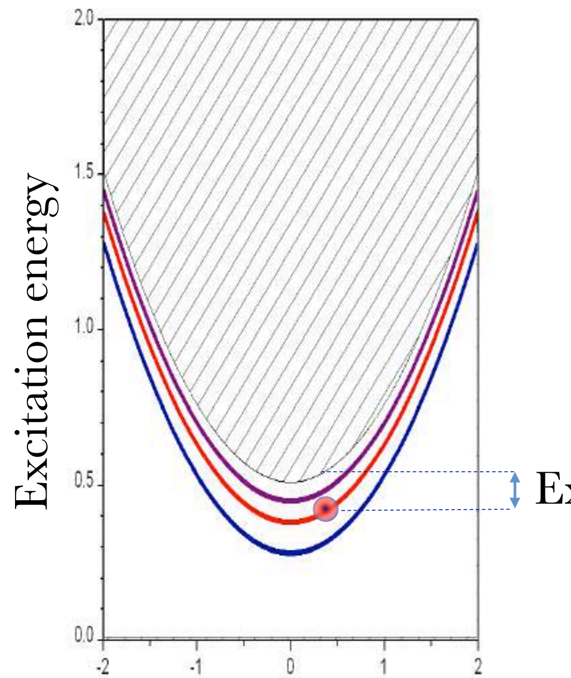
	$\mathbf{m_e}$	$\mathbf{m_h}$	**ε** Vis / IR	**Ry*** **(eV)**
Positronium	1	1	1	7
KCl	~1	~1	~5	~0.3
$SrTiO_3$	~1	~1	~5 / ~10^4	~0.3 / ~10^{-7}
Cu_2O	~1	~1	~7	~0.1

Exciton flavours - n,L,S quantum numbers

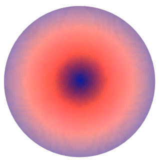




Single particle momentum

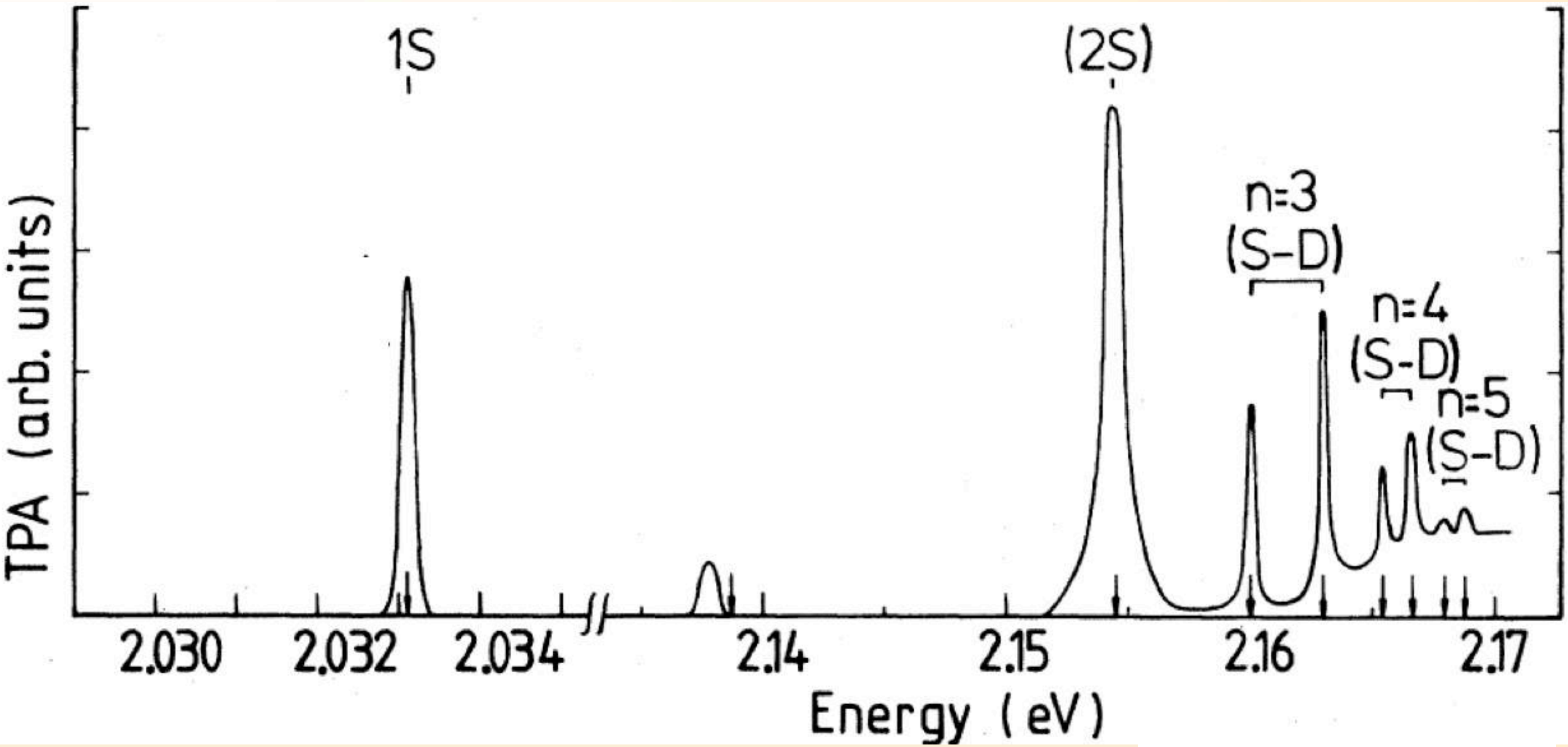


Collective momentum



Cu_2O

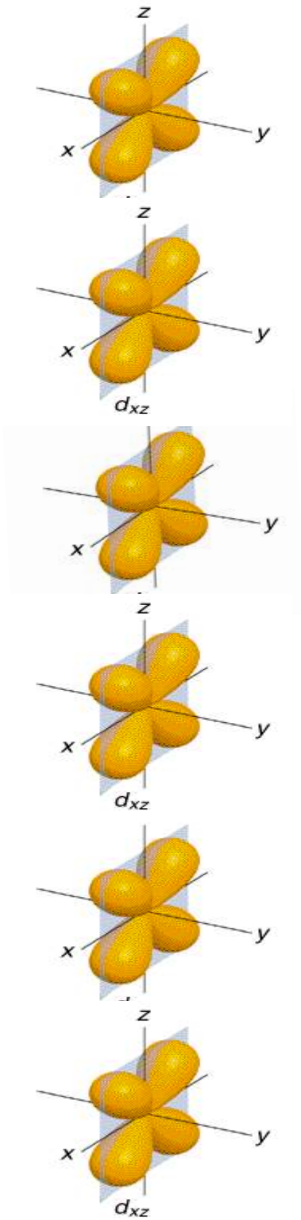
Cu_2O: Exciton flavours – quantum numbers

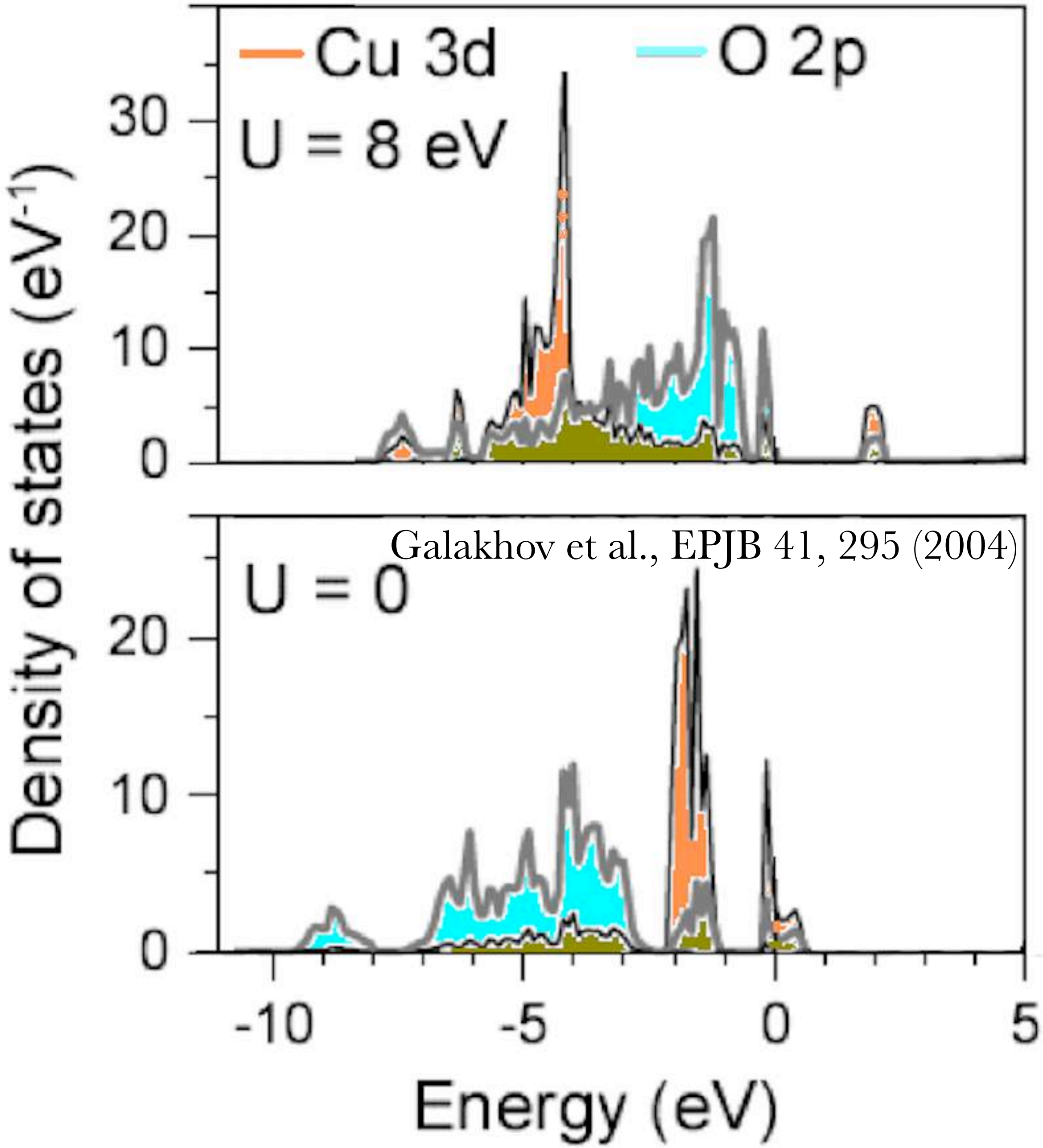


Band gap: 2.17 eV

Uihlein et al, PRB 23, 2734 (1981)

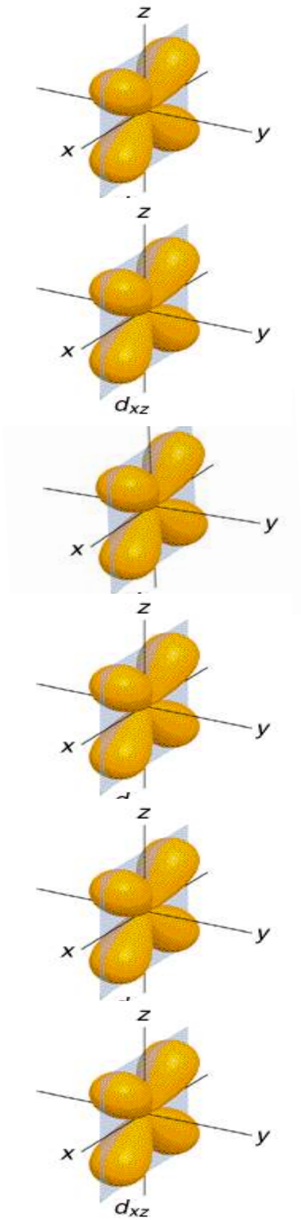
Excitons in a Mott-Insulator: $CuGeO_3$

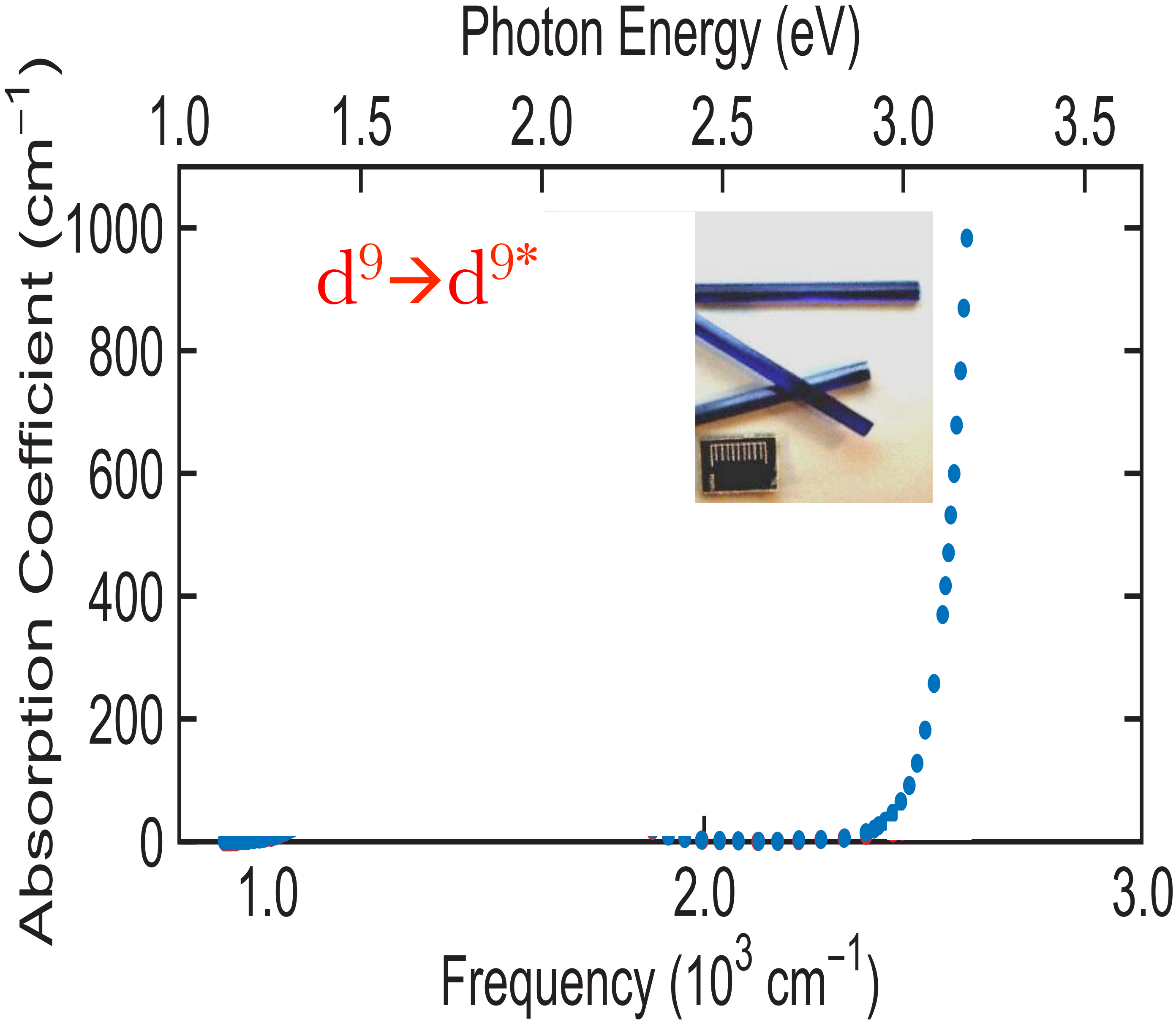




Galakhov et al., EPJB 41, 295 (2004)

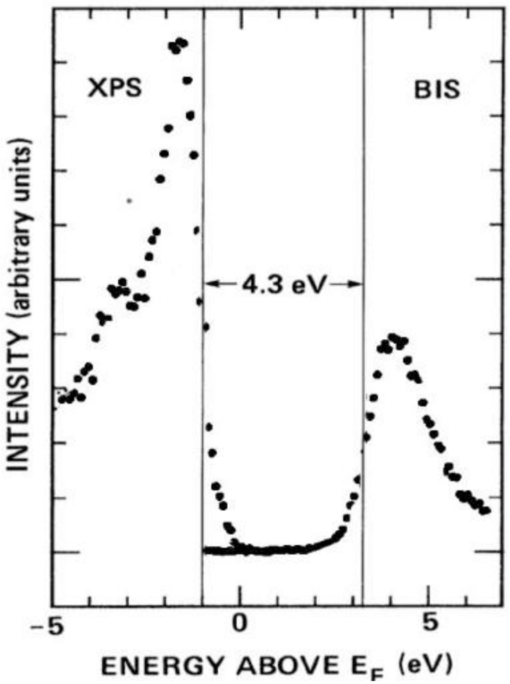
$CuGeO_3$: Intra-atomic excitons





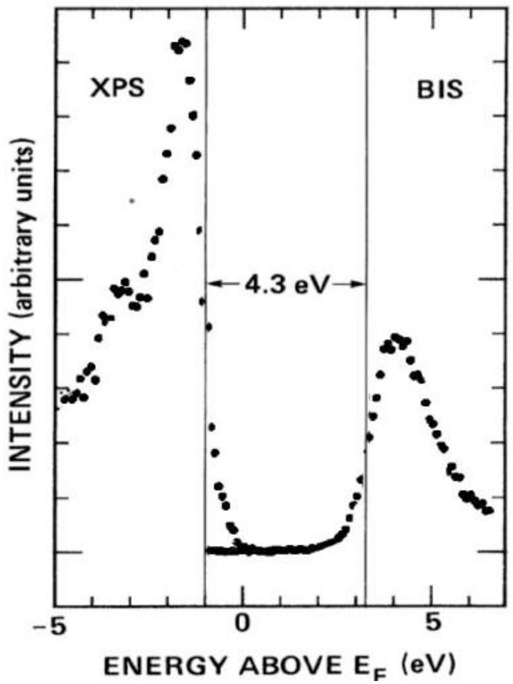
Bassi et al., 1996, PRB 54, R11030.

