Dielectric function and optical conductivity

Y. T. Lee*

Institute for Solid State Physics, The University of Tokyo, Kashiwanoha, Kashiwa, Chiba, Japan

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1 Drude Model

Drude's model provides a simple initial description for the optical response of metals. Based on the assumption of Drude's model, the electrons in a metal will be accelerated by external electrical and magnetic fields. The behavior of electrons can be described as the Eq. (1)

$$m_e a_e(\omega) = -eE(\omega) - e\frac{v_e(\omega)}{c} \times \overrightarrow{B} - m_e \frac{v_e(\omega)}{\tau} \tag{1}$$

where τ is the relaxation time, the constant c is the speed of light, $\overrightarrow{B}$ is an external magnatic field, and

$$a_e(\omega) = -i\omega \overrightarrow{v_e} e^{-i\omega t} \tag{2}$$

$$E(\omega) = \overrightarrow{E_0} e^{-i\omega t} \tag{3}$$

$$v_e(\omega) = \overrightarrow{v_e} e^{-i\omega t}. \tag{4}$$

After reorganizing the Eq. (1), the Eq. (5) can be obtained.

*ytlee@issp.u-tokyo.ac.jp

$$m_e(-i\omega\vec{v_e}e^{-i\omega t}) = -e\vec{E_0}e^{-i\omega t} - e\frac{\vec{v_e}e^{-i\omega t}}{c} \times \vec{B} - m_e\frac{\vec{v_e}e^{-i\omega t}}{\tau} \tag{5}$$

Here, we assume that there is no magnetic field in the system (i.e. $B = 0$) and remove $e^{-i\omega t}$, the Eq. (5) becomes

$$-im_e\omega\vec{v_e} = -e\vec{E_0} - m_e\frac{\vec{v_e}}{\tau} \tag{6}$$

$$m_e\frac{\vec{v_e}}{\tau} - im_e\omega\vec{v_e} = -e\vec{E_0} \tag{7}$$

$$\vec{v_e}(\frac{m_e}{\tau} - im_e\omega) = -e\vec{E_0} \tag{8}$$

$$\vec{v_e}(1 - i\omega\tau) = -e\vec{E_0}\tau/m_e \tag{9}$$

$$\vec{v_e} = -\frac{e\vec{E_0}\tau/m_e}{1 - i\omega\tau} = \frac{e\vec{E_0}\tau/m_e}{i\omega\tau - 1} \tag{10}$$

where $\vec{v_e}$ is the mean velocity of electrons, m_e is the electron's mass, and $\vec{E_0}$ is external electric field. If the density of mobile electrons is n, the current density $\vec{j}$ arising in response to $\vec{E_0}$ is

$$\vec{j} = -ne\vec{v_e} = \frac{ne^2\tau/m_e}{1 - i\omega\tau}\vec{E_0} \tag{11}$$

Because of

$$\vec{j} = \sigma(\omega)\vec{E_0}, \tag{12}$$

electrical conducivity $\sigma(\omega)$ is

$$\sigma(\omega) = \frac{ne^2\tau/m_e}{1 - i\omega\tau} \tag{13}$$

where $\sigma(\omega)$ is the linear relation between current density $\vec{j}$ and an external electric field $\vec{E_0}$. At low-frequency region, ω is a small number in the denominator of the Eq. (13) and ω can be ignored. Thus, the electrical conducivity $\sigma(\omega)$ is given by

$$\sigma = ne^2\tau/m_e. \tag{14}$$

On the other hand, at higher frequencies, current and field fall out of phase. Many features of the optical response of metals can be described by the Eq. (13) which is frequency-dependent.

2 Maxwell's equations

Maxwell's equations are fundamental equations that describe the interactions between particles (with charge Q and electronic density / particle density $n(t)$) and electro-magnetic fields in matter.

$$\nabla \cdot \vec{E} = 4\pi Qn(t) \tag{15}$$

$$\nabla \cdot \vec{B} = 0 \tag{16}$$

$$\nabla \times \vec{E(t)} = -\frac{1}{c}\frac{d\vec{B}}{dt} \tag{17}$$

$$\nabla \times \vec{B(t)} = \frac{4\pi}{c}\vec{j} + \frac{1}{c}\frac{d\vec{E}}{dt} \tag{18}$$

Based on the Eq. (15) and (18), the continuity equation (the Eq. (25)) with $\vec{j}$ the charge current density can be derived as below.

