Optical spectroscopy of complex electronic materials (I)

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Introduction about optical spectroscopy
(Optical constants, Kramers-Kronig transformation, intra- and interband transitions, Drude model of simple metal, Spectroscopy of correlated electrons, superconducting condensate…)

Some applications (CDW, high Tc superconductors, heavy fermions, 3D massless Dirac fermions)

Pump-probe and time domain THz spectroscopy
Some basics about optical spectroscopy technique

Emerging Experimental Techniques and Opportunities

- **Angle-Resolved Photoemission Spectroscopy (ARPES).** Wavelike quantum states of the electrons are defined in momentum space (k-space). ARPES allows direct determination of the complete momentum-space electronic structure, A(k, E), with remarkable energy and momentum resolution.

- **Spectroscopic Imaging-Scanning Tunneling Microscopy (SI-STM).** This is the complementary technique to ARPES that allows mapping of the energy-resolved quantum states in real space (r-space) with atomic resolution and yet over large sample areas.

- **Microwave/terahertz/infrared/optical spectroscopies.** These probe the electronic excitations and charge dynamics in both the frequency and time domains. This information is the key to understanding the dynamical interactions of the electrons.

- **Resonant elastic and inelastic x-ray spectroscopy.** Resonant elastic and inelastic x-ray scattering can now reveal spin and charge density waves and superlattices with tiny modulation amplitudes. This information is critically important for understanding spatially periodic electronic states of matter.

- **Neutron Scattering (NS).** High-intensity NS — for example, from the Spallation Neutron Source — will allow precision measurements of both magnetic ground states and the complete spectrum of magnetic excitations in high-temperature and exotic superconductors.

- **NMR/NQR/µSR.** NMR measures spin dynamics, NQR measures the charge heterogeneity and dynamics, and µSR measures nanoscale variation in local magnetic field strength. These are essentially local spin/charge probes, but without imaging capabilities.
Optical spectroscopy of solids

D. N. Basov, Richard D. Averitt, Dirk van der Marel, Martin Dressel, Kristjan Haule
Rev. Mod. Phys., 2011

Units: 1 eV = 8065 cm\(^{-1}\) = 11400 K
1 THz = 33 cm\(^{-1}\) ~ 4 meV
The electrodynamical properties of solids can be described by a number of so-called “optical constants”: complex refractive index, or complex dielectric constants, or complex conductivity. The charge excitations of an electronic system is directly reflected in those “constants”.

Those optical constants could be probed either directly (ellipsometry, ultrfast laser-based time domain terahertz spectroscopy,...) or indirectly (reflectance measurement over broad frequencies).
Optical constants

Consider an electromagnetic wave in a medium

$$E_y = E_0 e^{i(qx - \omega t)} = E_0 e^{i\omega(x/v-t)} = E_0 e^{i\omega \frac{nx}{c} - t}$$

where \( v \equiv \omega/q = c/n(\omega), \) \( n(\omega) \) : refractive index

If there exists absorption,

\[ E_y = E_0 e^{i\omega \frac{nx}{c} - t - \omega Kx/c} \]

K: attenuation factor

Intensity

\[ I \propto E_y^2 = E_0^2 e^{-\frac{2\omega Kx}{c}} \]

Introducing a complex refractive index:

\[ N(\omega) \equiv n(\omega) + iK(\omega) \]

\[ E_y = E_0 e^{i\omega \frac{N(\omega)x}{c} - t} \]
Reflectivity

\[
\frac{E_{\text{ref}}}{E_{\text{in}}} \equiv r = r(\omega)e^{i\theta(\omega)}
\]

\[
= \frac{n + iK - 1}{n + iK + 1} = \sqrt{(n-1)^2 + K^2} \frac{e^{i\theta(\omega)}}{(n+1)^2 + K^2}
\]

\[
R = |E_{\text{ref}} / E_{\text{in}}|^2 = |r(\omega)|^2 = \frac{(n-1)^2 + K^2}{(n+1)^2 + K^2}
\]

\[
tg \theta = \frac{2K}{n^2 + K^2 - 1}
\]