NiO: Intra-Atomic Excitons

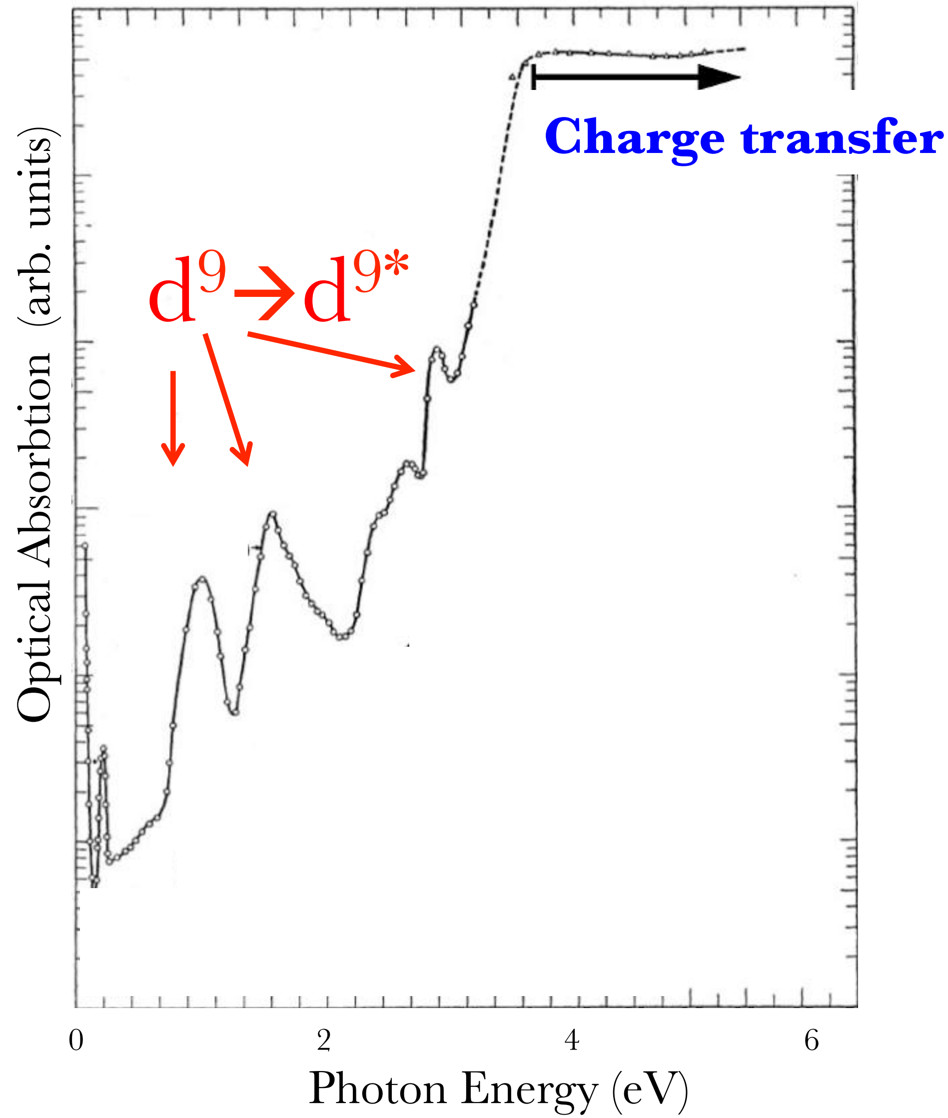


Sawatzky&Allen PRL 53, 2339 (1984)

NiO: Intra-Atomic Excitons



Sawatzky&Allen PRL 53, 2339 (1984)



Newman&Chrenko, Phys. Rev. 114, (1959)

Summary part I
Insulators – Interactions - Excitons

F-sum rule: The optical conductivity counts each electron

Interactions: EM-field "sees" *composite* particles (e.g. ions, phonons)

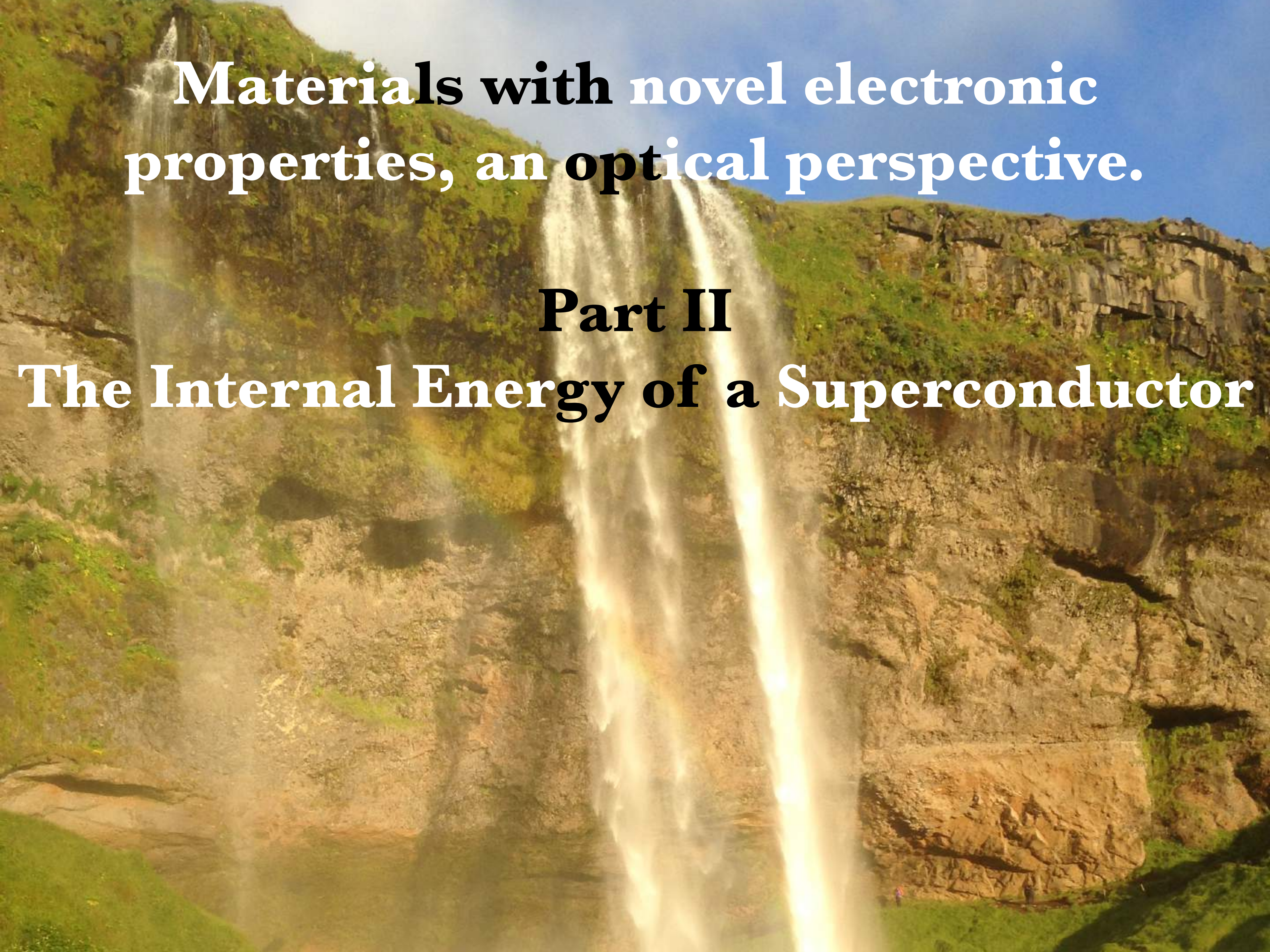
Excitons: Bound electron-hole pairs with zero charge

Normal insulators

- Extended e-h orbital
- Binding energy typically << gap

Mott Hubbard insulators

- Electron-hole paire confined to an atomic site
- Binding energy can be as big as the Mott-gap



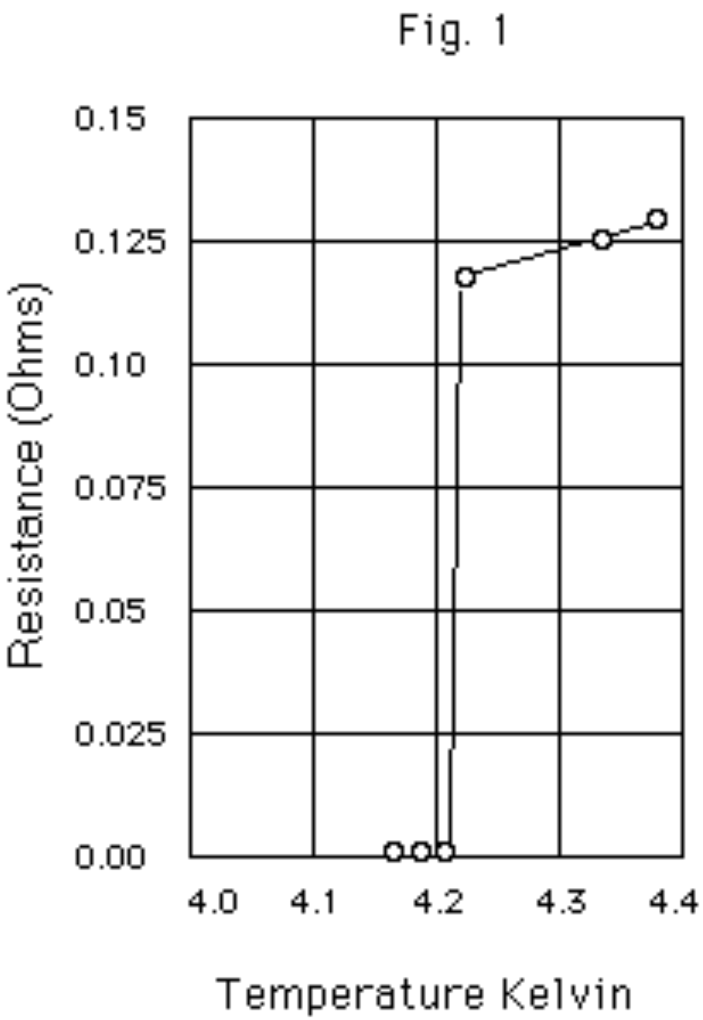
Materials with novel electronic properties, an optical perspective.

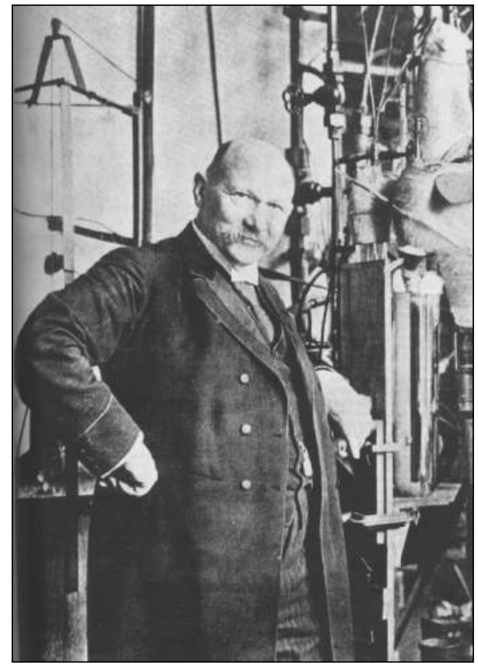
Part II
The Internal Energy of a Superconductor

Once Upon a Time in the West of the Netherlands

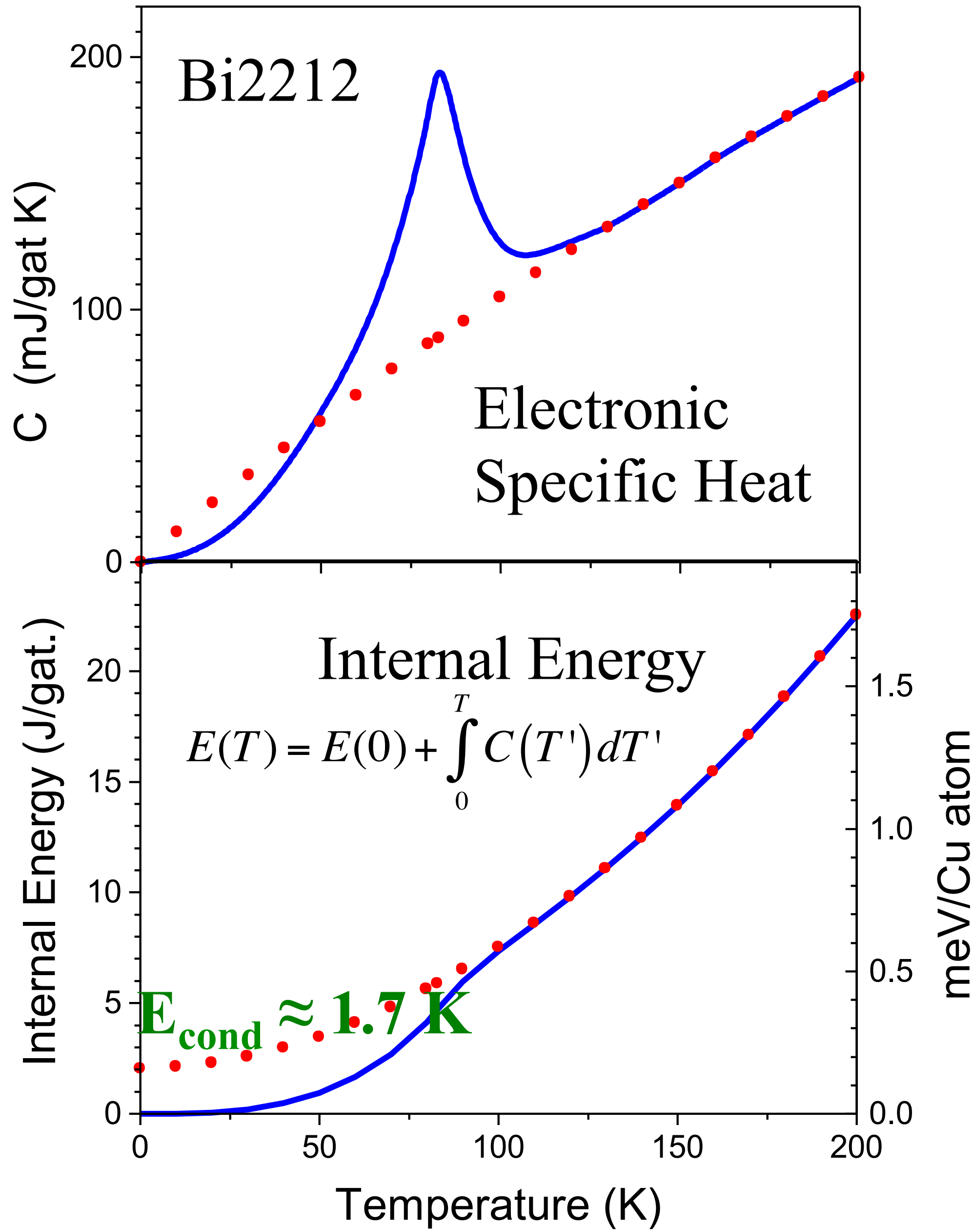
mercury

H. Kamerling Onnes





J. W. Loram et al., J. Phys. Chem. Solids 62, 59-64 (2001).



What about the Coulomb correlation energy ?