Step 1.

$$\nabla \cdot \vec{E} = 4\pi Q n(t) \tag{19}$$

$$\frac{\partial}{\partial t}\nabla \cdot \vec{E} = \nabla \cdot \frac{\partial \vec{E}}{\partial t} = 4\pi Q \frac{\partial n(t)}{\partial t} \tag{20}$$

Step 2.

$$\nabla \times \vec{B}(t) = \frac{4\pi}{c}\vec{j} + \frac{1}{c}\frac{d\vec{E}}{dt} \tag{21}$$

$$\nabla \cdot (\nabla \times \vec{B}(t)) = \frac{4\pi}{c}\nabla \cdot \vec{j} + \frac{1}{c}\nabla \cdot \frac{d\vec{E}}{dt} \tag{22}$$

$$0 = \frac{4\pi}{c}\nabla \cdot \vec{j} + \frac{1}{c}4\pi Q \frac{\partial n(t)}{\partial t} \tag{23}$$

$$\frac{4\pi}{c}\nabla \cdot \vec{j} = -\frac{4\pi Q}{c}\frac{\partial n(t)}{\partial t} \tag{24}$$

$$\nabla \cdot \vec{j} = -Q\frac{dn(t)}{dt} \tag{25}$$

Furthermore, according to

$$\vec{E} = -\nabla V \tag{26}$$

(where V is a scalar potential V) and the Eq. (15), the Poisson equation (the Eq. (28)) can be obatined as below.

$$\nabla \cdot \vec{E} = \nabla \cdot (-\nabla V) = 4\pi Q n(t) \tag{27}$$

$$\nabla^2 V = -4\pi Q n(t) \tag{28}$$

In order to distinguish the influence of external fields in matter, the electronic densities and currents in Maxwell's equations can be separated into "internal" and "external" regions (no overlap).

$$n = n_{int} + n_{ext} \tag{29}$$

$$\vec{j} = \vec{j}_{int} + \vec{j}_{ext} \tag{30}$$

On the other hand, the polarization $\vec{P}$ is defined by

$$\vec{P}(\vec{r}, t) = \int_{-\infty}^{t} dt' \vec{j}_{int}(r, t'). \tag{31}$$

Combining with the Eq. (25), one can get the Eq. (36) as below.

$$\nabla \cdot \vec{P}(\vec{r},t) = \nabla \cdot (\int_{-\infty}^{t} dt' \vec{j}_{int}(\vec{r},t')). \tag{32}$$

$$= \int_{-\infty}^{t} dt' \left(\nabla \cdot \vec{j}_{int}(\vec{r},t')\right) \tag{33}$$

$$= \int_{-\infty}^{t} dt' \left(-Q\frac{dn_{int}(\vec{r},t')}{dt'}\right) \tag{34}$$

$$= -Q\int_{-\infty}^{t} dt' \frac{dn_{int}(\vec{r},t')}{dt'} \tag{35}$$

$$\nabla \cdot \vec{P}(\vec{r},t) = -Qn_{int}(\vec{r},t) \tag{36}$$

Because the displacement field $\vec{D}$ is defined as

$$\vec{D} = \vec{E} + 4\pi\vec{P} \quad \text{or} \quad \vec{E} = \vec{D} - 4\pi\vec{P}, \tag{37}$$

Maxwell's equation can be rewritten as the Eq. (38)-(41).

$$\nabla \cdot \vec{D} = 4\pi Q n_{ext}(t) \tag{38}$$

$$\nabla \cdot \vec{B} = 0 \tag{39}$$

$$\nabla \times \vec{E}(t) = -\frac{1}{c}\frac{d\vec{B}}{dt} \tag{40}$$

$$\nabla \times \vec{B}(t) = \frac{4\pi}{c}\vec{j}_{ext} + \frac{1}{c}\frac{d\vec{D}}{dt} \tag{41}$$

In comparison with the Eq. (15)-(18), one can find that only the Eq. (38) and the Eq. (41) are different from the Eq. (15) and the Eq. (18), respectively. Both of them can be derived easily as below.