If \(n, K\) are known, we can get \(R, \theta\); vice versa.
Dielectric function

\[ D(q, \omega) \equiv \varepsilon(q, \omega)E(q, \omega) \]

photon, \( q \to 0, \varepsilon = \varepsilon(\omega, q \to 0) = \varepsilon(\omega) \)

\[ \therefore \sqrt{\varepsilon(\omega)} = N(\omega) \]

\[ \Rightarrow \varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega) = (n(\omega) + iK(\omega))^2 \]

\[ \varepsilon_1(\omega) = n^2(\omega) - K^2(\omega) \]
\[ \varepsilon_2(\omega) = 2n(\omega) \cdot K(\omega) \]

or

\[ n = \frac{1}{\sqrt{2}} \sqrt{\frac{1}{\varepsilon_1(\omega)} + \frac{1}{\varepsilon_2(\omega)} + \frac{1}{\varepsilon_1(\omega)}} \]
\[ k = \frac{1}{\sqrt{2}} \sqrt{\frac{1}{\varepsilon_1(\omega)} + \frac{1}{\varepsilon_2(\omega)} - \frac{1}{\varepsilon_1(\omega)}} \]

\( 0 \quad \pi/a \sim 1 \text{Å}^{-1} \)

Infrared
\( q = 2\pi/\lambda \sim 10^{-4} \text{Å}^{-1} \)
conductivity

\[ \sigma = \sigma_1(\omega) + \sigma_2(\omega) \]

By electrodynamics, \( \varepsilon(\omega) = 1 + \frac{4\pi i \sigma(\omega)}{\omega} \)

In a solid, considering the contribution from ions or from high energy electronic excitations

\[ \varepsilon(\omega) = \varepsilon_\infty + \frac{4\pi i \sigma(\omega)}{\omega} \]
Now, we have several pairs of optical constants:

\[
\begin{align*}
\{ & n(\omega), K(\omega) \\
& R(\omega), \theta(\omega) \\
& \varepsilon_1(\omega), \varepsilon_2(\omega) \\
& \sigma_1(\omega), \sigma_2(\omega) \}
\end{align*}
\]

Usually, \( R(\omega) \) can be obtained from measurement.
**Kramers-Kronig relation**

-- the relation between the real and imaginary parts of a response function.

For optical reflectance

\[ r(\omega) = \sqrt{R(\omega)}e^{i\theta} \]
\[ \Rightarrow \ln r(\omega) = (1/2) \ln R(\omega) + i\theta \]

- **Low-\(\omega\) extrapolations:**
  - Insulator: \(R\)~ constant
  - Metal: Hagen-Rubens
  - Superconductor: two-fluids model

- **High-\(\omega\) extrapolations:**
  - \(R\)~ \(\omega^{-p}\) (\(p \approx 0.5-1\), for intermediate region)
  - \(R\)~ \(\omega^{-n}\) (\(n = 4\), above interband transition)
Reflectivity measurement
From $R(\omega)$ to $\sigma_1(\omega)$

$R(\omega) \Rightarrow \theta(\omega) \Rightarrow \begin{cases} n(\omega), \kappa(\omega) \\ \varepsilon_1(\omega), \varepsilon_2(\omega) \\ \sigma_1(\omega), \sigma_2(\omega) \end{cases}$

Si-beamsplitter for 113v

Bruker 113, 66v, 80v, and grating spectrometers

Energy range: 17 -50000 cm\(^{-1}\) (2 meV~6 eV)
In-situ overcoating technique

FT-IR spectrometer

In situ evaporation

\[
\left( \frac{R_g}{R_r} \right) \left( \frac{R_{gs}}{R_r} \right)^{-1} \equiv \frac{R_g}{R_{gs}}
\]

C. C. Homes et al.
Applied Optics 32,2976(1993)
Infrared light cannot be absorbed directly by electron-hole excitation.

(a) Impurity-assisted absorption

(b) boson-assisted absorption

Holstein process, if phonons are involved.