Virial Theorem and Superconductivity

G. V. Chester, PR 103, 1693 (1956).
A. J. Leggett, PNAS 96, 8365 (1999).

$$\hat{H} = \hat{H}_{kin} + \hat{V}_C \quad \textit{(electrons and nuclei)}$$

Coulomb interaction energy: $E_C = \langle \hat{V}_C \rangle$

Virial Theorem: $\langle \hat{H}_{kin} \rangle = -\frac{1}{2} E_C \Rightarrow E = \langle \hat{H} \rangle = \frac{1}{2} E_C$

Thermodynamics: $E^{sc} < E^n$

The Virial theorem than implies that : $E_C^{sc} < E_C^n$

Charge susceptibility and Coulomb energy

P. Nozières and D. Pines, PR 111 (1958)

Pair distribution function: $g(r,t;r',t')$

(Probability that another particle is at coordinate r', if there is already one at r)

Structure factor $(t=t')$: $S_q = \int e^{iqr} g(r,0)\,dr$

Dynamic structure factor
(see Lucia Reining's course on friday)

Fluctuation dissipation theorem: $S_q = \frac{\hbar}{2\pi}\int_0^\infty \operatorname{Im}\chi(q,\omega)\left[1+2n_B(\omega/T)\right]d\omega$

Linear response theory: $V_q \operatorname{Im}\chi(q,\omega) = \operatorname{Im}\frac{-1}{\varepsilon(q,\omega)} = L(q,\omega)$

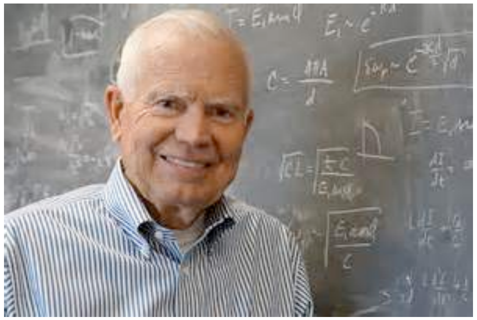
($L(q,\omega)$ is called "Energy Loss Function")

Many-body Coulomb energy:

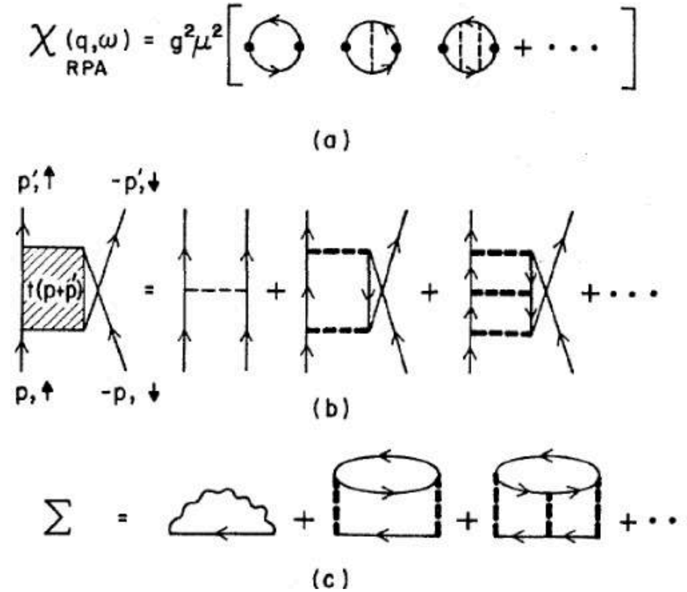
$$\begin{cases} \left\langle V_{C,q}\right\rangle_T = \frac{\hbar}{2\pi}\int_0^\infty L(q,\omega)(1+2n_B)\,d\omega \\ E_C(T) = \sum_q \left\langle V_{C,q}\right\rangle_T \end{cases}$$

How to get a bound fermion pair out of repulsion

Effective pairing interaction mediated by spin fluctuations



D. J. Scalapino



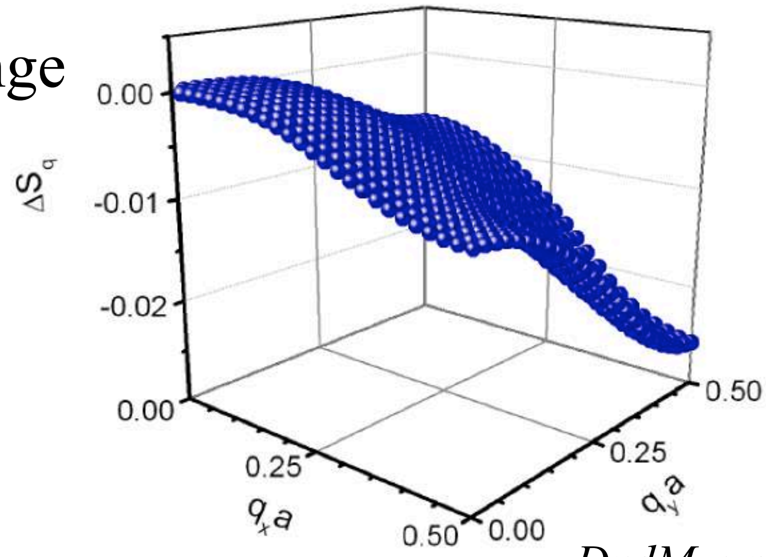
1) The main energy saving comes from the exchange of *S=1* fluctuations

2) The important momentum range is *q≈ (π/a,π/a)*

T. A. Maier, M. Jarrell, and D. J. Scalapino, Phys. Rev. B 75, 134519 (2007)
N. Berk and J. Schrieffer, Phys. Rev. Lett. 17, 433 1966.

How to get a bound fermion pair out of repulsion

Superconductivity induced change of the structure factor:



DvdM, unpubl. (2016)

$$\Delta S_q = S_{q,SC} - \Delta S_{q,N}$$

→ *d-wave gap*

→ $V_q = e^2/q^2$ *(Coulomb)*

Eugene Demler & Shou-Cheng Zhang, Nature (1998):

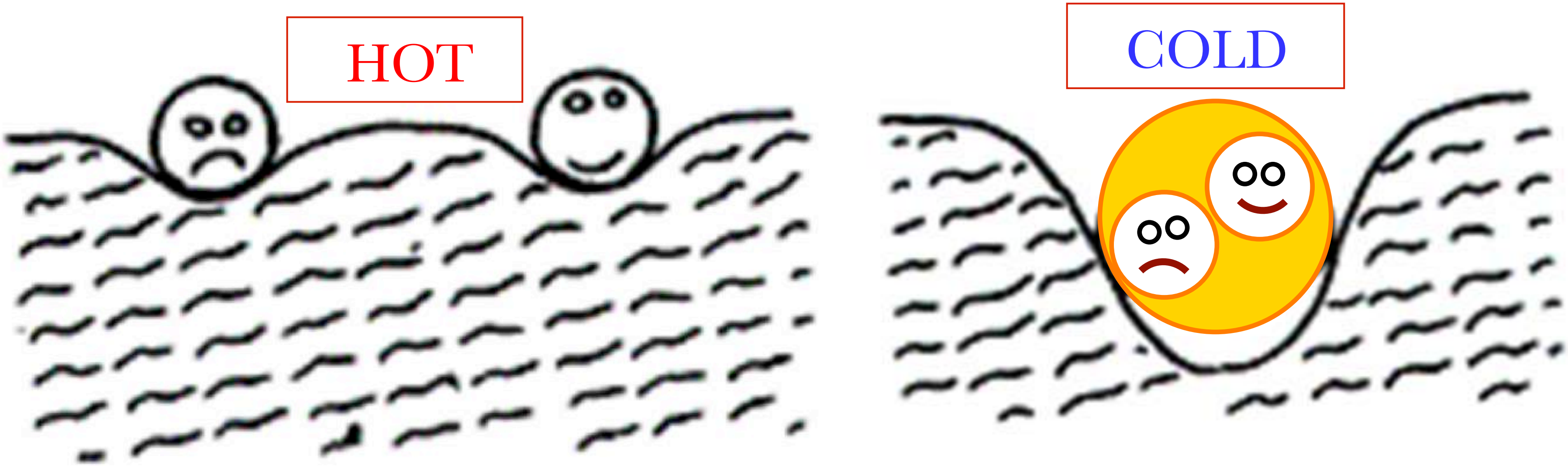
$$E_J = \frac{3J\hbar}{4\pi}\sum_q \left(\cos q_x a + \cos q_x a\right)\int_0^\infty d\omega S\left(q,\omega\right) + \ldots$$

$S\left(q,w\right)$ from **experimental** neutron scattering data: $E_J = 0.016J \sim 18K$

Conclusion: $\mathbf{E_J}$ **is big enough in comparison to** $\mathbf{E_{cond}} \sim \mathbf{1K}$

What does BCS theory tell about the Coulomb energy ?

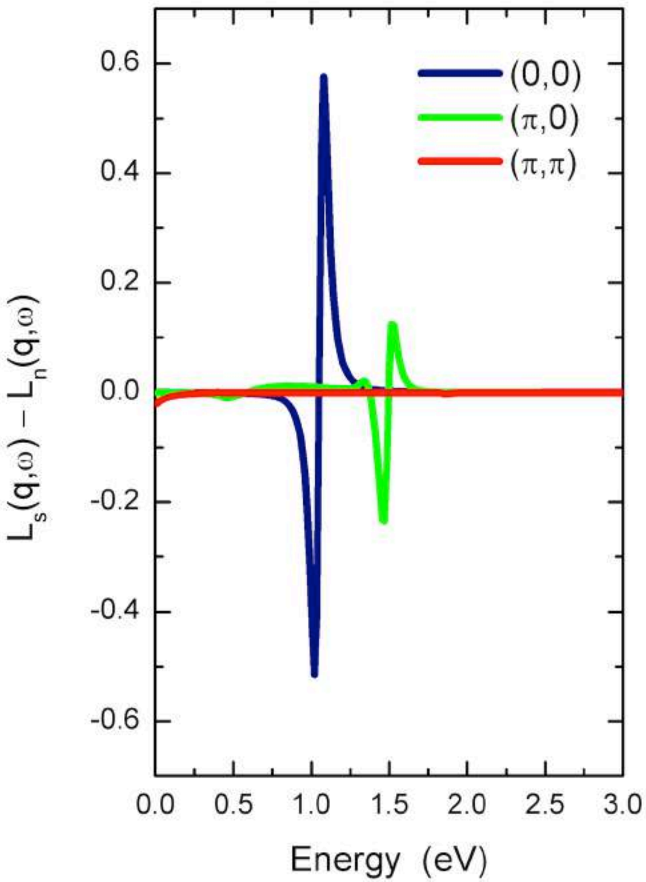
***Provided that* electrons <u>attract</u> each other.....**

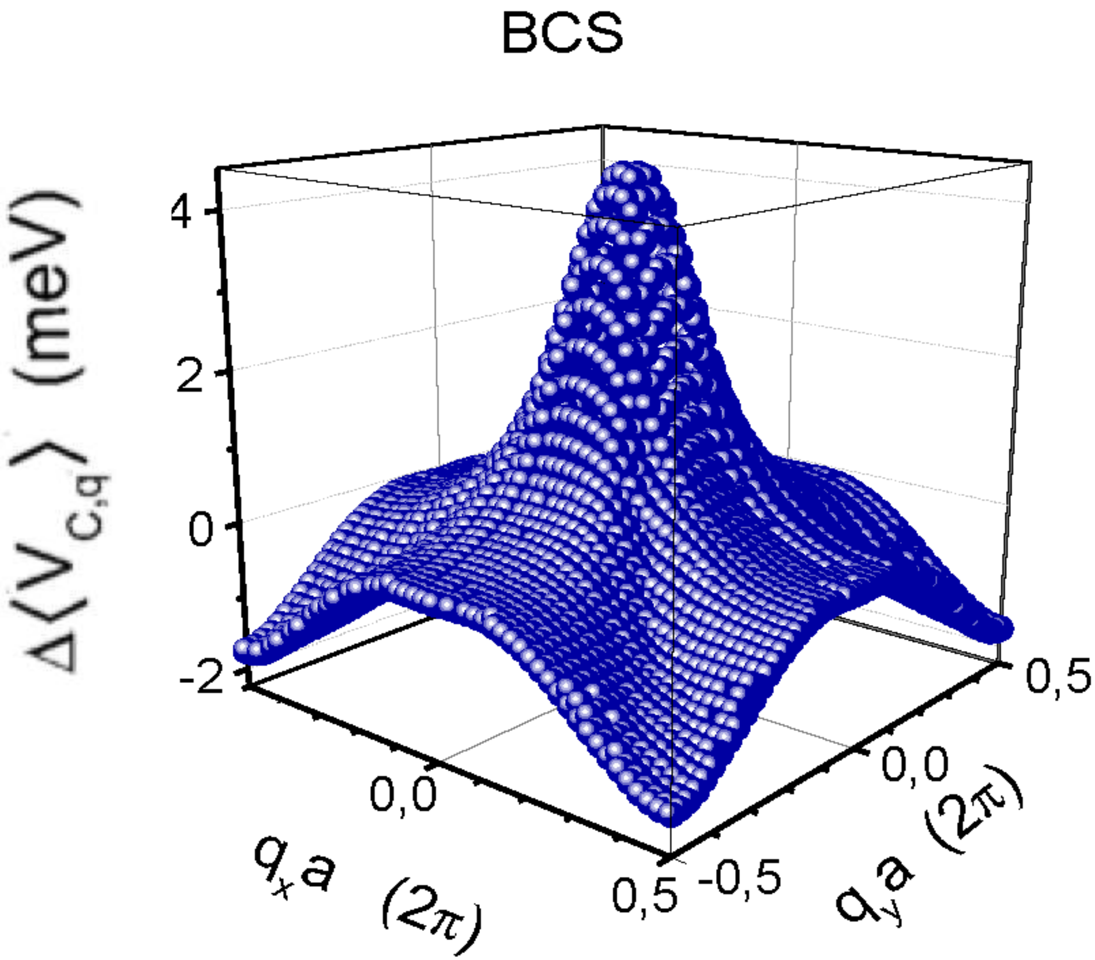


....they form Cooper pairs when $T < T_{BCS}$

What does BCS theory tell about the Coulomb energy ?

$$\left\langle V_{C,q}\right\rangle = \frac{\hbar}{2\pi}\int_0^{\infty} L(q,\omega)(1+2n_B)d\omega$$



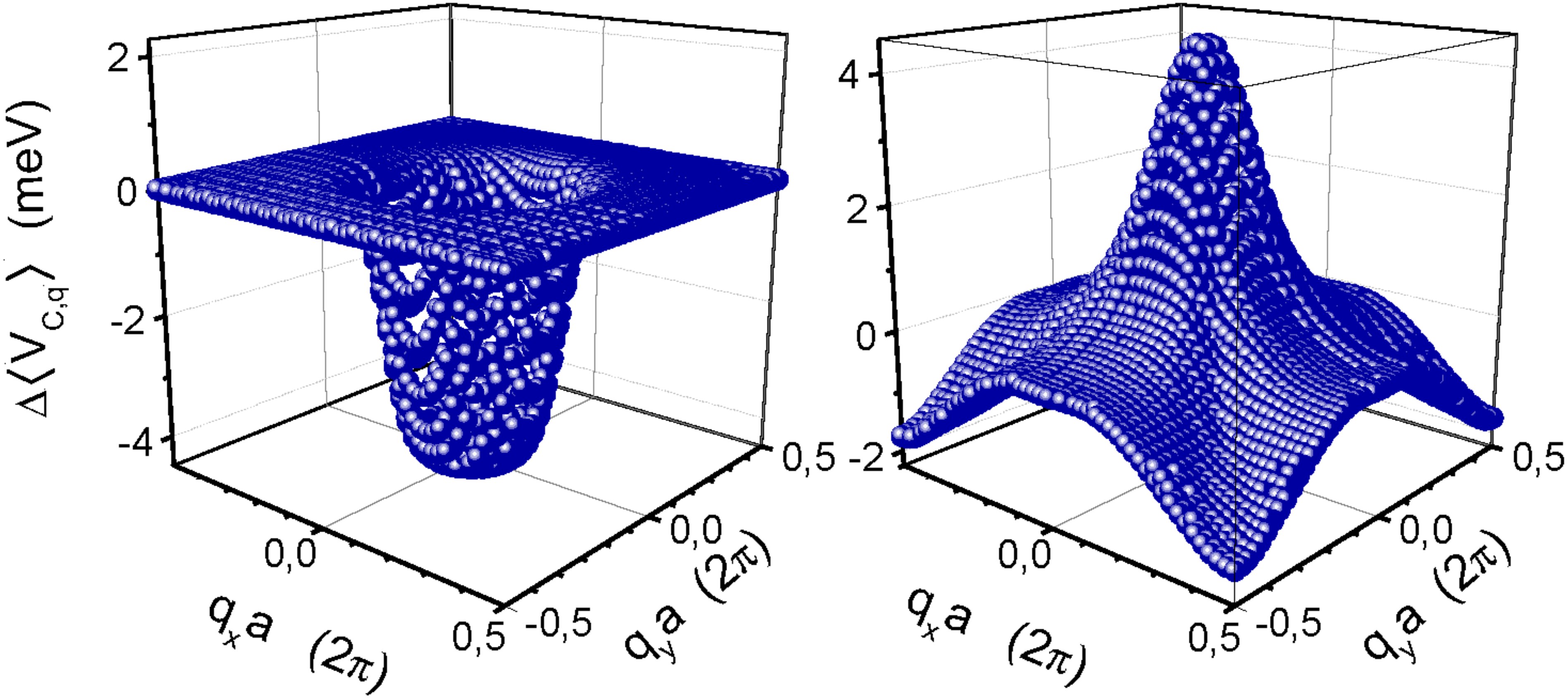


DvdM, unpubl. (2016)