Prove the Eq. (38)*:*

$$\nabla \cdot \vec{E} = \nabla \cdot (\vec{D} - 4\pi\vec{P}) \tag{42}$$

$$\nabla \cdot \vec{E} = \nabla \cdot \vec{D} - 4\pi\nabla \cdot \vec{P} \tag{43}$$

$$\nabla \cdot \vec{D} = \nabla \cdot \vec{E} + 4\pi\nabla \cdot \vec{P} \tag{44}$$

$$\nabla \cdot \vec{D} = 4\pi Q n(\vec{r},t) + 4\pi(-Qn_{int}(\vec{r},t)) \tag{45}$$

$$\nabla \cdot \vec{D} = 4\pi Q \left[n(\vec{r},t) - n_{int}(\vec{r},t)\right] \tag{46}$$

$$\nabla \cdot \vec{D} = 4\pi Q \left[n_{ext}(\vec{r},t) + n_{int}(\vec{r},t) - n_{int}(\vec{r},t)\right] \tag{47}$$

$$\nabla \cdot \vec{D} = 4\pi Q n_{ext}(\vec{r},t) \tag{48}$$

Prove the Eq. (41)*:*

$$\nabla \times \overrightarrow{B}(t) = \frac{4\pi}{c}\overrightarrow{j} + \frac{1}{c}\frac{d\overrightarrow{E}}{dt} = \frac{4\pi}{c}(\overrightarrow{j}_{ext} + \overrightarrow{j}_{int}) + \frac{1}{c}\frac{d\overrightarrow{E}}{dt} \tag{49}$$

$$= \frac{4\pi}{c}\overrightarrow{j}_{ext} + \frac{4\pi}{c}\overrightarrow{j}_{int} + \frac{1}{c}\frac{d\overrightarrow{E}}{dt} \tag{50}$$

$$= \frac{4\pi}{c}\overrightarrow{j}_{ext} + \frac{4\pi}{c}\frac{d\overrightarrow{P}(\overrightarrow{r},t)}{dt} + \frac{1}{c}\frac{d\overrightarrow{E}}{dt} \tag{51}$$

$$= \frac{4\pi}{c}\overrightarrow{j}_{ext} + \frac{1}{c}\frac{d(\overrightarrow{E} + 4\pi\overrightarrow{P}(\overrightarrow{r},t))}{dt} \tag{52}$$

$$\nabla \times \overrightarrow{B}(t) = \frac{4\pi}{c}\overrightarrow{j}_{ext} + \frac{1}{c}\frac{d\overrightarrow{D}}{dt} \tag{53}$$

The advantage of this form of Maxwell's eqautions is that all of source terms come from external. In the interior of a matter, n_{ext} and $\overrightarrow{j}_{ext}$ vanish even though they can lead to fields inside the matter. As shown by the Eq. (15)-(18) and the Eq. (38)-(41), $\overrightarrow{E}$ is the total field in the material, whereas $\overrightarrow{D}$ is the field due to external sources. Thus, the value of $\overrightarrow{D}$ at any point **r** is independent of the material and is the same as if the material were absent.

Note:

$$\overrightarrow{P}(\overrightarrow{r},t) = \int_{-\infty}^{t} dt' \overrightarrow{j}_{int}(r,t'). \tag{54}$$

$$\frac{d\overrightarrow{P}(\overrightarrow{r},t)}{dt} = \frac{d\int_{-\infty}^{t} dt' \overrightarrow{j}_{int}(\overrightarrow{r},t')}{dt} = \overrightarrow{j}_{int}(\overrightarrow{r},t) \tag{55}$$

Now, we knew that the currect density $\overrightarrow{j}$ is defined in the Eq. (12), and it can be substituted into the Eq. (18) (Ampere-Maxwell law).

$$\nabla \times \overrightarrow{B} = \frac{4\pi}{c}\overrightarrow{j} + \frac{1}{c}\frac{d\overrightarrow{E}}{dt} = \frac{4\pi}{c}\overrightarrow{j} - \frac{i\omega}{c}E_0 e^{-i(\omega t - \overrightarrow{q}\cdot\overrightarrow{r})} \tag{56}$$

$$= \frac{4\pi}{c}\sigma(\omega)\overrightarrow{E} - \frac{i\omega}{c}\overrightarrow{E} \tag{57}$$

$$= -\frac{i\omega}{c}\overrightarrow{E}(1 + \frac{i4\pi\sigma(\omega)}{\omega}) \tag{58}$$

$$= -\frac{i\omega}{c}\overrightarrow{E}(\epsilon_1(\omega) + i\epsilon_2(\omega)) = -\frac{i\omega}{c}\overrightarrow{E}\epsilon(\omega) \tag{59}$$

where $\overrightarrow{E} = E_0 e^{-i(\omega t - \overrightarrow{q}\cdot\overrightarrow{r})}$.