Kubo-Greenwood formula

\[ \epsilon_2(\omega) = \frac{8\pi^2 e^2}{m^2 \omega^2} J(\hbar\omega) \left| \tilde{p}_{vc}(\hbar\omega) \right|^2 \]

\[ \sigma_1(\omega) = \frac{1}{4\pi} \omega \epsilon_2(\omega) \]
Drude model

--free carrier response

\[ \sigma(\omega) = \frac{\sigma_0}{1 - i\omega\tau} \]

\[ \sigma_0 = \frac{ne^2\tau}{m^*} = \frac{\omega_p^2\tau}{4\pi} \]

\[ \sigma_1(\omega) = \frac{\sigma_0}{1 + \omega^2\tau^2} = \frac{\omega_p^2\tau}{4\pi} \frac{1}{1 + \omega^2\tau^2} \]

\[ \sigma_2(\omega) = \frac{\sigma_0\omega\tau}{1 + \omega^2\tau^2} = \frac{\omega_p^2\tau}{4\pi} \frac{\omega\tau}{1 + \omega^2\tau^2} \]

Paul Drude (1863-1906)
Schematic picture for Drude response

\[
\sigma(\omega) = \frac{\sigma_0}{1 - i\omega\tau} = \frac{\omega_D^2}{4\pi} \frac{1}{1/\tau - i\omega}
\]

\[
\varepsilon(\omega) = \varepsilon_\infty + \frac{4\pi i}{\omega} \sigma(\omega)
\]

\[
\Rightarrow \varepsilon_1 = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + 1/\tau^2}
\]

\[
\text{Im}\left\{ \frac{-1}{\varepsilon(\omega)} \right\} = \frac{\omega_p^2 \omega / \tau}{(\omega^2 - \omega_p^2)^2 + \omega^2 \tau^2}
\]

\[
\omega_p' = \omega_p / \sqrt{\varepsilon_\infty}
\]

\[
\sigma_1(\omega) = \frac{\omega_D^2}{4\pi} \frac{1/\tau}{(1/\tau)^2 + \omega^2}
\]

\[
\int_0^\infty \sigma_1(\omega) d\omega = \frac{\omega_p^2}{8}
\]
Phenomenological Drude-Lorentz model

\[ \sigma_1(\omega) = \sum_i \frac{\omega_{p_i}^2 / 4\pi}{\gamma_i^2 + \omega^2} + \sum_j \frac{(S_j^2 / 4\pi) \gamma_j \omega^2}{(\omega_{0j}^2 - \omega^2)^2 + \gamma_j^2 \omega^2} \]

Drude model:
free electron gas, \( \tau = \text{const.} \)

Lorentz model:
localized electron
CuxTiSe2 system
Parent compound 1T-TiSe$_2$

- 1T-TiSe$_2$ was one of the first CDW-bearing materials
- Broken symmetry at 200 K with a 2x2x2 superlattice
Band structure and lattice instability of TiSe$_2$

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Ti: 3d$^2$4s$^2$
Se: 4s$^2$4p$^4$

Ti: 3d band
Se: 4p band

FIG. 1. Energy-band structure of TiSe$_2$ in the local exchange and correlation model.
Electron-Hole Coupling and the Charge Density Wave Transition in TiSe$_2$

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No band crossing

Fermi level below CDW.

It is insulating!!
Photoemission of bands above the Fermi level: The excitonic insulator phase transition in $1T$-TiSe$_2$


But does not satisfy the charge neutrality! X
Issue

- ARPES experiments did not resolve conclusively whether the compound is a semimetal or semiconductor with a small indirect gap.

- The mechanism of the CDW transition: not due to the Fermi Surface nesting
**TiSe$_2$ single crystal**

G. Li et al., PRL (07a)
$\omega_p' = \omega_p / \sqrt{\varepsilon_\infty}$
Drude  CDW gap

Free carriers with very long relaxation time exist in the CDW gapped state

FS is not fully gapped??
\[ \epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega/\tau} + \sum_{i=1}^{2} \frac{S_i^2}{\omega_i^2 - \omega^2 - i\omega/\tau_i}. \]  

(1)

It contains a Drude term and two Lorentz terms, which approximately capture the contributions by free carriers and interband transitions. As shown in the inset of
1T-TiSe2 is a semimetal with very low carrier density at all T.

- Carrier density changes with T, decreases from room T to 150 K then increases slightly with further decreasing T.

- Development of an energy gap ~0.15 eV below 200 K.