A. J. Leggett's theory is radically different (PNAS 96, 8365 (1999))

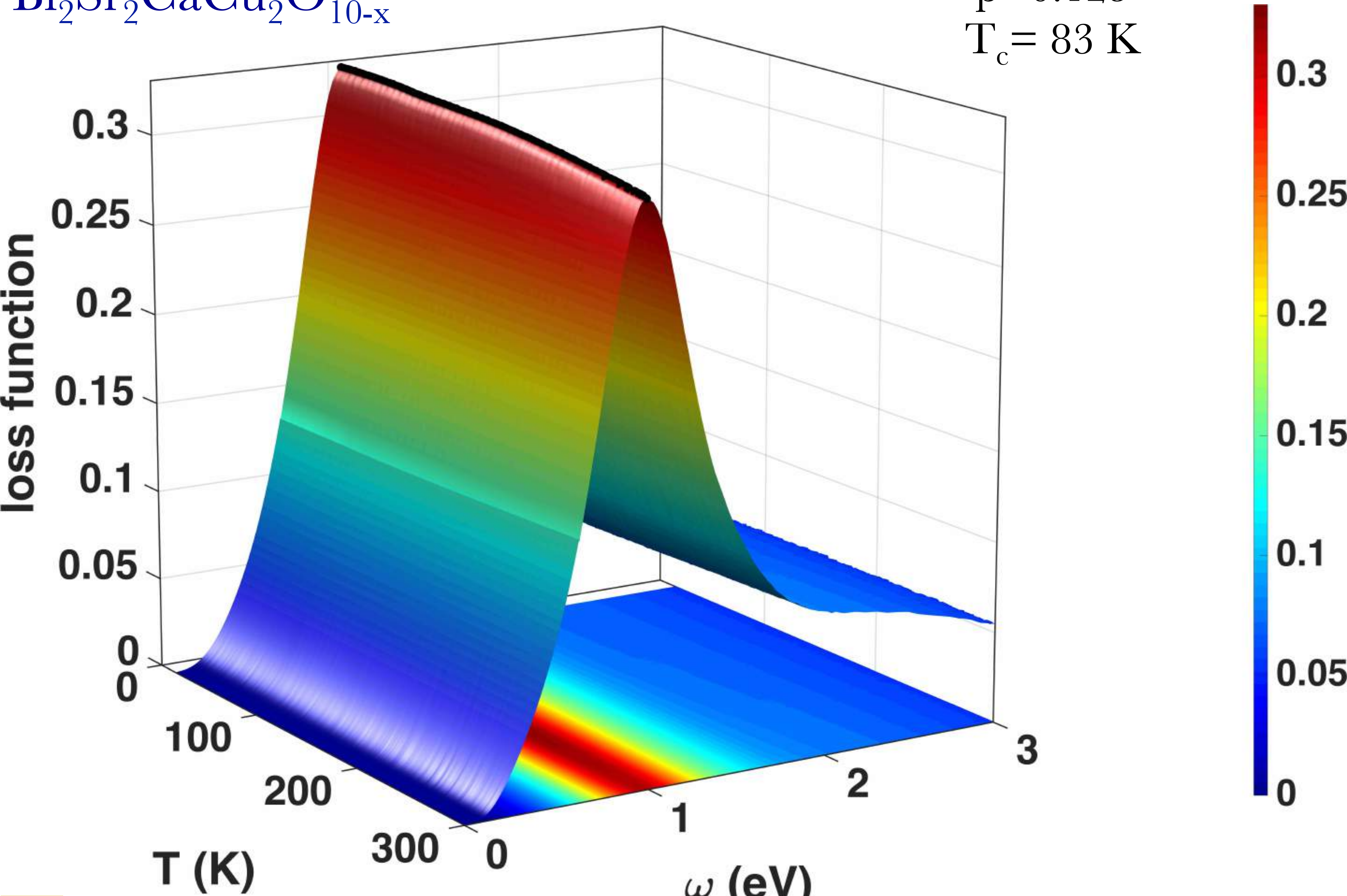
The main energy saving comes from the loss function peak for *$q < 1/\xi \approx 0.3$ Å^{-1}*   ("Willie Sutton principle »)

Leggett's theory   BCS theory



$Bi_2Sr_2CaCu_2O_{10-x}$

p=0.125
T_c= 83 K



J.Levallois et al.,arXiv:1512.00672.

$Bi_2Sr_2CaCu_2O_{10-x}$

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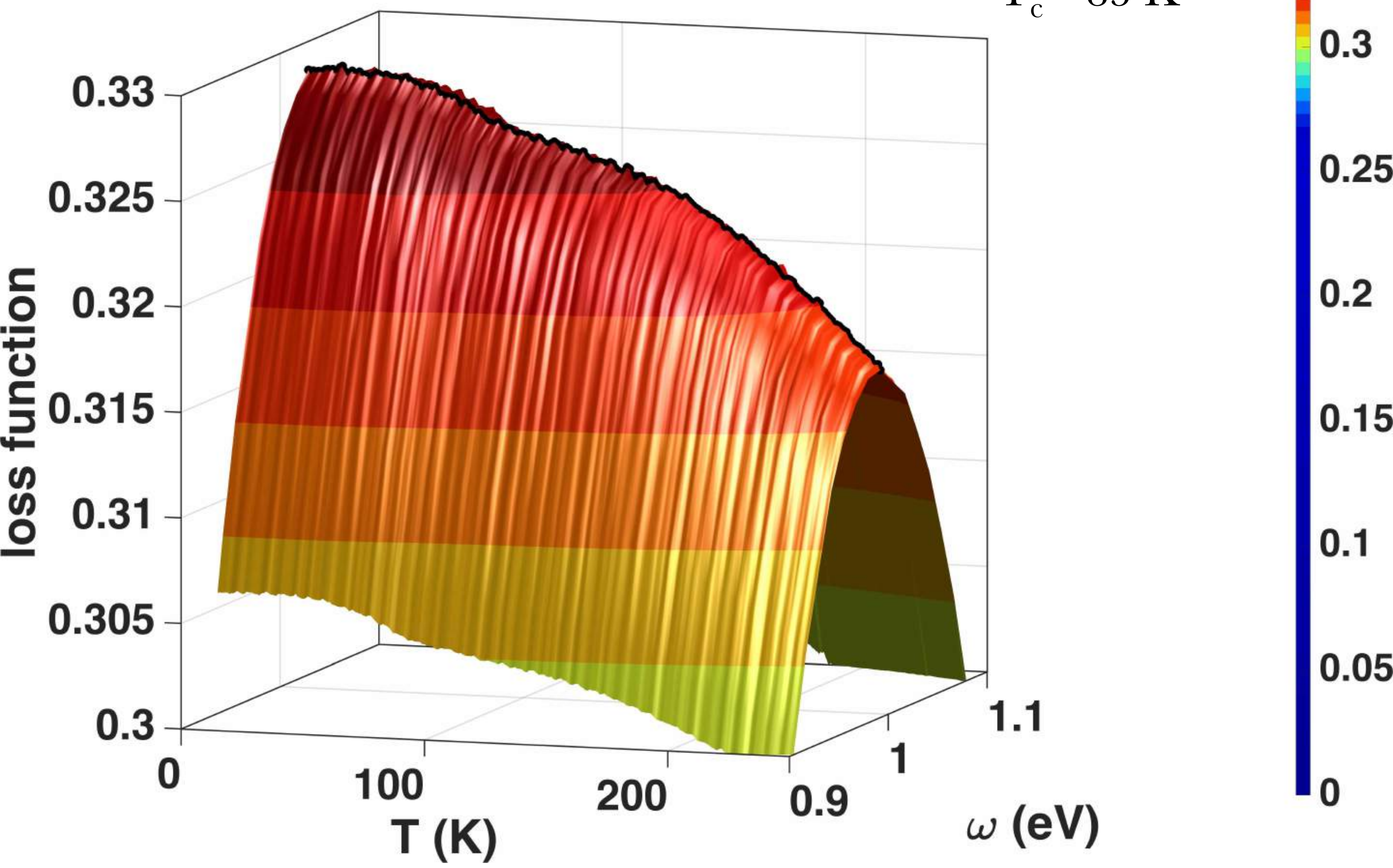
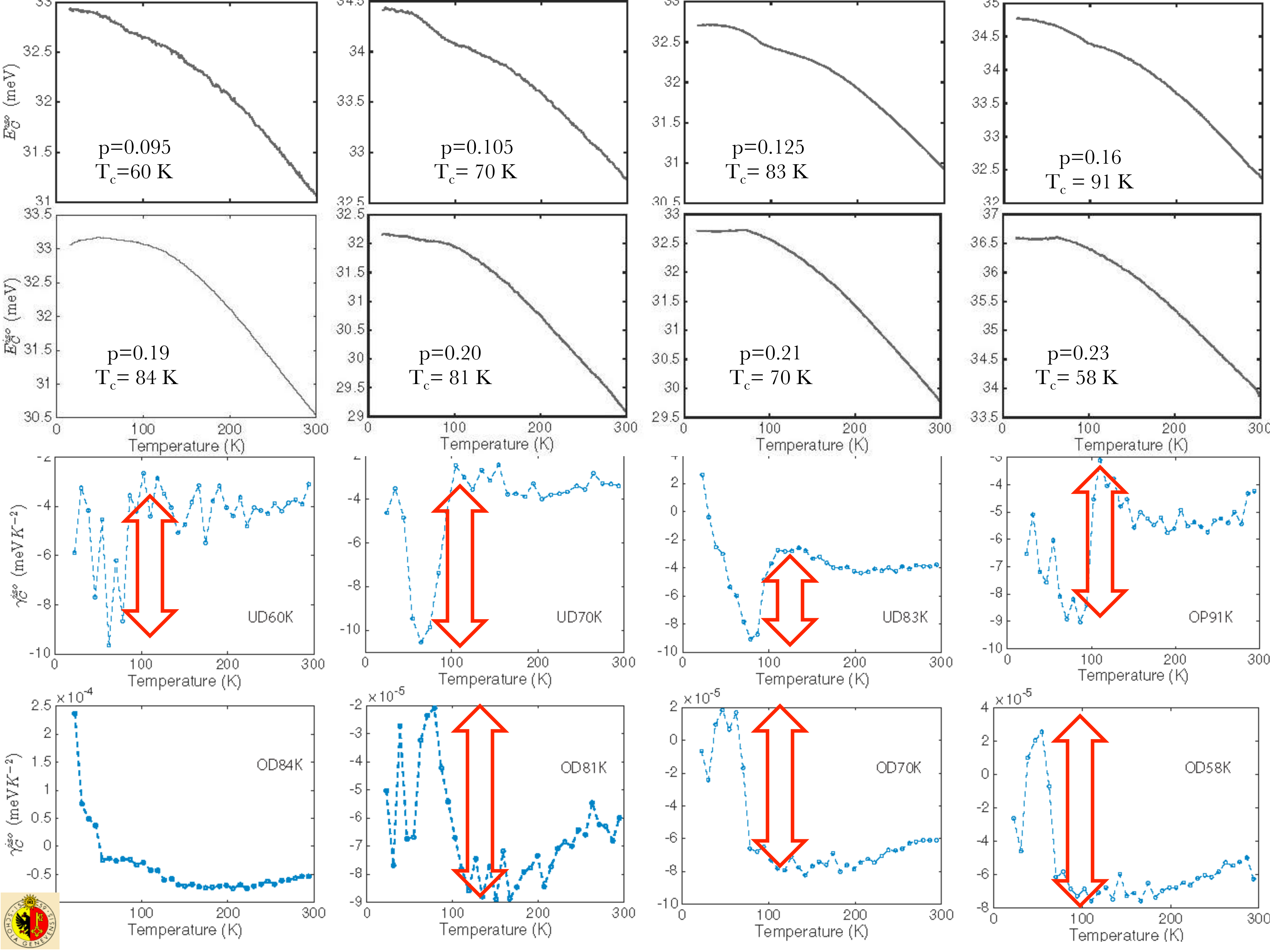
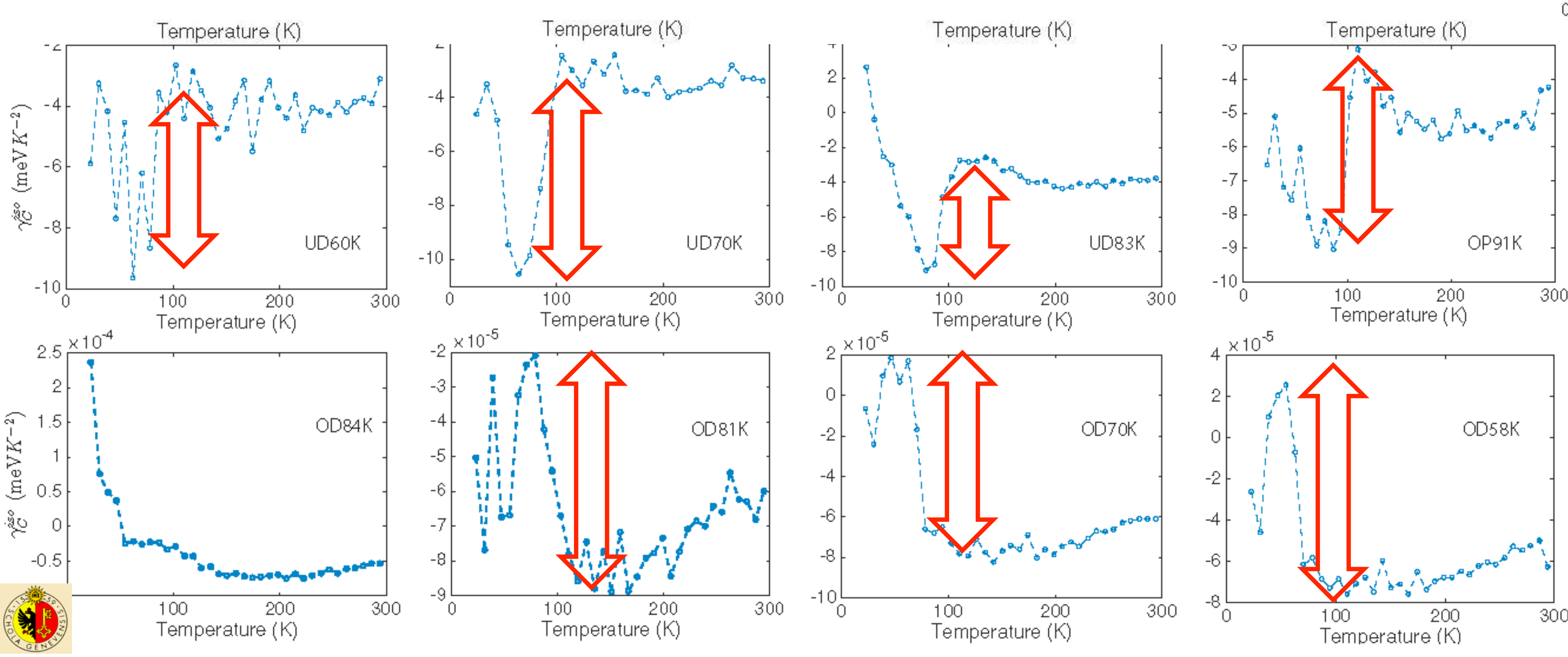


J.Levallois et al.,arXiv:1512.00672.