$$\nabla \times \overrightarrow{B} = \frac{1}{c}\frac{d\overrightarrow{D}}{dt} = \frac{\epsilon}{c}\frac{d\overrightarrow{E}}{dt} = \frac{-i\omega\overrightarrow{E}\epsilon(\omega)}{c} \tag{60}$$

Based on (59) and (60), the conductivity in the Eq. (58) is connected to the dielectric constant. And, the dielectric constant ($\epsilon(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega)$) is defined as the Eq. (61).

$$\epsilon(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega) = \epsilon_1(\omega)(1 + i\frac{\epsilon_2(\omega)}{\epsilon_1(\omega)}) \tag{61}$$

$$= \epsilon_1(\omega)(1 + i\tan\delta(\omega)) \tag{62}$$

$$= 1 + i\frac{4\pi}{\omega}\sigma(\omega) \tag{63}$$

where $\epsilon_1(\omega)$ and $\epsilon_2(\omega)$ are the real and imaginary part of dielectric constant, respectively, and $\tan\delta$ is the ratio of $\epsilon_2(\omega)$ to $\epsilon_1(\omega)$ (i.e. $\epsilon_2(\omega)/\epsilon_1(\omega)$). Once the conductivity in the Eq. (63) is obatined, the dielectric function $\epsilon(\omega)$ can be evaluated to check how all charges respond to electrical fields. And, the value of loss tengent vector , i.e. $\tan\delta(\omega)$, can be compared with experimental results (such as Cole-Cole diagram).

2.1 Traveling Waves

If the electric fields is perpendicular to the matter, one can finds the relations as below by following the Eq. (40) and (53).

$$\nabla \times \nabla \times \overrightarrow{E}(t) = \nabla \times (-\frac{1}{c}\frac{d\overrightarrow{B}}{dt}) \tag{64}$$

$$(-i\overrightarrow{q})((-i\overrightarrow{q})\cdot\overrightarrow{E}) - (-iq)^2\overrightarrow{E} = (i\overrightarrow{q})(0) - (-iq)^2\overrightarrow{E} = -\frac{1}{c}\frac{d(\nabla\times\overrightarrow{B})}{dt} = -\frac{1}{c^2}\frac{\partial\epsilon(\omega)\overrightarrow{E}}{\partial t^2} \tag{65}$$

$$q^2\overrightarrow{E} = -\frac{\epsilon(\omega)}{c^2}\frac{\partial^2\overrightarrow{E}}{\partial t^2} \tag{66}$$

where $\overrightarrow{q}\cdot\overrightarrow{E} = 0$. And, one can know the relations as below

$$\overrightarrow{E} = E_0 e^{-i(\omega t - \overrightarrow{q}\cdot\overrightarrow{r})} \tag{67}$$

$$\frac{\partial\overrightarrow{E}}{\partial t} = -i\omega E_0 e^{-i(\omega t - \overrightarrow{q}\cdot\overrightarrow{r})} = -i\omega\overrightarrow{E} \tag{68}$$

$$\frac{\partial^2\overrightarrow{E}}{\partial t^2} = -\omega^2 E_0 e^{-i(\omega t - \overrightarrow{q}\cdot\overrightarrow{r})} = -\omega^2\overrightarrow{E} \tag{69}$$

$$\frac{\partial\overrightarrow{E}}{\partial r} = i\overrightarrow{q}E_0 e^{-i(\omega t - \overrightarrow{q}\cdot\overrightarrow{r})} = i\overrightarrow{q}\overrightarrow{E} \tag{70}$$

$$\frac{\partial^2\overrightarrow{E}}{\partial r^2} = i^2\overrightarrow{q}^2 E_0 e^{-i(\omega t - \overrightarrow{q}\cdot\overrightarrow{r})} = -\overrightarrow{q}^2\overrightarrow{E}, \tag{71}$$