- Dramatic different carrier damping at different T.
The electron-hole coupling acts to mix the electron band and hole band that are connected by a particular wave vector.
Exciton-driven CDW

(b) Normal Phase

(c) CDW Phase

$\omega_p^2 = 4\pi e^2 \left( \frac{n_h}{m_h} + \frac{n_e}{m_e} \right)$

$R_H = \frac{1}{e} \frac{n_h \mu_h^2 - n_e \mu_e^2}{(n_h \mu_h + n_e \mu_e)^2}$

$\sigma = e(n_h \mu_h + n_e \mu_e)$

G. Li et al., PRL (07a)
FIG. 1. Energy-band structure of TiSe$_2$ in the local exchange and correlation model.
Cu-doped Cu$_x$TiSe$_2$
Emergence of Fermi Pockets in a New Excitonic Charge-Density-Wave Melted Superconductor

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[Graphical representation of the Fermi pockets and band structure]

Single band metal
Plasma frequency increases with decreasing T!!
Rarely seen phenomenon!!
\[
\epsilon(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\omega/\tau} + \sum_{i=1}^{2} \frac{S_i^2}{\omega_i^2 - \omega^2 - i\omega/\tau_i}.
\] (1)

It contains a Drude term and two Lorentz terms, which approximately capture the contributions by free carriers and interband transitions. As shown in the inset of
Plasma frequency increases with decreasing $T$??

\[ \omega_p^2 = \frac{4\pi ne^2}{m^*} \]

In terms of two bands:

\[ \omega_p^2 = 4\pi e^2 \left( \frac{n_e}{m_e^*} + \frac{n_h}{m_h^*} \right) \]

• $n$ increases with decreasing $T$??

• $m^*$ decreases with decreasing $T$, undressing effect??
Hall coefficient is almost T-independent for superconducting sample!!
Possible shift of chemical potential with $T$

$$\mu = \mu_0[1 - \alpha (T/T_F)^2],$$

$E_F \sim 80-100 \text{ meV at } 300 \text{ K}$

heavy mass at L point due to very strong electron-phonon scattering, (linked with lattice instability in parent compound).

smaller mass at Fermi crossing point due to reduced scattering (away from L point).

$\Sigma(\varepsilon_k)$ changes along dispersive band!

G. Li et al., PRL (07b)
Recently, we found similar phenomena in a number of low-carrier density metals, e.g. LaSb (semimetal):
From simple metal to correlated systems

Simple metal

High-\(T_c\) cuprates

![Graph of R vs \(\omega\) for gold](image)

![Graph of R vs \(\omega\) for YBCO and Bi2212](image)
Correlation effect in optical conductivity

Simple metal

\[ \omega_p^2 = \frac{4\pi Ne^2}{m_b} \]

Correlation effect: reducing the kinetic energy of electrons, or Drude spectral weight.

Correlated metal

Correlation effect:

\[ \omega_p^{\text{Drude}} = \frac{4\pi Ne^2}{m^*} \]

\[ \omega_p^{\text{MIR}2} = \frac{4\pi Ne^2}{m_b} \]

\[ m^* / m_b = \frac{\omega_p^{\text{Tot}2}}{\omega_p^{\text{Drude}2}} \]


\[ K_{\text{exp}} / K_{\text{band}} = \frac{\int_0^{\omega_{\text{opt}}} \sigma_1(\omega) d\omega}{\int_0^{\omega_{\text{band}}} \sigma_1(\omega) d\omega} \]

\[ K_{\text{exp}} / K_{\text{band}} = \frac{\omega_p^2}{\omega_p^2 + (\omega_p^{\text{MIR}})^2} \]
Hubbard U physics: $\rho(\omega)$

\[ \rho(\omega) \equiv -\frac{1}{\pi} \sum_k \text{Im} G(k, \omega + i0^+) \]

FIG. 30. Local spectral density $\pi D \rho(\omega)$ at $T=0$, for several values of $U$, obtained by the iterated perturbation theory approximation. The first four curves (from top to bottom, $U/D = 1, 2, 2.5, 3$) correspond to an increasingly correlated metal, while the bottom one ($U/D = 4$) is an insulator.

served. As $T$ is lowered, there is an enhancement of the spectrum at intermediate frequencies of order 0.5 eV; more notably, a sharp low-frequency feature emerges that extends from 0 to 0.15 eV.
Sum rule

f-sum rule:

\[ \int_0^\infty \sigma_1(\omega) d\omega = \frac{\pi n e^2}{2 m_e} = \omega_p^2 / 8 \]

It has the explicit implication that at energies higher than the total bandwidth of a solid, electrons behave as free particles.