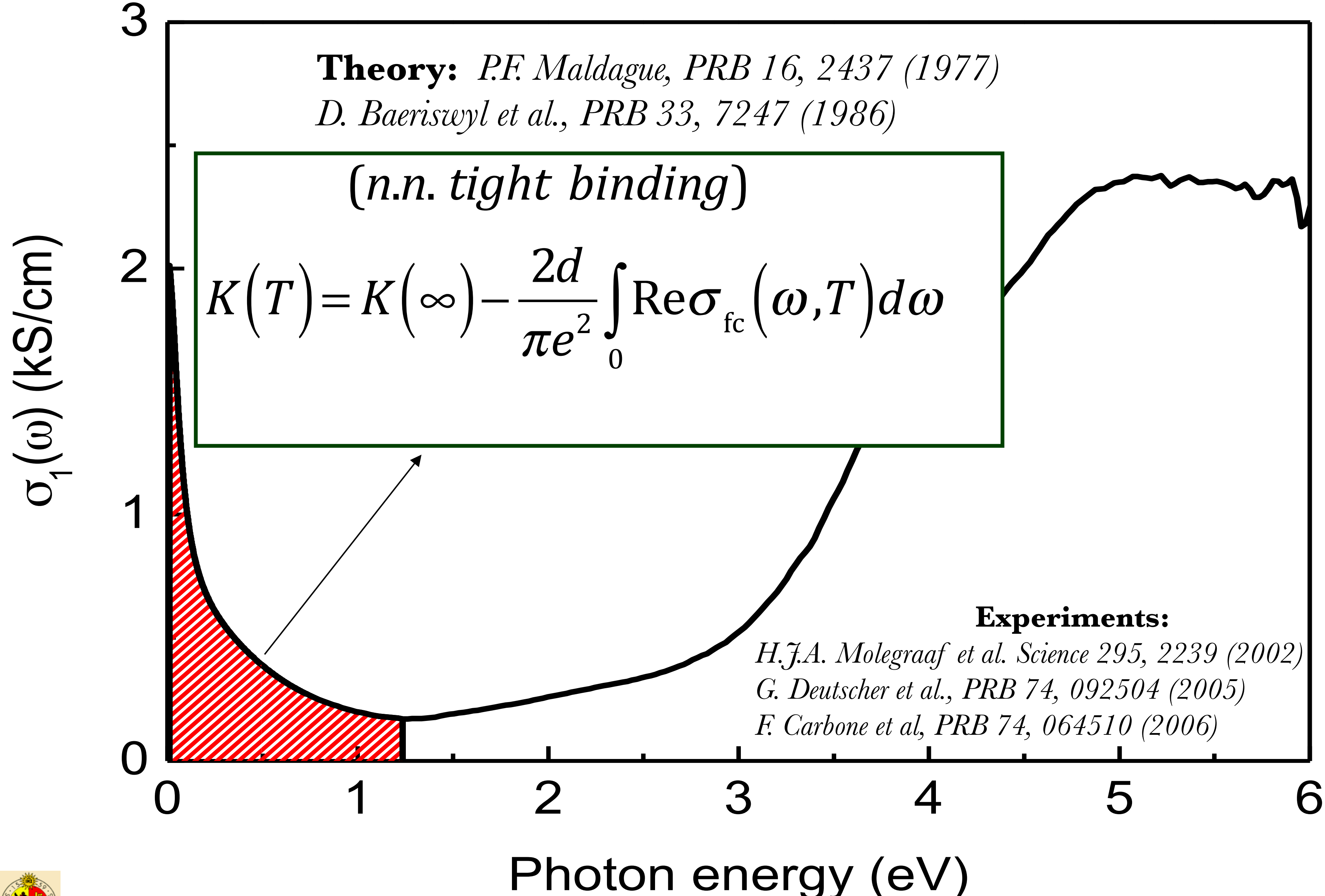


p=0.095
Tc=60 K
p=0.105
Tc= 70 K
p=0.125
Tc= 83 K
p=0.16
Tc = 91 K
p=0.19
Tc= 84 K
p=0.20
Tc= 81 K
p=0.21
Tc= 70 K
p=0.23
Tc= 58 K
(meV)
Temperature (K)
(meVK−2)
UD60K
UD70K
UD83K
OP91K
OD84K
OD81K
OD70K
OD58K

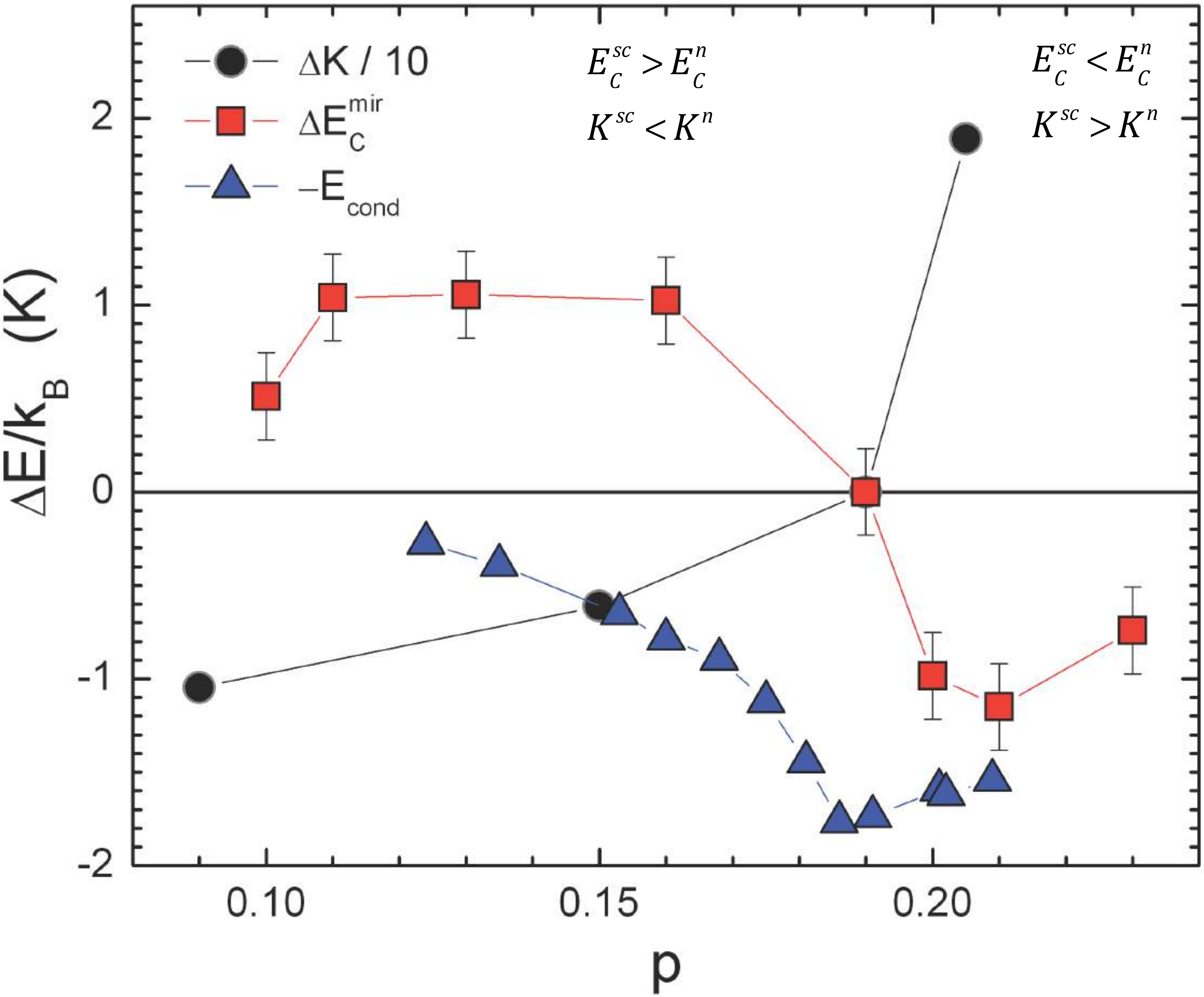
$$E_C^{sc}(0) - E_C^{n}(0) \cong \frac{1}{2}\Delta\gamma_C T_c^2$$



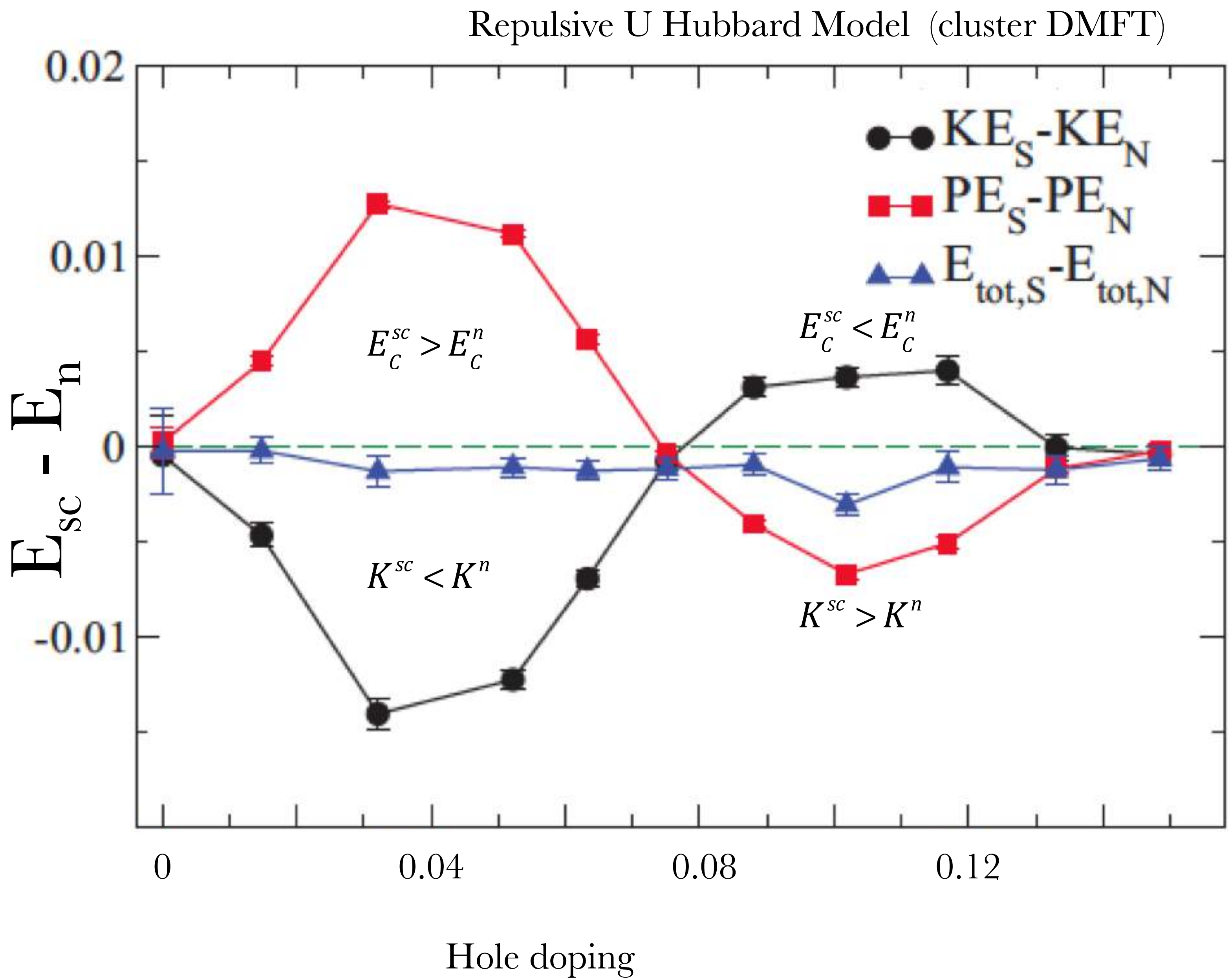
Relation between σ(ω) and K(T)



Quantitative: Coulomb, kinetic and total condensation energy



J Levallois, MK Tran, D Pouliot, CN Presura, LH Greene, J.N Eckstein, J Uccelli, E Giannini, GD Gu, AJ Leggett & DvdM, Phys. Rev. X ***6****, 031027 (2016)*



E. Gull and A. J. Millis, PRB 86, 241106 (2012)

SUMMARY PART II

Study of correlation functions : makes sense for correlated electrons

E_C (T) behaviour at T_c is opposite for $p < 0.19$ and $p > 0.19$.

Underdoped region disagrees with Legget's MIR scenario

Overdoped region agrees *qualitatively* with Legget's MIR scenario

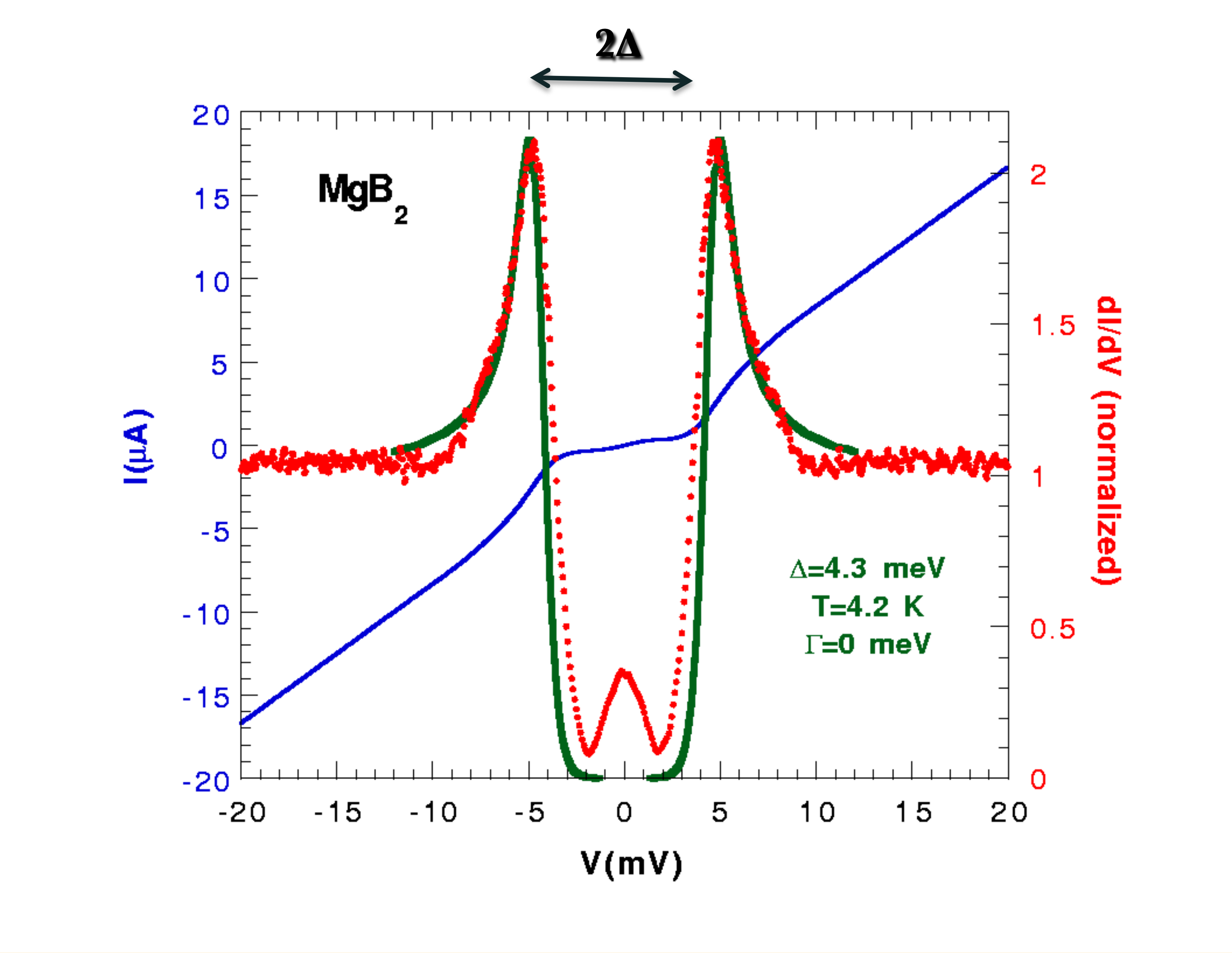
***Quantitatively* the saving of small-momentum Coulomb energy is obviously insufficient**

The doping dependence of $\Delta K(T)$ is opposite to that of $\Delta E_C(T)$

These trends agree qualitatively with the Hubbard model (CDMFT)