then the Eq. (66) becomes

$$q^2\overrightarrow{E} = -\frac{\epsilon(\omega)}{c^2}(-\omega^2\overrightarrow{E}) \tag{72}$$

$$q^2 = \frac{\omega^2\epsilon(\omega)}{c^2}. \tag{73}$$

$$q = \frac{\omega\sqrt{\epsilon(\omega)}}{c}. \tag{74}$$

Since $\overrightarrow{q} = \omega\tilde{n}/c$, the real part of refactive index is $\tilde{n} = \sqrt{\epsilon(\omega)}$. Therefore, the real and imaginery part of dielectric function can be related by

$$\epsilon_1(\omega) = \tilde{n}^2 - \kappa^2 \tag{75}$$

$$\epsilon_2(\omega) = 4\pi\mathrm{Re}[\sigma]/\omega \tag{76}$$

where κ is the imaginery part of refractive index.

For longitudinal waves, $\overrightarrow{E}$ is parallel to $\overrightarrow{q}$, the Eq. (65) becomes

$$(-i\overrightarrow{q})((-i\overrightarrow{q})\cdot\overrightarrow{E}) - (-iq)^2\overrightarrow{E} = \epsilon(\overrightarrow{q},\omega)\frac{\omega^2}{c^2}\overrightarrow{E} \tag{77}$$

$$i^2q^2\overrightarrow{E} - (-iq)^2\overrightarrow{E} = \epsilon(\overrightarrow{q},\omega)\frac{\omega^2}{c^2}\overrightarrow{E} \tag{78}$$

$$-q^2\overrightarrow{E} + q^2\overrightarrow{E} = 0 = \epsilon(\overrightarrow{q},\omega)\frac{\omega^2}{c^2}\overrightarrow{E} \tag{79}$$

$$\epsilon(\overrightarrow{q},\omega) = 0 \tag{80}$$

3 Dielectric function

3.1 Dielectric loss

The total current $I(t)$ is defined as

$$I(t) = \frac{dD}{dt} \tag{81}$$

where D is the electric displacement induced by an electric field $\overrightarrow{E}$. And, D is always in phase with applied electric field $\overrightarrow{E}$ because the electric displacement D is induced by this field. Then, we can get the relation between D and E as below.

$$D(t) = \mathrm{Re}\left[\epsilon\overrightarrow{E}\right] = \mathrm{Re}\left[\epsilon_o\epsilon_r^* E_0 e^{i\omega t}\right] \tag{82}$$

$$= \mathrm{Re}\left[\epsilon_o(\epsilon_1 - i\epsilon_2)E_0(\cos\omega t + i\sin\omega t)\right] \tag{83}$$

$$= \epsilon_o E_0(\epsilon_1\cos\omega t + \epsilon_2\sin\omega t) \tag{84}$$

Then, the Eq. (82) is substituted into the Eq. (81).

$$I(t) = \frac{dD}{dt} = \frac{d(\epsilon_o\epsilon_r^* E_0 e^{i\omega t})}{dt} \tag{85}$$

$$= \epsilon_o\epsilon_r^* i\omega E_0 e^{i\omega t} \tag{86}$$

$$= \epsilon_o(\epsilon_1 - i\epsilon_2)i\omega E_0 e^{i\omega t} \tag{87}$$

$$= \epsilon_o(i\epsilon_1 + \epsilon_2)\omega E_0(\cos\omega t + i\sin\omega t) \tag{88}$$

$$= \epsilon_o\omega E_0[(\epsilon_2\cos\omega t - \epsilon_1\sin\omega t) + i(\epsilon_1\cos\omega t + \epsilon_2\sin\omega t)] \tag{89}$$

In reality, one can measure the real part of current density, not imaginary part. Thus, the $I_{re}(t)$ represents the current density.