Kubo partial sum-rule:

\[ \int_0^W \sigma_1(\omega) d\omega = W_K = \frac{\pi e^2}{2N} \sum_k \nabla^2_{k_x} \epsilon_k n_k \]

The upper limit of the integration is much larger than the bandwidth of a given band crossing the Fermi level but still smaller than the energy of interband transitions. For \( \epsilon_k = k^2 / 2m_e \), the Kubo sum rule reduces to the f-sum rule.

\( W_K \) depends on \( T \) and on the state of the system because of \( n_k \)--"violation of the conductivity sum rule", first studied by Hirsch.

In reality, there is no true violation: the change of the spectral weight of a given band would be compensated by an appropriate change in the spectral weight in other bands, and the total spectral weight over all bands is conserved.
Non-Drude spectra of strongly correlated electrons

General feature: a sharp peak at $\omega=0$

+ a long tail extending to high energies

Two possible interpretations

$\sigma_1(\omega)$

Drude

MIR

$1/\tau(\omega)$
**Extended Drude Model**

**Drude Model**

\[
\sigma(\omega) = \frac{\omega_p^2}{4\pi} \frac{1}{1/\tau - i\omega}
\]

Let \( M(\omega, T) = 1/\tau(\omega, T) - i\omega\lambda(\omega, T) \)

\[
\sigma(\omega, T) = \frac{\omega_p^2}{4\pi} \frac{1}{M(\omega, T) - i\omega}
\]

\[
= \frac{\omega_p^2}{4\pi} \frac{1}{1/\tau(\omega, T) - i\omega[1 + \lambda(\omega, T)]}
\]

\[
= \frac{1}{4\pi} \frac{\omega_p^*}{1/\tau^*(\omega, T) - i\omega}
\]

\[
1/\tau(\omega, T) = (\omega_p^2 / 4\pi) \text{Re}[1/\sigma(\omega, T)]
\]

\[
m^*/m = 1 + \lambda(\omega) = (\omega_p^2 / 4\pi\omega) \text{Im}[1/\sigma(\omega, T)]
\]

**e.g. Marginal Fermi Liquid model:**

\[
M(\omega, T) = 1/\tau(\omega, T) - i\omega\lambda(\omega, T)
\]

\[
= g^2 N^2(0) \left( \frac{\pi}{2} \frac{x + i\omega \ln \frac{x}{\omega_c}}{x} \right)
\]

Where \( x = \max(|\omega|, T) \),

or \( x = (\omega^2 + \alpha(\pi T)^2)^{1/2} \)
The extended Drude model in terms of optical self-energy

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

According to Littlewood and Varma,

$$\gamma(\omega) = -2i\Sigma^{op}$$
$$= -2i[\Sigma_1(\omega) + i\Sigma_2(\omega)]$$

Optical self-energy

Relation to the $1/\tau(\omega)$ and $m^*/m$

$$\gamma_1(\omega) = 1/\tau(\omega) = 2\Sigma_2$$
$$\gamma_2(\omega) = \omega(1-m^*/m) = -2\Sigma_1$$
Hwang, Timusk, Gu,
Energy gaps for symmetry broken states: Coherence factors

Coherent factors play an extremely important role in determining the energy gap structures of broken symmetry states.

\[ \alpha_s = \int |M|^2 F(\Delta, E, E + \hbar \omega)N_s(E)N_s(E + \hbar \omega)[f(E) - f(E + \hbar \omega)]dE \]

From the textbook of Tinkham

\[ F(\Delta, E, E') = \frac{1}{2}(1 \pm \frac{\Delta^2}{EE'}) \]

Superconductor vs density wave state

\[ \frac{\sigma_1^S}{\sigma_1^N} = \frac{1}{\hbar \omega} \int_{-\infty}^{\infty} \frac{|E(E + \hbar \omega) \mp \Delta^2|}{(E^2 - \Delta^2)^{1/2}[(E + \hbar \omega)^2 - \Delta^2]^{1/2}}[f(E) - f(E + \hbar \omega)]dE \]

\[ 2\Delta / T_{DW} \sim 3.5 \text{ under the weak coupling BCS theory} \]
Optical spectra of a superconductor

T=0, London electrodynamics gives

\[ \sigma = \frac{1}{8} \omega_p^2 \delta(\omega) + i \omega_p^2 / 4\pi \omega \Rightarrow \frac{1}{\lambda_L^2} = \frac{8}{c^2} \int_0^\infty (\sigma_1^n - \sigma_1^s) d\omega \quad \text{or} \quad \frac{1}{\lambda_L^2} = \frac{4\pi}{c^2} \omega \sigma_2(\omega) \]

Ferrell-Glover-Tinkham sum-rule: missing area is equal to the superconducting condensate.

