Optical Spectroscopy [Armitage]

- Most physical systems are understood by their response to natural frequency responses
  - ▲ For solid, & electromagnetic waves, freq. goes from radio to X-ray (meV ~ KeV)
  - ▲ Conversions: 200 GHz ~ 10K ~ 1 meV ~ $10\,cm^{-1}$

- Relevant Energy Scales

  10 eV ⟶ 10 KeV : chemical bonding

  0.5 eV ⟶ 10 eV : on-site Coulomb $|U|$, nearest-neighbor hopping $|t|$

  0 ⟶ 0.5 eV : collective magnetic modes

  0 ⟶ 0.1 eV : collective phonon modes

  ~ 50 meV : superconducting gaps in cuprates

  1 meV ⟶ 10 meV : scattering rate in clean metals

  0.1 meV ⟶ 5 meV : SC-gaps in conventional SC.

  ~0.01 meV : scattering rate in heavy fermion

- We shall consider experiments that determine complex conductivity $\sigma(q,\omega)$ in limit $q \to 0$.
  (we shall also consider complex dielectric $\epsilon(q,\omega)$)

WARNING: We shall use the language of non-interacting electrons in describing most results (e.g. electron scattering rate, masses, etc.) Formally these should treat as convenient parametrization

- We shall treat EM-field classically, described by Maxwell's eqn.
  We shall assume linear regime:
  $$\begin{cases} P = \chi_e E \\ M = \chi_m H \\ J = \sigma E \end{cases}$$

NOTE: Nonlinear regime can be important in, e.g. semi-conductors

Then, $\epsilon = 1 + 4\pi\chi_e$ and $\mu = 1 - 4\pi\chi_m$. We shall assume $\mu = 1$ throughout.

Then, $\vec{D} = \int_{-\infty}^{t}\int \epsilon(\vec{r},\vec{r}';t,t')\vec{E}(\vec{r}',t')\,d^3\vec{r}'dt'$

$\vec{J} = \int_{-\infty}^{t}\int \sigma(\vec{r},\vec{r}';t,t')\vec{E}(\vec{r}',t')\,d^3\vec{r}'dt'$

Fourier transform gives $\epsilon(q,\omega)$ and $\sigma(q,\omega)$, and these two are related by $\sigma(q,\omega) = \frac{i\omega}{4\pi}(1-\epsilon(q,\omega))$

Often we shall see $\epsilon(q,\omega) = \epsilon_1 + \frac{4\pi\sigma_1}{\omega}$

We shall consider $q \to 0$, hence assuming D, E, J, M, & P are locally homogeneous and porportional to each other.

The response function satisfies causality, from which we obtain the Kramers–Kronig relations:

$$\left(\mathcal{P}\int_{-\infty}^{\infty} d\omega' \frac{\sigma(\omega')}{\omega'-\omega}\right) - i\pi\sigma(\omega) = 0$$

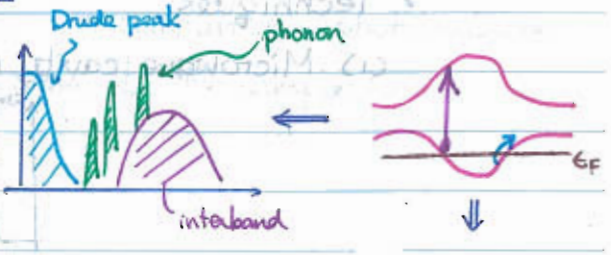
Since in time domain $\mathrm{Im}\{\sigma(\tau)\} = 0$, $\sigma(-\omega) = \sigma^*(\omega)$ and hence:

$$\sigma_1(\omega) = \frac{2}{\pi}\mathcal{P}\int_0^{\infty} d\omega' \frac{\omega'\sigma_2(\omega')}{\omega'^2-\omega^2}$$

$$\sigma_2(\omega) = -\frac{2\omega}{\pi}\mathcal{P}\int_0^{\infty} d\omega' \frac{\sigma_1(\omega')}{\omega'^2-\omega^2}$$

Generally,

$$\sigma_1^T(\omega) = \frac{\pi e^2}{m^2\omega}\frac{2}{(2\pi)^3}|\langle s|\vec{p}|s'\rangle|^2 D_{ss'}(\hbar\omega)$$