Once a voltage $V(= V - 0)$ is applied into the system, one can know the relation between $\overrightarrow{E}$ and V (i.e. $E = V/l$). Thus, the work W within a period of ω can be estimated by

$$W = \frac{\int_0^{2\pi/\omega} I_{re}(t)V(t)dt}{\Omega \int_0^{2\pi/\omega} dt} \tag{90}$$

$$= \frac{\omega}{2\pi A l}\int_0^{2\pi/\omega} I_{re}(t)E(t)ldt = \frac{\omega l}{2\pi A l}\int_0^{2\pi/\omega} I_{re}(t)E_0\cos\omega t dt \tag{91}$$

$$= \frac{\omega}{2\pi A}\int_0^{2\pi/\omega} \epsilon_o\omega E_0(\epsilon_2\cos\omega t - \epsilon_1\sin\omega t)E_0\cos\omega t dt \tag{92}$$

$$= \frac{\omega^2\epsilon_o E_0^2}{2\pi A}\int_0^{2\pi/\omega} (\epsilon_2\cos\omega t - \epsilon_1\sin\omega t)\cos\omega t dt \tag{93}$$

$$= \frac{\omega^2\epsilon_o E_0^2}{2\pi A}\int_0^{2\pi/\omega} \left(\epsilon_2\cos^2\omega t - \epsilon_1\sin\omega t\cos\omega t\right) dt \tag{94}$$

$$= \frac{\omega^2\epsilon_o E_0^2}{2\pi A}\left[\int_0^{2\pi/\omega} \epsilon_2\cos^2\omega t dt - \int_0^{2\pi/\omega} \epsilon_1\sin\omega t\cos\omega t dt\right] \tag{95}$$

$$= \frac{\omega^2\epsilon_o E_0^2}{2\pi A}\left[\frac{\epsilon_2\pi}{\omega} + 0\right] \tag{96}$$

$$= \frac{\omega\epsilon_o\epsilon_2 E_0^2}{2A} \tag{97}$$

where Ω is the volumne within two plates, l is the distance between two plates, and A is the same surface area of two plates perpendicular to electric field. Therefore, one can know this work, called dielectric loss, is caused by electric fields and corresponding induced current density / electric displacement to absorb energy from external sources.

If ϵ_2 is 0, it will have no dielectric loss. But, the electric current still exists because the second term of the Eq. (62) contributes with 90 degree of phase-shift, i.e. the Eq. (101).

$$I(t) = \omega\epsilon_0\vec{E}(\epsilon_2\cos(\omega t) - \epsilon_1\sin(\omega t)) \tag{98}$$

$$= \omega\epsilon_0\vec{E}[0 - \epsilon_1\sin(\omega t)] \tag{99}$$

$$= \omega\epsilon_0\vec{E}[0 + \epsilon_1\cos(\omega t + 90^\circ)] \tag{100}$$

$$= \omega\epsilon_0\epsilon_1\vec{E}\cos(\omega t + 90^\circ). \tag{101}$$

According to the Eq. (62), the loss tangent angle δ is

$$\delta = \tan^{-1}\left(\frac{\omega\epsilon_0\epsilon_2\vec{E}}{\omega\epsilon_0\epsilon_1\vec{E}}\right) = \tan^{-1}\left(\frac{\epsilon_2}{\epsilon_1}\right) \tag{102}$$

3.2 Kramers-Krong relations

The Cauchy's residue theorem is that

$$\oint \frac{\chi(\omega')}{\omega' - \omega}d\omega' = 0 \tag{103}$$

for a closed contour within this area. One can choose the contour to trace the real axis with a hump over the pole at $\omega' = \omega$ and a large semi-circle in the upper half complex plane. Then, this integral can be decomposed into three contributions along (1) real axis, (2) half-circle at ω' pole, and (3) half-circle of upper half complex plane.

The integral through semi-circle path vanishs because $\chi(\omega')$ vanishes as fast as $1/|\omega'|$. Thus, one can get the relation as below.

$$0 = \oint \frac{\chi(\omega')}{\omega' - \omega} d\omega' = P \int_{-\infty}^{\infty} \frac{\chi(\omega')}{\omega' - \omega} d\omega' - i\pi\chi(\omega) \tag{104}$$

It can be reorganized as the Eq. (105).

$$\chi(\omega) = \frac{1}{i\pi} P \int_{-\infty}^{\infty} \frac{\chi(\omega')}{\omega' - \omega} d\omega' \tag{105}$$

Therefore, one can apply this equation into dielectric function by replacing $\chi(\omega)$ with $\epsilon(\omega) - \epsilon^{\infty}$.