**dirty limit:** \( \xi > 1 \iff 2\Delta < \Gamma \)

Absorption starts at \( 2\Delta \).

**clean limit:** \( \xi < 1 \iff 2\Delta > \Gamma \)

Absorption starts at \( 2\Delta + \Omega \).

\[ \therefore \text{pippard coherence length} \quad \xi = \frac{v_F}{\pi \Delta}, \quad \Gamma = \frac{1}{\tau} = \frac{v_F}{l} \]
Kinetic energy lowering with superconducting condensate

\[ \omega_{ps}^2 = 8 \int_{0+}^{\omega_c} \left[ \sigma_1(\omega, T \approx T_c) - \sigma_1(\omega, T \ll T_c) \right] d\omega + \Delta W_K \]
Structure of energy gap

Density wave

URu2Si2

E. Fawcett et al.: Spin-density-wave antiferromagnetism in chromium alloys
Superconductors

Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$

Structure of energy gap

G. Li et al PRL 08

Cuprates, Homes data
Coherent peak below $T_c$.

O. Klein et al. PRB 94

T. Fischer et al. PRB 2010

R. V. Arguilar et al., arXiv:107.3677

THz data are different from NMR? S+- pairing?
More about extended Drude model

— Extraction the boson (phonon) spectral function from optical conductivity
Mode coupling effect in infrared spectra of high-$T_c$ cuprates

- Mode coupling effect in ARPES and tunneling
- Feature in IR spectra of high-$T_c$ superconductors

Mode coupling??

Extraction the boson (phonon) spectral function
requires Eliashberg theory:

- phonon dynamics (retardation) taken into account \( \alpha^2 F(\Omega) \)

- gap is a function of frequency \( \Delta(\omega) = F[\{\alpha^2 F(\Omega)\}, \mu^*] \)

- density of states is modified: \( \frac{dI}{dV} \propto N(\omega) = N(\epsilon_F) \text{Re} \left\{ \frac{1}{\sqrt{\omega^2 - \Delta^2(\omega)}} \right\} \)
Phonon structure in the tunneling conductance of Bi$_2$Sr$_2$CaCu$_2$O$_8$

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(Received 17 March 1995)

Clear phonon structures were observed in the tunneling conductance of a Bi$_2$Sr$_2$CaCu$_2$O$_8$-GaAs junction. The spectral function of the electron-phonon interaction gives a value of 87.3 K for the critical temperature and 22.2 meV for half a gap. There is no particularly large phonon structure, and the high $T_c$ cannot be attributed to a particular phonon mode in the electron-phonon mechanism. The gap edge structure is sharp, and an $s$-wave state is probable. However, if the angular distribution of the tunneling current is highly anisotropic we cannot definitely exclude a $d$-wave state.

Fig. 2. The normalization procedure was illustrated. The subtracted conductances at 4.2 and 88 K were plotted together with the GPDS of Bi2212 (Ref. 10). These subtracted conductances were normalized. See the text.

Fig. 4. Spectral functions obtained by Vedeneev et al. (Ref. 3), Gonnelli et al. (Ref. 4), Miyakawa et al. (Ref. 5), and the present result are compared with GDPS and the atomic vibrations (Ref. 10). O1; O in the CuO$_2$ plane, O2; apical O, O3; O in the Bi-O planes.
Tunneling spectroscopy of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$: Eliashberg analysis of the spectral dip feature

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Eliashberg strong-coupling theory, extended to a $d$-wave symmetric gap function, is used to fit quantitatively a published tunneling spectrum of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ near optimal doping. The shape, location, and strength of the high-bias spectral dip feature is adequately reproduced using a single-peak $\alpha^2F(\omega)$ centered at 36.5 meV. $\alpha^2F(\omega)$ also self-consistently determines the measured gap value $\Delta = 32$ meV. Possible origins of the bosonic spectrum that give rise to high-$T_C$ superconductivity are discussed.
Mode coupling in optical spectra?