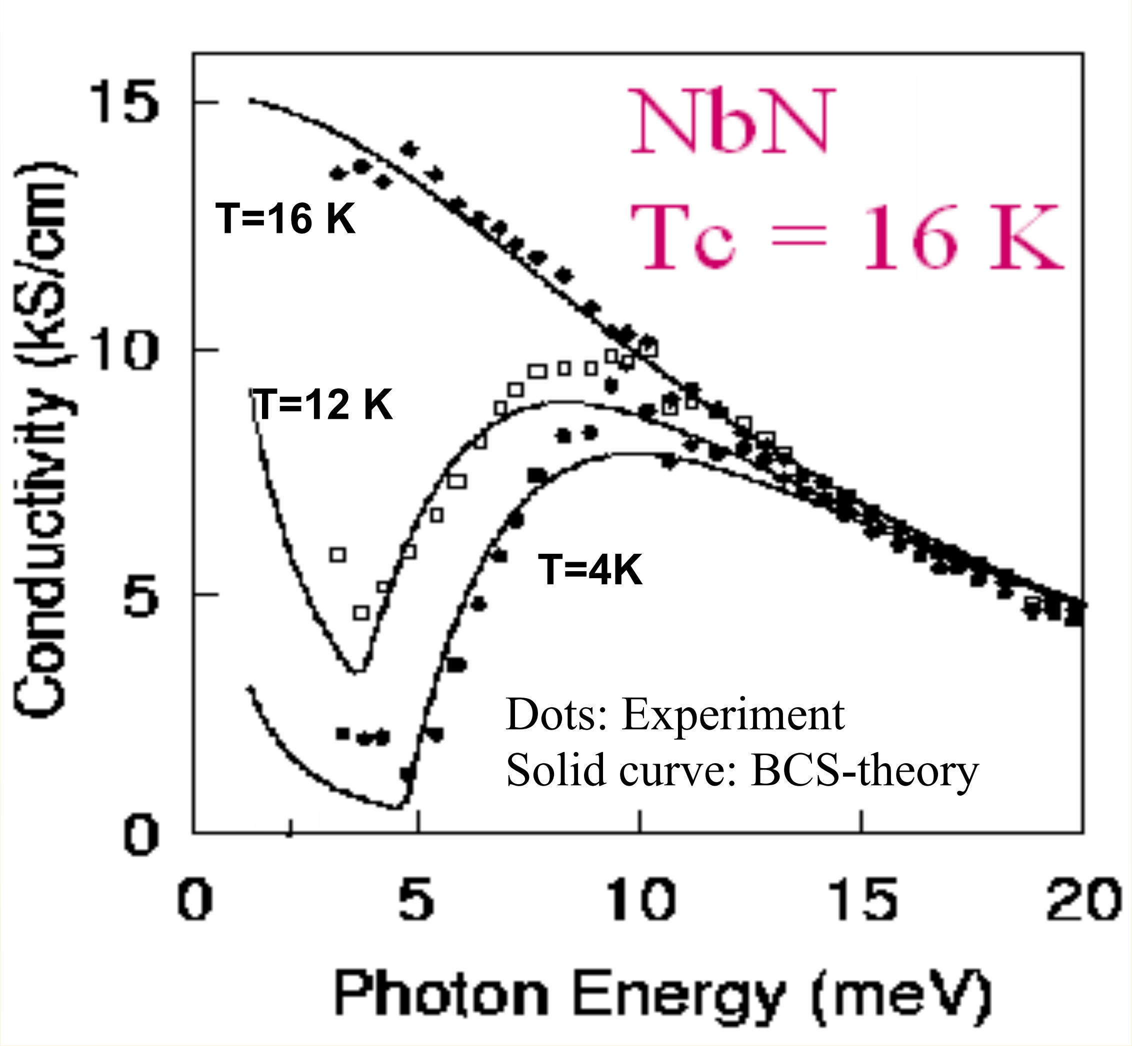
Materials with novel electronic properties, an optical perspective.
Part III
Superconductors – Interactions -Plasmons

Superconducting gap in the single particle density of states



Tunneling spectroscopy of MgB_2 (T_c = 39 K)
H.Schmidt et al (2001)

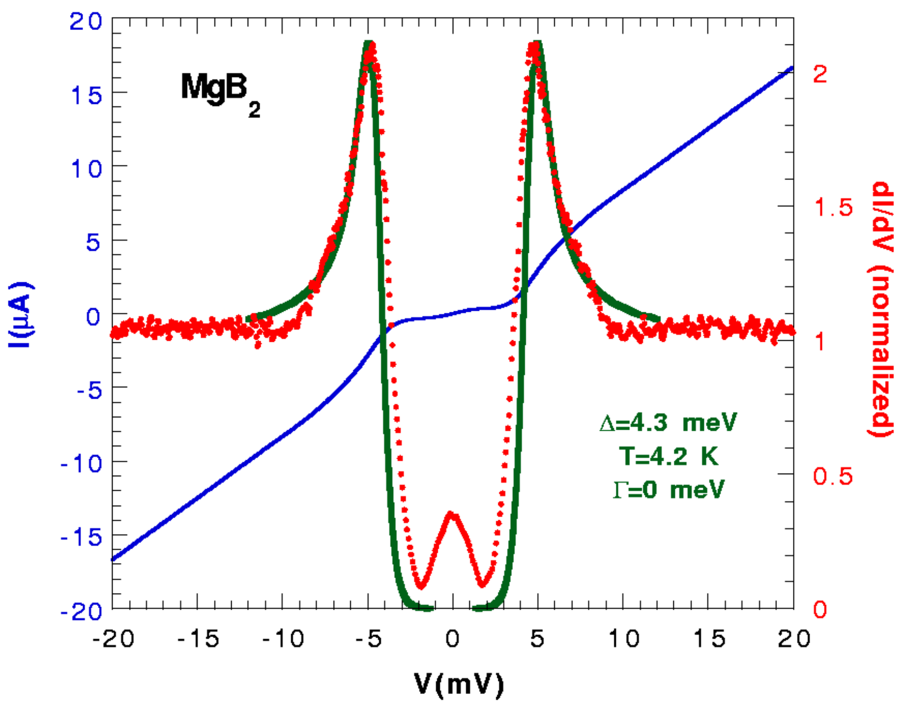
Superconducting gap in the optical conductivity
$2\Delta=3.52\ k_BT_c$

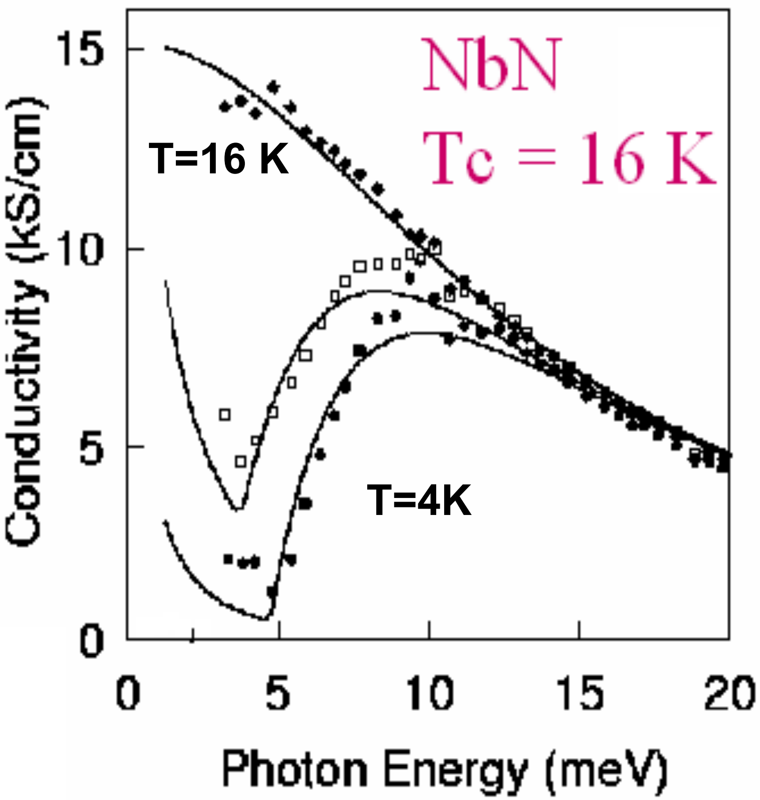


Excitation energies > 2Δ: individual electrons and holes.

However, the *supercurrent* is carried by *paired* electrons !

Are collective excitations of the *pairs* possible below 2Δ ,
- a bit like excitons in the gap of an insulator ?





P. W. Anderson, *Phys. Rev.* **112**, 1900 (1958)

Bad metal (e.g. amorphous bismuth):

No plasmon in the normal state:

$\omega_{p,L} = 0$

Transverse EM-waves: $\omega_{pT} = pv$

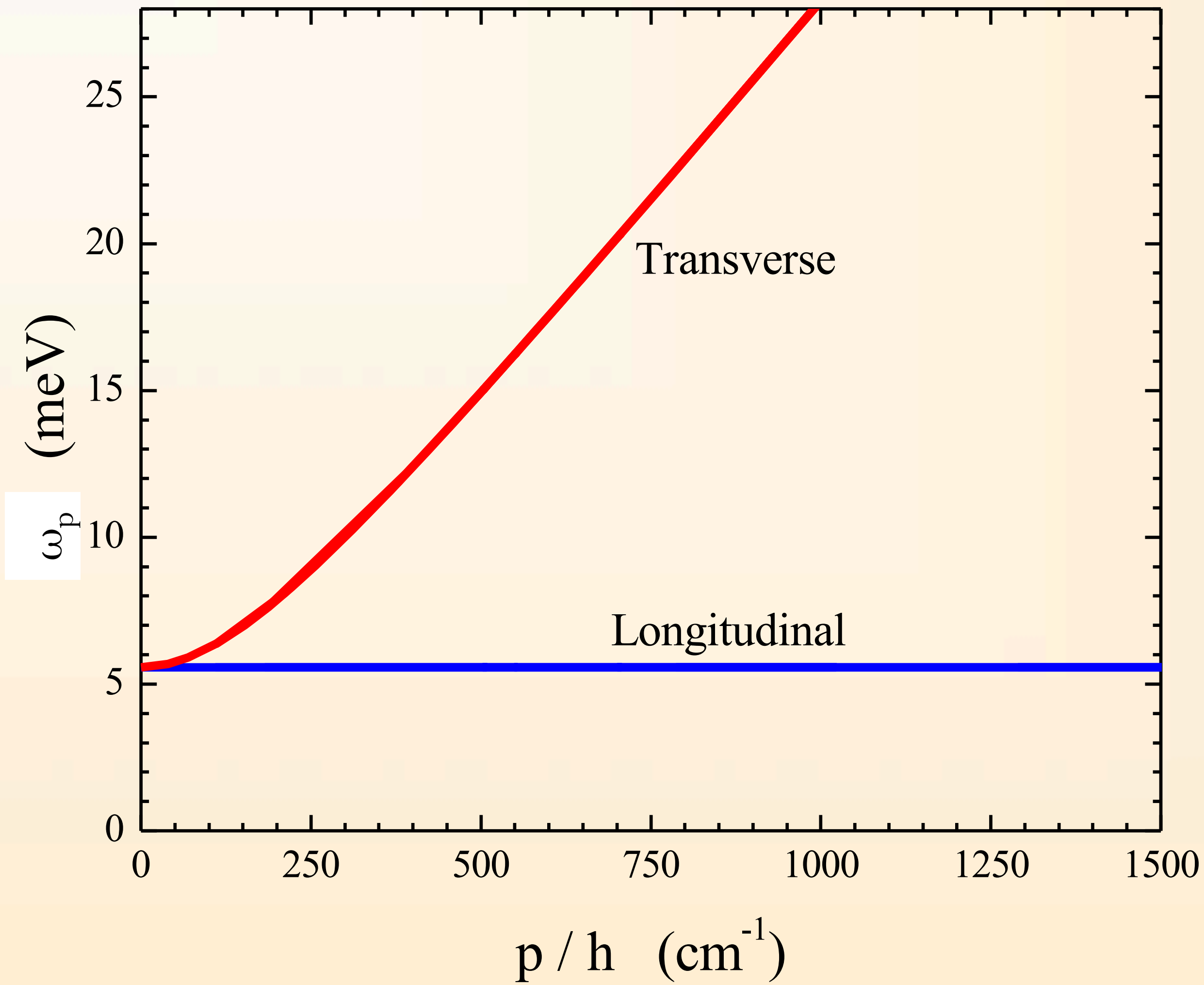
Plasmon in the superconducting state

(superfluid density = $\omega_s^{\,2}$)

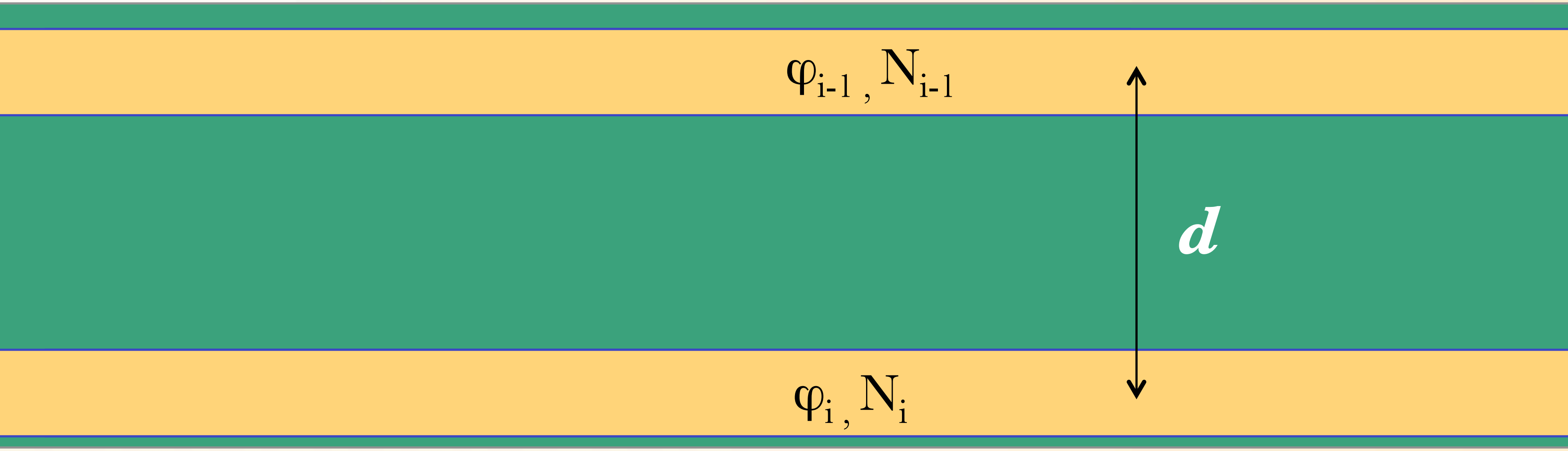
Plasmon: $\omega_{p,s} = c\, \omega_s$

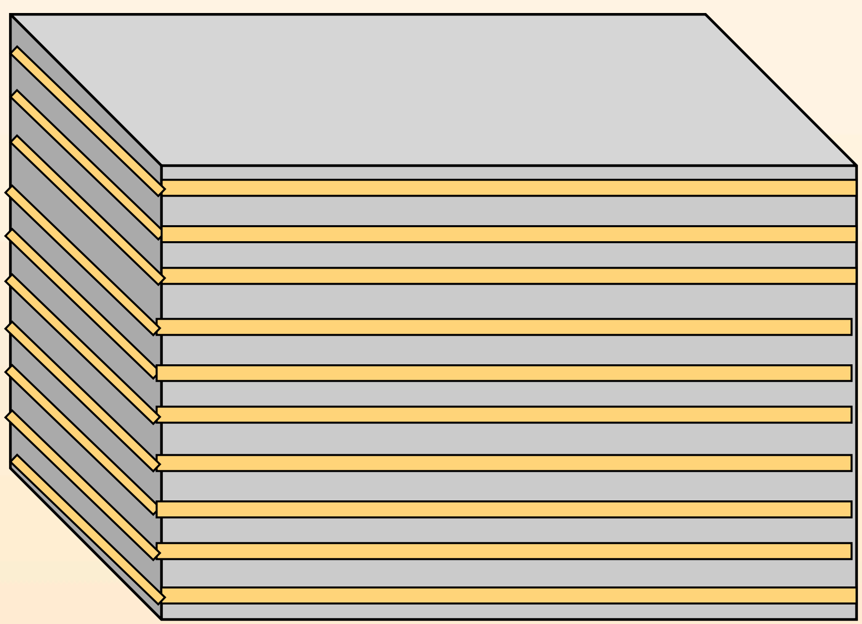
Transverse EM: $\omega_{p,T} = (\omega_{p,s}^{\,2} + p^2 v^2)^{1/2}$