$$\epsilon(\omega) - \epsilon^{\infty} = \frac{1}{i\pi} P \int_{-\infty}^{\infty} \frac{\epsilon(\omega) - \epsilon^{\infty}}{\omega' - \omega} d\omega' \tag{106}$$

And, the Eq. (106) can be separated into real and imaginary part.

$$[\epsilon_{re}(\omega) - \epsilon_{re}^{\infty}] + i[\epsilon_{im}(\omega) - \epsilon_{im}^{\infty}] = \frac{1}{i\pi} P \int_{-\infty}^{\infty} \frac{[\epsilon_{re}(\omega) - \epsilon_{re}^{\infty}] + i[\epsilon_{im}(\omega) - \epsilon_{im}^{\infty}]}{\omega' - \omega} d\omega' \tag{107}$$

$$= \left[\frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{[\epsilon_{im}(\omega) - \epsilon_{im}^{\infty}]}{\omega' - \omega} d\omega'\right] - i \left[\frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{[\epsilon_{re}(\omega) - \epsilon_{re}^{\infty}]}{\omega' - \omega} d\omega'\right] \tag{108}$$

$$\mathrm{Re}[\epsilon(\omega) - \epsilon^{\infty}] = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\mathrm{Im}[\epsilon(\omega') - \epsilon^{\infty}]}{\omega' - \omega} d\omega' \tag{109}$$

$$\mathrm{Im}[\epsilon(\omega) - \epsilon^{\infty}] = \frac{-1}{\pi} P \int_{-\infty}^{\infty} \frac{\mathrm{Re}[\epsilon(\omega') - \epsilon^{\infty}]}{\omega' - \omega} d\omega' \tag{110}$$

Because $\epsilon(t)$ is real, $\epsilon(\omega) = \epsilon^*(-\omega)$. It indicates that the real and imarinary part of $\epsilon(\omega)$ are even and odd, respectively. Thus, both equations can be rewritten as below.

$$\epsilon_{re}(\omega) - \epsilon^{\infty} = P \int_{-\infty}^{\infty} \frac{2\omega' d\omega'}{\pi} \frac{\epsilon_{im}(\omega')}{\omega'^2 - \omega^2} \tag{111}$$

$$\epsilon_{im}(\omega) = -P \int_{-\infty}^{\infty} \frac{2\omega d\omega'}{\pi} \frac{\epsilon_{re}(\omega') - \epsilon^{\infty}}{\omega'^2 - \omega^2} \tag{112}$$

When one measures the imaginary part of the dielectric function (i.e. absorption), it can be used to find the real part (i.e. dispersion) or vice versa.

Note:

Prove the Eq. (111) *from the Eq.* (109)*:*

$$\begin{aligned}
\mathrm{Re}[\epsilon(\omega)-\epsilon^{\infty}] &= \frac{1}{\pi}P\int_{-\infty}^{\infty}\frac{\mathrm{Im}[\epsilon(\omega')-\epsilon^{\infty}]}{\omega'-\omega}d\omega' && (113)\\
&= \frac{1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Im}[\epsilon(\omega')]}{\omega'-\omega}d\omega' + \frac{1}{\pi}P\int_{-\infty}^{0}\frac{\mathrm{Im}[\epsilon(\omega')]}{\omega'-\omega}d\omega' && (114)\\
&= \frac{1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Im}[\epsilon(\omega')]}{\omega'-\omega}d\omega' + \frac{1}{\pi}P\int_{\infty}^{0}\frac{\mathrm{Im}[\epsilon(-\omega'')]}{-\omega''-\omega}(-d\omega'') && (115)\\
&= \frac{1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Im}[\epsilon(\omega')]}{\omega'-\omega}d\omega' + \frac{1}{\pi}P\int_{\infty}^{0}\frac{-\mathrm{Im}[\epsilon(\omega'')]}{\omega''+\omega}d\omega'' && (116)\\
&= \frac{1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Im}[\epsilon(\omega')]}{\omega'-\omega}d\omega' + \frac{1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Im}[\epsilon(\omega'')]}{\omega''+\omega}d\omega'' && (117)\\
&= \frac{1}{\pi}P\int_{0}^{\infty}\mathrm{Im}[\epsilon(\omega'')]\left[\frac{1}{\omega''-\omega}+\frac{1}{\omega''+\omega}\right]d\omega'' && (118)\\
&= \frac{1}{\pi}P\int_{0}^{\infty}\mathrm{Im}[\epsilon(\omega'')]\left[\frac{\omega''+\omega}{\omega''^2-\omega^2}+\frac{\omega''-\omega}{\omega''^2-\omega^2}\right]d\omega'' && (119)\\
&= \frac{2}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Im}[\epsilon(\omega'')]\omega''d\omega''}{\omega''^2-\omega^2} = P\int_{0}^{\infty}\frac{2\omega''d\omega''}{\pi}\frac{\mathrm{Im}[\epsilon(\omega'')]}{\omega''^2-\omega^2} && (120)
\end{aligned}$$