YBCO
C C Homes
D N Basov
T. Timusk

\[ R(\omega) \Rightarrow \sigma_1(\omega) \Rightarrow 1/\tau(\omega) \]

TI-2212 thin film \( T_c = 108 \text{ K} \)

N. L. Wang et al., PRB03
In the clean limit, we must consider the boson-assisted electron-hole excitations.

(a) Impurity-assisted absorption

(b) boson-assisted absorption

Infrared light cannot be absorbed directly by electron-hole excitation.
The electron-boson (phonon) interaction

\[ 1/\tau(\omega) = \frac{2\pi}{\omega} \int_{0}^{\infty} d\Omega (\omega - \Omega) \alpha_{tr}^2(\Omega) F(\Omega) \]

T=0 K
P.B.Allen 1971

\[ \alpha^2 F(\omega) = \frac{\omega_p^2 \omega^2}{(\omega_0^2 - \omega^2)^2 + \gamma^2 \omega^2} \]

\[ \omega_0 = 500 \text{ cm}^{-1} \]
\[ \gamma = 100 \text{ cm}^{-1} \]
\[ \omega_p^2 = 50000 \text{ cm}^{-2} \]
Electron-phonon coupling at finite temperature

\[
\frac{1}{\tau(\omega, T)} = \frac{\pi}{\omega} \int_{0}^{\infty} d\Omega \alpha^2 F(\Omega, T) \left[ 2\omega \coth\left(\frac{\Omega}{2T}\right) - (\omega + \Omega) \coth\left(\frac{\omega + \Omega}{2T}\right) + (\omega - \Omega) \coth\left(\frac{\omega - \Omega}{2T}\right) \right]
\]

Formula based on the Kubo formula for the conductivity

by Shulga 1991
Inversion of reflectance is a VERY ill-defined problem, but the “image” of $\alpha^2 F(\Omega)$ is in the data.

$T = 0$ Holstein Theory:

$$\sigma(\nu) = \frac{\omega_p^2 i}{4\pi \nu} \int_0^\nu d\omega \frac{1}{\nu + \frac{i}{\tau} - \Sigma(\nu - \omega + i\delta) - \Sigma(\omega + i\delta)}$$

where

$$\Sigma(\omega + i\delta) = \int_0^\infty d\Omega \alpha^2 F(\Omega) \log \left| \frac{\Omega - \omega}{\Omega + \omega} \right| - i\pi \int_0^{\omega} d\Omega \alpha^2 F(\Omega)$$

Numerical inversion is possible but requires high precision (Pb)

“Poor man’s” inversion — use perturbation theory

Find:

$$\alpha^2 F(\nu) = \frac{\omega_p^2}{4\pi} \frac{1}{2\pi d\nu^2} \{\nu Re \left( \frac{1}{\sigma(\nu)} \right) \} \quad \text{or} \quad W(\omega) = \frac{1}{2\pi} \frac{d^2}{d\omega^2} \left[ \omega \frac{1}{\tau(\omega)} \right]$$
\[ \alpha^2 F(\nu) = \frac{\omega_P^2}{4\pi} \frac{1}{2\pi} \frac{d^2}{d\nu^2} \left\{ \nu \text{Re} \left( \frac{1}{\sigma(\nu)} \right) \right\} \]
$\alpha^2 F(\nu) = \frac{\omega_F^2}{4\pi} \frac{1}{2\pi d\nu^2} \{ \nu \text{Re} \left( \frac{1}{\sigma(\nu)} \right) \}$

$G(\nu)$

$\alpha^2 F(\nu)$


$T_c = 19 \text{ K}$

$\lambda = 0.8$

$\mu^* = 0.3$

very BCS-like
Coupling to 41 meV mode

J.P. Carbotte, E. Schachinger and D.N. Basov, Nature 401, 354(1999)

\[ W(\omega) = \frac{1}{2\pi} \frac{d^2}{d\omega^2} \left[ \omega \frac{1}{\tau(\omega)} \right] \]

The peak in \( W(\omega) \): \( \Delta + \Omega \)

Because of d-wave pairing, the peak is shifted by \( \Delta \) (not \( 2\Delta \)!)