Plasmons (longitudinal)
&
Plasma-polaritons (transverse)



High T_c cuprates





Josephson Plasmon

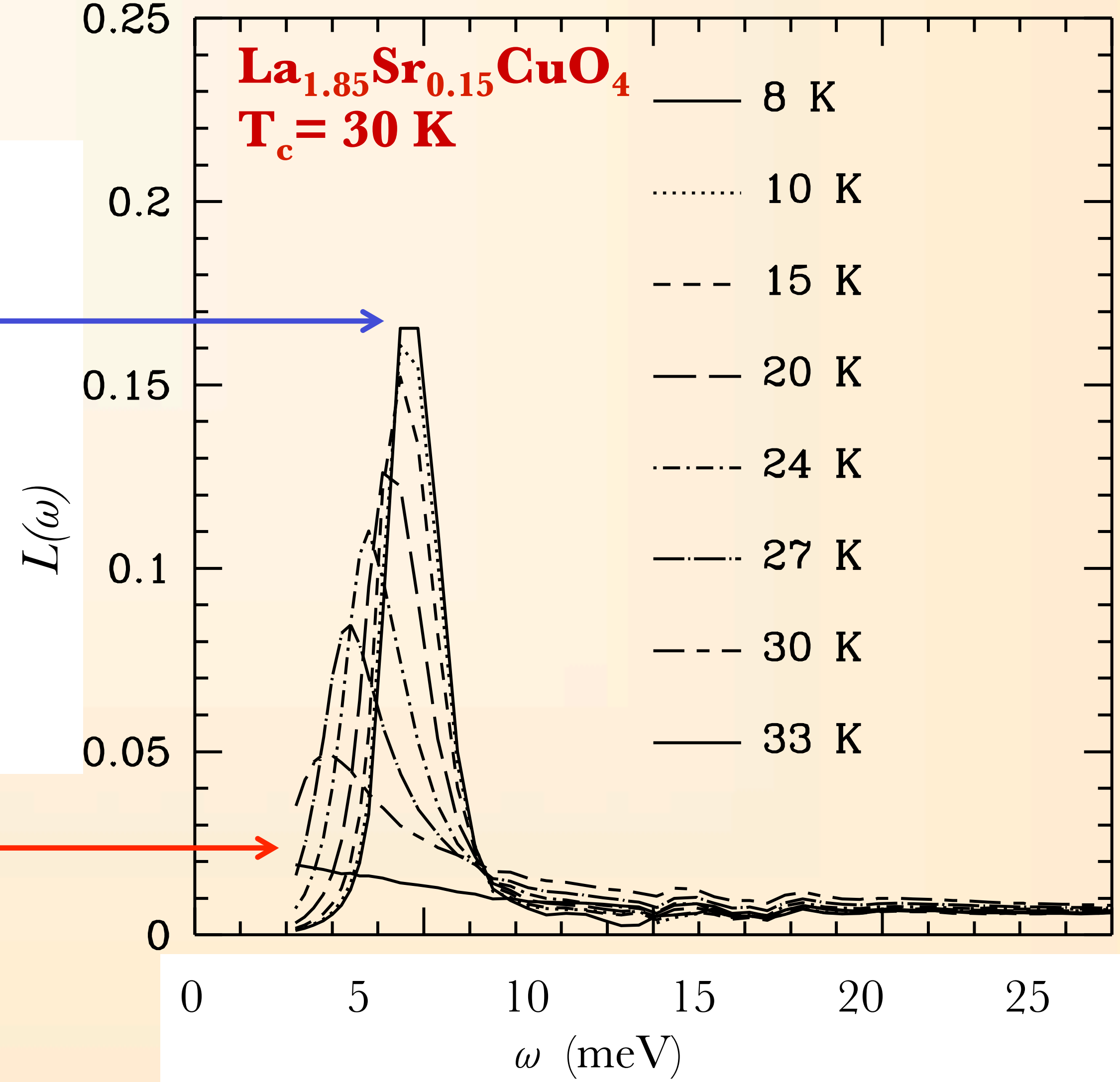
$$\omega_{p,s}{}^2 = 4\pi\, d\, e^2\, E_J$$

$$\text{Superconductor: } \sigma(\omega) = \frac{i\omega_{p,s}^2 + \gamma^2}{4\pi\omega} \Rightarrow \text{Loss function: } L(\omega) \equiv \mathrm{Re}\frac{\omega}{4\pi\sigma(\omega) - i\omega} = \frac{\gamma^2\omega^2}{\left[\omega_{p,s}^2 - \omega^2\right]^2 + \gamma^4}$$

$T < T_c$:

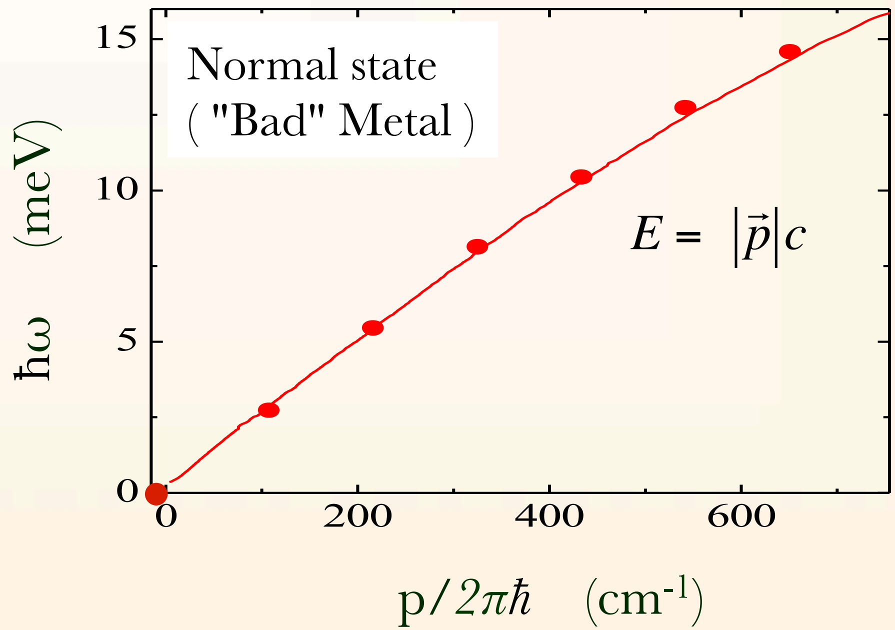
Resonance at $\omega = \omega_{p,s}$

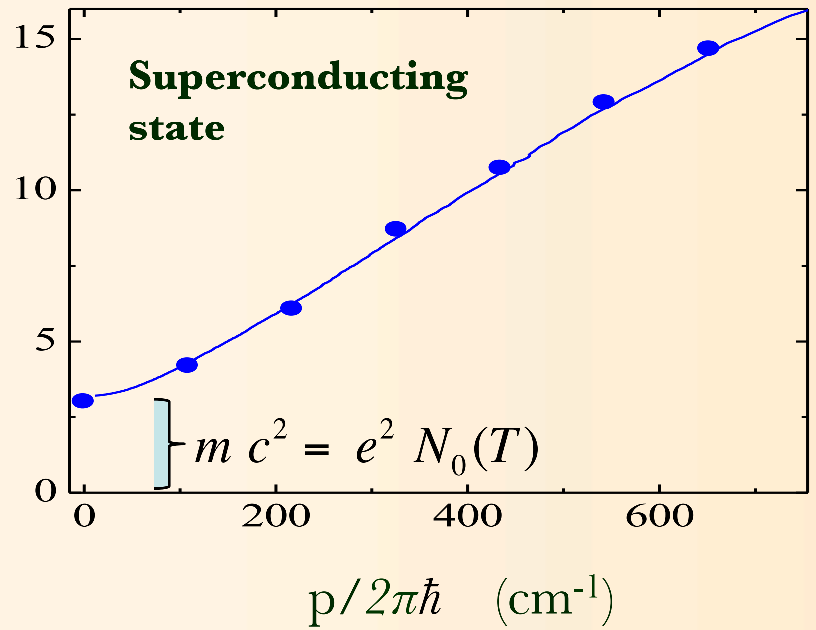
$T > T_c$: No plasmon



J. H. Kim *et al.*, *Physica C* **247**, 297 (1995).

The Higgs Mechanism in a Superconductor



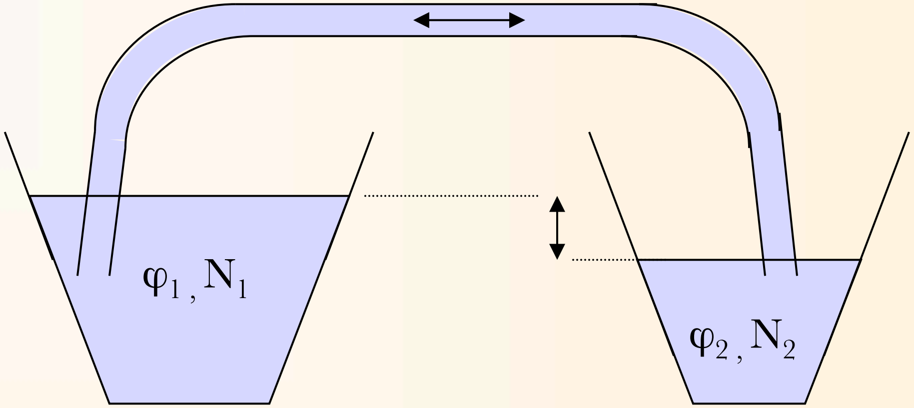


Coupling of *Δ* and *A* : *Photon mass in $La_{2-x}Sr_xCuO_4$ < 6 meV*

DvdM, *J. of Superconductivity: Incorporating Novel Magnetism*, **559**, 17 (2004) .

Leggett excitons in two-band superconductors

A.J. Leggett, Progr Theor. Phys. 36, 901 (1966)



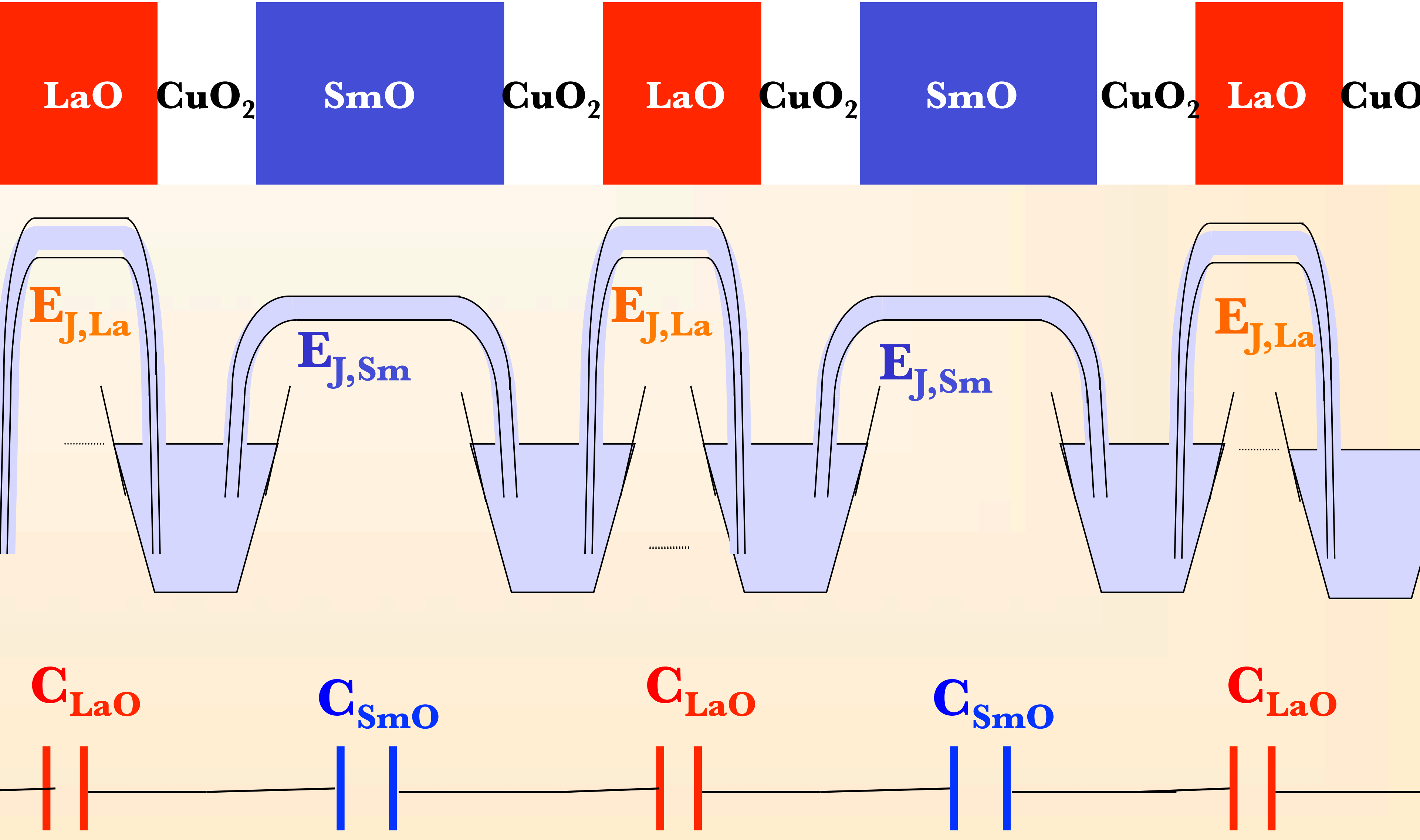
Josephson coupling: E_J

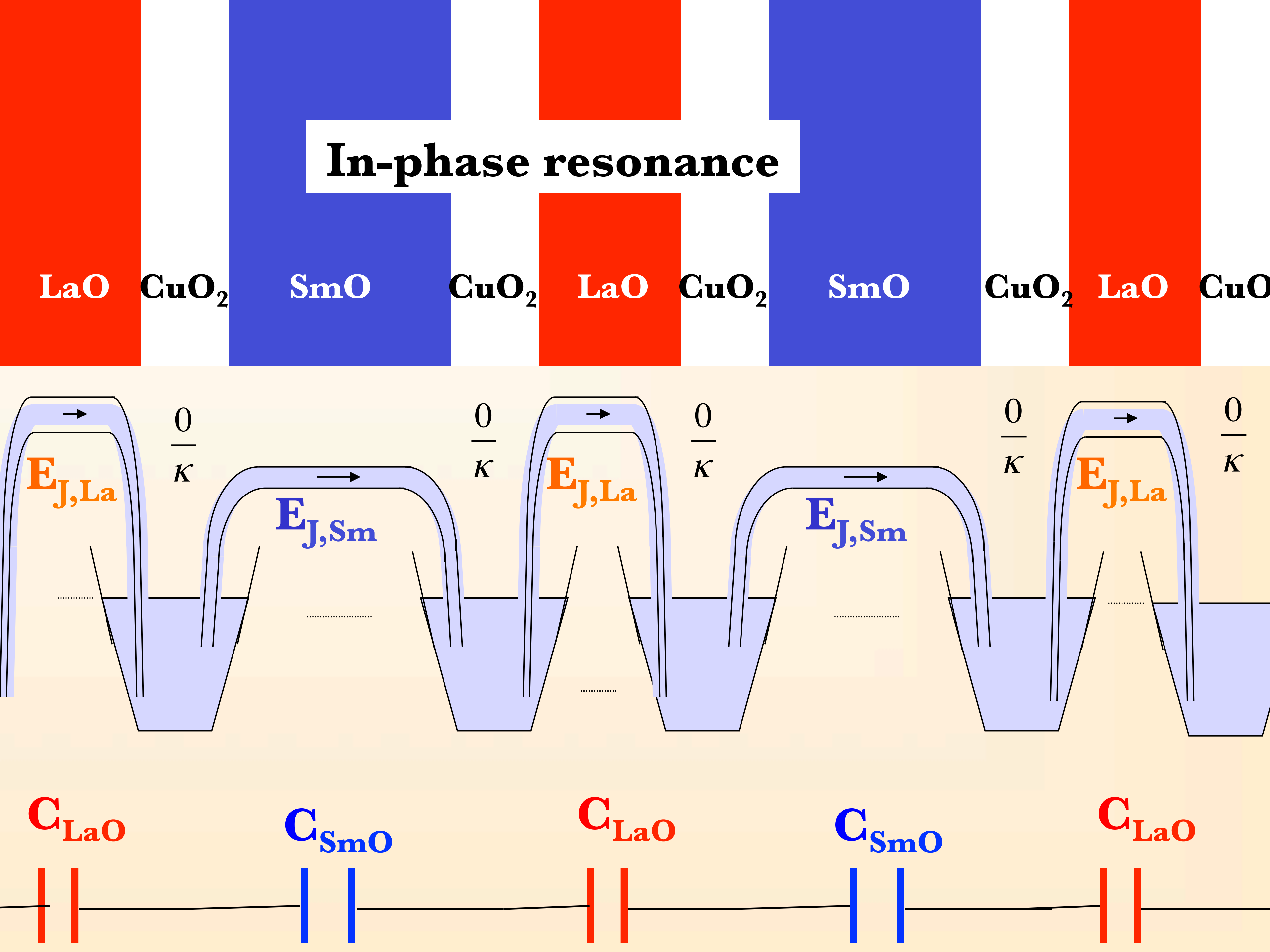
Restoring force = incompressibility: $1/\kappa = 1/N(0)$