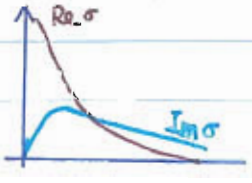


- Drude Model:

$$m\ddot{x} = -eE - m\dot{x}/\tau$$

$$\Longrightarrow x = \left(\frac{e\tau E_0}{m}\frac{1}{1-i\omega\tau}\right)e^{-i\omega t} \quad \text{for } E = E_0 e^{-i\omega t}$$

$$\Longrightarrow \sigma = \frac{Ne^2\tau}{m}\frac{1}{1-i\omega\tau} = \frac{Ne^2\tau}{m}\frac{1+i\omega\tau}{1+\omega^2\tau^2}$$



$$\mathrm{Im}\,\sigma \sim \begin{cases} \omega & \text{for } \omega \to 0 \\ 1/\omega & \text{for } \omega \nearrow \infty \end{cases}$$

$$\mathrm{Re}\,\sigma \sim 1/\omega^2 \text{ for } \omega \nearrow \infty$$

When $\frac{1}{\tau} \to 0$, $\begin{cases} \sigma_1 = \frac{\pi}{2} \frac{Ne^2}{m} \delta(\omega=0) \\ \sigma_2 = \frac{Ne^2}{m\omega} \end{cases}$

Optical mass obtained in Drude model is in general agreement with band mass (from heat capacity).

The Drude model can be improved by introducing classical restoring force $F = -\kappa x$. This gives Drude-Lorentz model:

$$\sigma(\omega) = \frac{Ne^2}{m} \frac{\omega}{i(\omega_0^2 - \omega^2) + i\omega/\tau} \qquad [\omega_0^2 = \kappa/m]$$

- Kubo Formalism

$$\mathcal{H}_{int} \approx -\frac{1}{c} \int d^3\vec{r}\ J^T(\vec{r})\ A^T(\vec{r})$$

From Fermi's golden rule & from $\mathcal{P}$ (dissipation power) $= \sigma E^2$, we get:

$$\sigma_1^T(q,\omega) = \sum_s \frac{1}{\hbar\omega} \int dt\ \langle s | J^T(q,0)\ J^{T*}(q,t) | s \rangle\ e^{-i\omega t}$$

Taking Fermi statistics into account, get:

$$\sigma_1^T(q,\omega) = \frac{\pi e^2}{m^2\omega} \frac{2}{(2\pi)^3} |\langle s|p|s' \rangle|^2 D_{s's}(\hbar\omega)$$

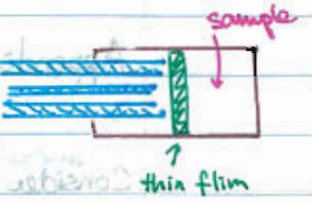
($D_{s's}(\hbar\omega)$: joint density of state)

- Techniques

(1) Microwave cavity resonator

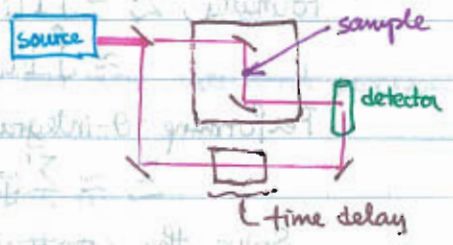
- ▲ Large range of freq.
- ▲ Only discrete frequencies are available.
- ▲ Highly sensitive.
- ▲ measured quantity: freq. shift & freq. boardening.

(2) Broadband Corbino Spectroscopy



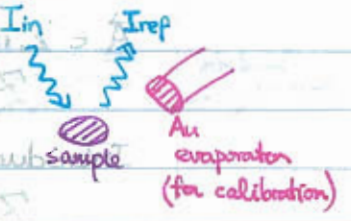
- 10 MHz – 200 GHz
- Measure power & phase shift of reflected wave

(3) THz time domain measurements



- Measurement in time domain
- 80 GHz – 2.5 THz

(4) Fourier Transform Infrared Reflectivity.



- Only measure reflectivity. Not adaquate to get complex σ. Need to use Krameus–Kronig, hence can be unreliable.
- Ellipsometry: send in polaized light. s & p channel will have different reflechivity ⇒ outgoing light is elliptically polaized.
- Reflectivity is suface sensitive. But it can get through ~10–100 layers of atoms (in comparison, photoemission probe only the first few atoms of the sample).