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The peak in $W(\omega)$: $\Delta + \Omega$

Because of d-wave pairing, the peak is shifted by $\Delta$ (not $2\Delta$!)

$$W(\omega) = \frac{1}{2\pi} \frac{d^2}{d\omega^2} \left[ \omega \frac{1}{\tau(\omega)} \right]$$
the bosonic spectral function cannot be negative. The negative values are linked with the overshoot in $1/\tau(\omega)$.

A mode is unable to cause a overshoot in $1/\tau$
Allen’s formula for the scattering rate in the superconducting state

\[
1/\tau(\omega) = \frac{2\pi}{\omega} \int_0^{\omega-2\Delta} d\Omega (\omega - \Omega) \alpha^2 F(\Omega) E \left[ \sqrt{1 - \frac{4\Delta^2}{(\omega - \Omega)^2}} \right]
\]

E(x) is the second kind elliptic integral

P.B. Allen 1971

FIG. 9. Model spectral function $\alpha^2 F(\omega)$ (thin line) is used to calculate the scattering rate $1/\tau_{cal}(\omega)$ from Eq. (13). For $\Delta=0$ the calculated scattering rate resembles $1/\tau(\omega)$ of underdoped YBa$_2$Cu$_3$O$_{6.60}$ (Fig. 7). However, for finite values of the gap the calculated scattering rate resembles $1/\tau(\omega)$ of optimally doped YBa$_2$Cu$_3$O$_{6.95}$: there is an overshoot following the suppressed region (Fig. 7).
\[
\frac{1}{\tau(\omega, T)} = \frac{\pi}{\omega} \int_0^{+\infty} d\Omega \alpha^2 F(\Omega) \int_{-\infty}^{+\infty} dz \left[ N(z - \Omega) + N(-z + \Omega) \right] \\
\left[ n_B(\Omega) + 1 - f(z - \Omega) \right] \left[ f(z - \omega) - f(z + \omega) \right]
\]

\[
N(z) = \left[ N(0) + \left(1 - N(0)\right) \frac{z^2}{\Delta^2} \theta(\Delta - |z|) + \theta(|z| - \Delta) \right]
\]

\[
\alpha^2 F(\Omega) = PK(\Omega) + BG(\Omega)
\]

\[
PK(\Omega) = \frac{A}{\sqrt{2\pi}(d/2.35)} e^{-\left(\Omega - \Omega_{PR}\right)^2/[2(d/2.35)^2]}
\]

\[
BG(\Omega) = \frac{I_s \Omega}{\Omega_0^2 + \Omega^2}
\]

**Gap Function**

**Y123-Ortholl**

**$YBa_2Cu_3O_{6.50}$ Ortholl**
Angle-Resolved Photoemission Spectroscopy (ARPES)

$A(k, \omega) = \frac{1}{\pi} \frac{\text{Im} \sum (\omega, k)}{[\omega - \varepsilon_k - \text{Re} \sum (\omega, k)]^2 + [\text{Im} \sum (\omega, k)]^2}$

**PEAK POSITION:** Dispersion  (velocity; Effective mass; etc.)

**PEAK WIDTH:** $\text{Im} \Sigma$ or $1/\tau$ scattering rate
ARPES – Band Mapping and Fermi Surface

Energy Distribution Curve (EDC)

Photoelectron Intensity (Arb. Unit)

Kinetic Energy
\[ \lambda = - (\partial \text{Re} \Sigma / \partial \omega)_{E_F} \]
Dispersion of LSCO along the (0,0)-(\(\pi,\pi\)) Nodal Direction

Fine structures

X. J. Zhou et al., PRL (05)
Extraction of Bosonic Spectral Function from Re\(\Sigma\)

In metals, the real part of self-energy is related to the bosonic spectral function by:

\[
\text{Re} \Sigma(k, \varepsilon, T) = \int_0^\infty d\omega \alpha^2 F(\omega; \varepsilon, k) K\left(\frac{\varepsilon}{kT}, \frac{\omega}{kT}\right)
\]

where

\[
K(y, y') = \int_{-\infty}^\infty dx \frac{2y'}{x^2 - y'^2} f(x + y)
\]

with \(f(x)\) being the Fermi-Dirac distribution function

**Maximum Entropy Method** \(\Rightarrow \alpha^2 F(\omega)\)
Comparison of the Extracted Spectral Function with Known Structure

Hayden et al.,

McQueeney et al.,
PRL 87(2001) 077001

(1). el-ph
(2). multiple phonons
Thanks!