→ Collective mode: $\hbar\omega_L = (E_J / N(0))^{0.5}$

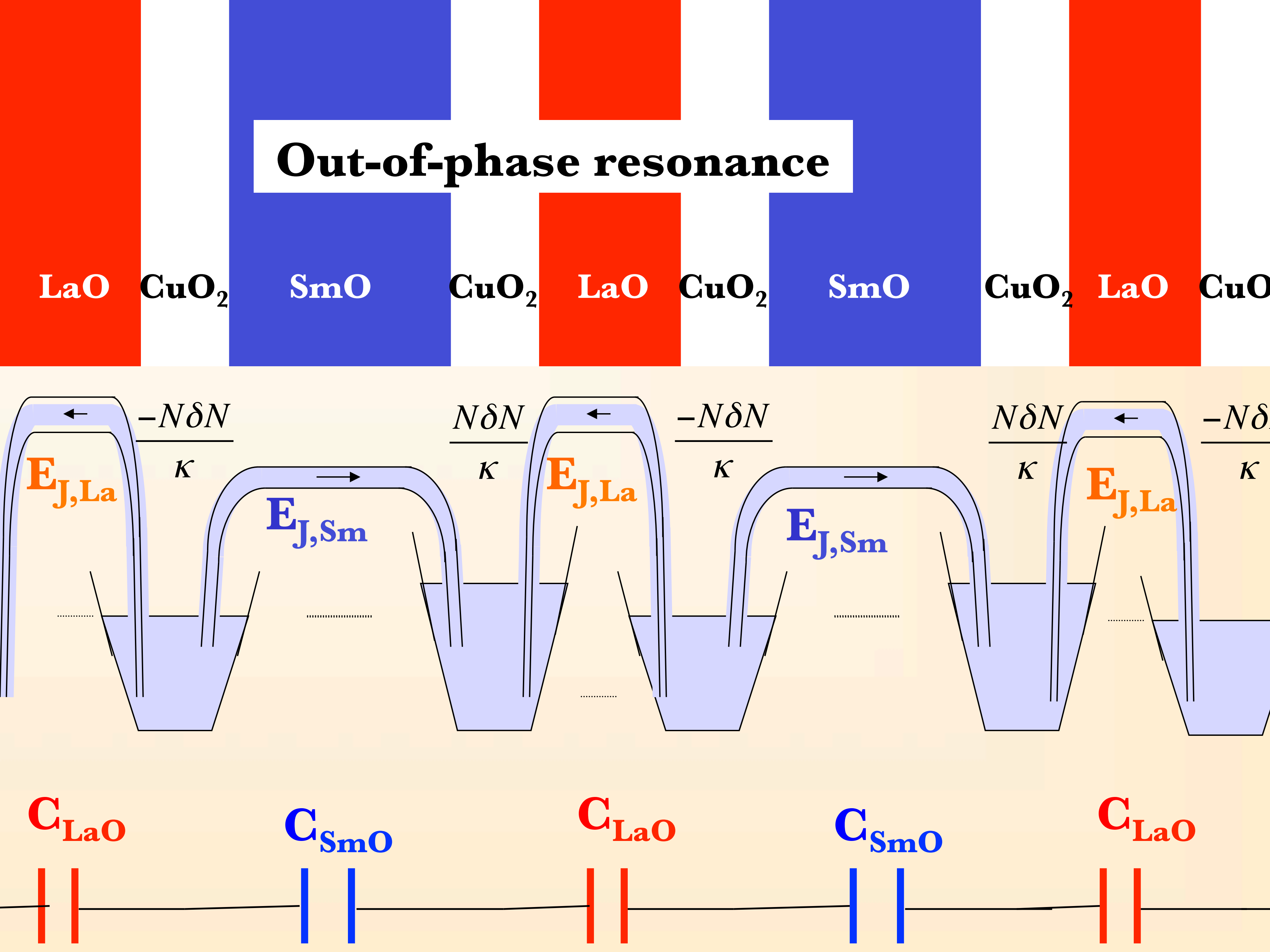
Layered superconductors have two restoring forces: Capacitance + Incompressibility

D. van der Marel and A. A. Tsvetkov, Phys. Rev. B 64 (2001) 024530.





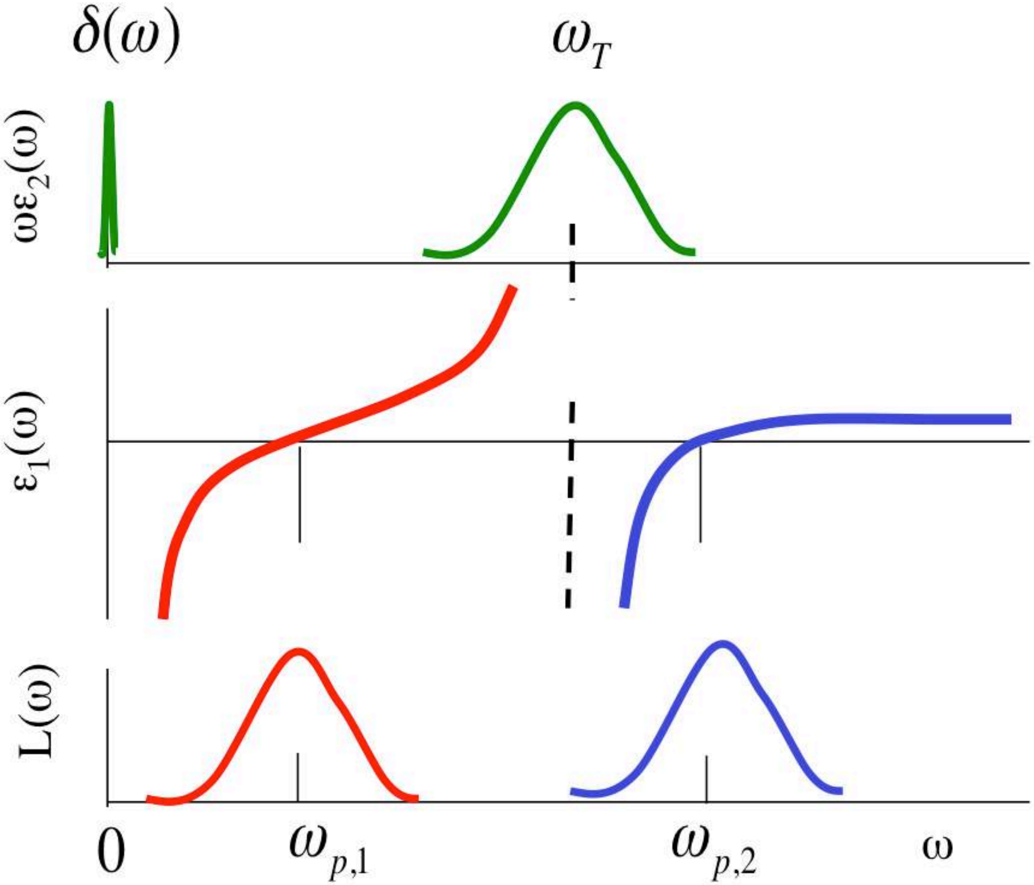
In-phase resonance
LaO
CuO2
SmO
CuO2
LaO
CuO2
SmO
CuO2
LaO
CuO
EJ,La
0
κ
EJ,Sm
0
κ
EJ,La
0
κ
EJ,Sm
0
κ
EJ,La
0
κ
CLaO
CSmO
CLaO
CSmO
CLaO

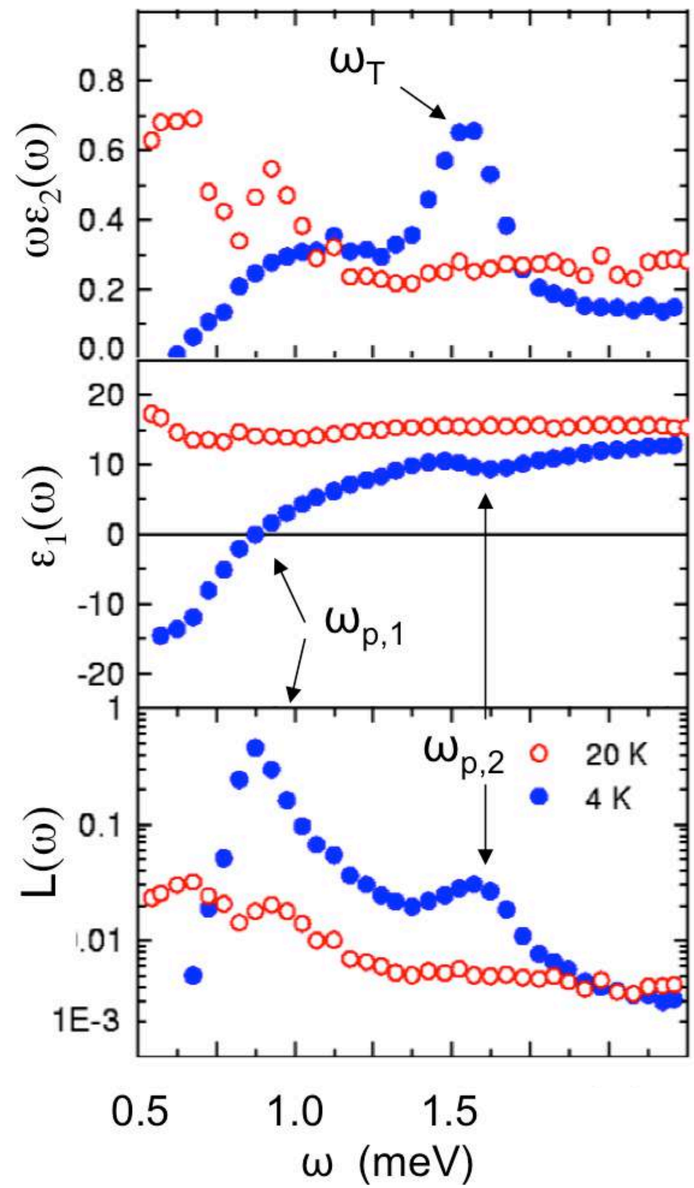


Out-of-phase resonance
LaO
CuO$_2$
SmO
CuO$_2$
LaO
CuO$_2$
SmO
CuO$_2$
LaO
$-N\delta N / \kappa$
$N\delta N / \kappa$
$-N\delta N / \kappa$
$N\delta N / \kappa$
$E_{J,La}$
$E_{J,Sm}$
$E_{J,La}$
$E_{J,Sm}$
$E_{J,La}$
C_{LaO}
C_{SmO}
C_{LaO}
C_{SmO}
C_{LaO}

Transverse optical plasmons & Leggett excitons

DvdM & A. Tsvetkov, PRB 64, 024530 (2001)



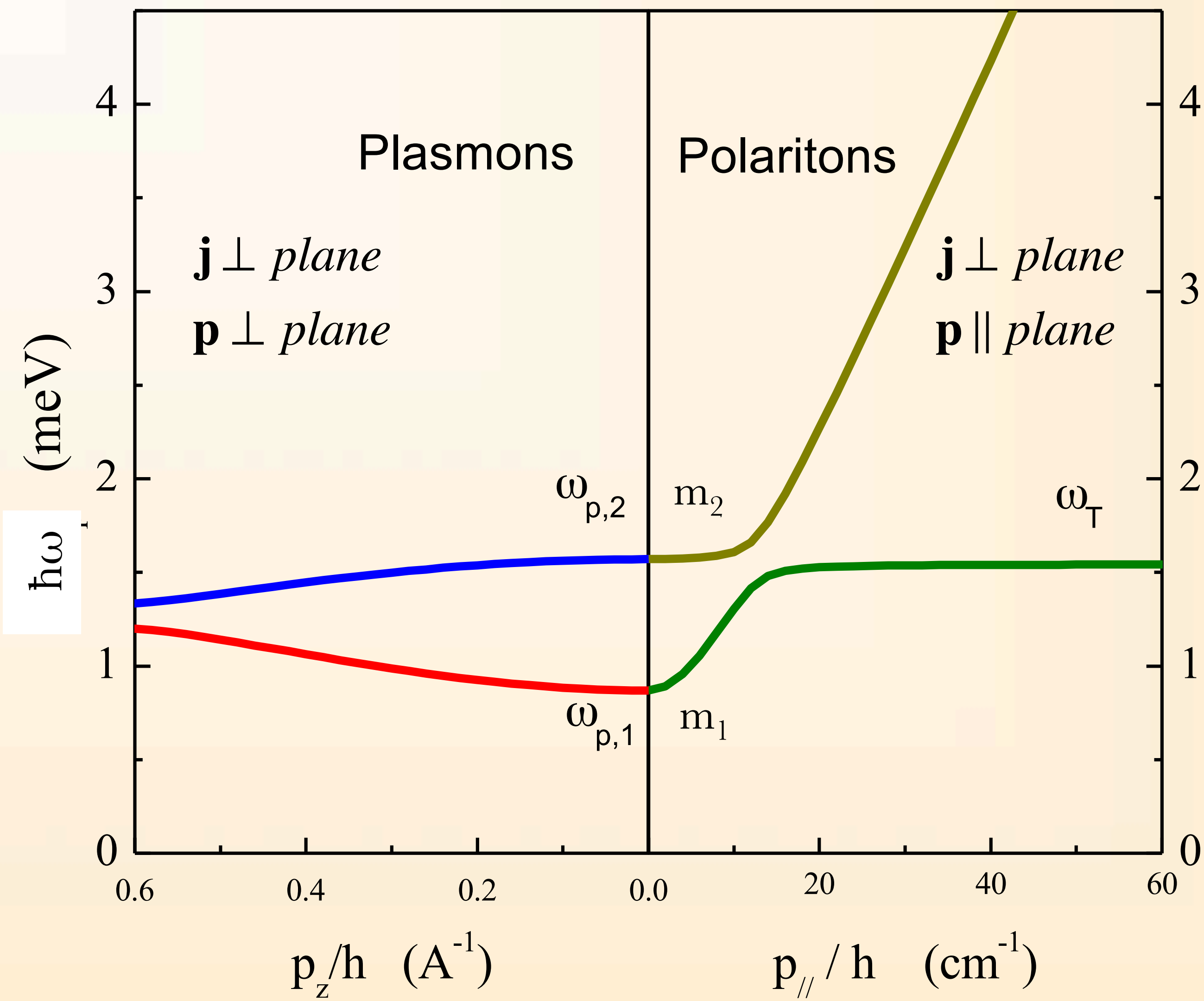


$$\varepsilon(\omega) = \varepsilon_s \frac{\left(\omega^2 - \omega_{p,1}^2\right)\left(\omega^2 - \omega_{p,2}^2\right)}{\omega^2\left(\omega^2 - \omega_T^2 + i0^+\right)}; \quad \omega_{p,1}^2 < \omega_T^2 < \omega_{p,2}^2$$

T. Kakeshita *et al.*, *PRL* **86**, 4140; H. Shibata *et al.*, *PRL* **86**, 21225; D.Dulic *et al.* *PRL* **86**, 4144 (2001)

Transverse optical plasmons & Leggett excitons

DvdM & A. Tsvetkov, PRB 64, 024530 (2001)



Leggett excitons in two-band superconductors

Symmetry breaking

BCS: $\Delta_1 \neq 0$: $\Delta_2 \neq 0$

2 Higgs bosons & 1 Leggett exciton

Coupling of Δ_1, Δ_2 and A :

Two photon flavors in $SmLaCuO_4$:

$$m_1 c^2 = 0.8\ meV$$

$$m_2 c^2 = 1.6\ meV$$

Summary part III

Superconductors – Interactions - Plasmons

Several kinds of multi-particle excitations exist inside the superconducting gap !

Plasmons, Leggett-excitons, Higgs particles, polaritons, many of which have been detected.

Some of these had –and continue to have- deep implications for the basic fabric of a superconductor.

This type of physics has also pointed –and continues to point- the way for high energy physics; the Higgs mechanism is a famous example building further on ideas born in the realms superconductivity.