where $\mathrm{Im}[\epsilon^{\infty}]=0$.

Prove the Eq. (112) *from the Eq.* (110)*:*

$$\begin{aligned}
\mathrm{Im}[\epsilon(\omega)-\epsilon^{\infty}] &= \frac{-1}{\pi}P\int_{-\infty}^{\infty}\frac{\mathrm{Re}[\epsilon(\omega')-\epsilon^{\infty}]}{\omega'-\omega}d\omega' && (121)\\
&= \frac{-1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Re}[\epsilon(\omega')-\epsilon^{\infty}]}{\omega'-\omega}d\omega' + \frac{-1}{\pi}P\int_{-\infty}^{0}\frac{\mathrm{Re}[\epsilon(\omega')-\epsilon^{\infty}]}{\omega'-\omega}d\omega' && (122)\\
&= \frac{-1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Re}[\epsilon(\omega')-\epsilon^{\infty}]}{\omega'-\omega}d\omega' + \frac{-1}{\pi}P\int_{\infty}^{0}\frac{\mathrm{Re}[\epsilon(-\omega'')-\epsilon^{\infty}]}{-\omega''-\omega}(-d\omega'') && (123)\\
&= \frac{-1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Re}[\epsilon(\omega')-\epsilon^{\infty}]}{\omega'-\omega}d\omega' + \frac{-1}{\pi}P\int_{\infty}^{0}\frac{\mathrm{Re}[\epsilon(-\omega'')-\epsilon^{\infty}]}{\omega''+\omega}d\omega'' && (124)\\
&= \frac{-1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Re}[\epsilon(\omega')-\epsilon^{\infty}]}{\omega'-\omega}d\omega' + \frac{1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Re}[\epsilon(-\omega'')-\epsilon^{\infty}]}{\omega''+\omega}d\omega'' && (125)\\
&= \frac{-1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Re}[\epsilon(\omega')-\epsilon^{\infty}]}{\omega'-\omega}d\omega' + \frac{1}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Re}[\epsilon(\omega'')-\epsilon^{\infty}]}{\omega''+\omega}d\omega'' && (126)\\
&= \frac{1}{\pi}P\int_{0}^{\infty}\mathrm{Re}[\epsilon(\omega')-\epsilon^{\infty}]\left[\frac{-1}{\omega''-\omega}+\frac{1}{\omega''+\omega}\right]d\omega'' && (127)\\
&= \frac{1}{\pi}P\int_{0}^{\infty}\mathrm{Re}[\epsilon(\omega')-\epsilon^{\infty}]\left[\frac{-\omega''-\omega}{\omega''^2-\omega^2}+\frac{\omega''-\omega}{\omega''^2-\omega^2}\right]d\omega'' && (128)\\
&= \frac{-2}{\pi}P\int_{0}^{\infty}\frac{\mathrm{Re}[\epsilon(\omega'')-\epsilon^{\infty}]\omega d\omega''}{\omega''^2-\omega^2} = -P\int_{0}^{\infty}\frac{2\omega d\omega''}{\pi}\frac{\mathrm{Re}[\epsilon(\omega'')-\epsilon^{\infty}]}{\omega''^2-\omega^2} && (129)
\end{aligned}$$

3.3 Kubo-Greenwood formula

3.3.1 Born Approximation

Begin with Born approximation, which states that an eigenstate $|l\rangle$ of $\hat{H}$ comes in contact with a weak time-dependent potential $\hat{U}(t)$ evloves into

$$\left|\tilde{l}(t)\right\rangle \approx N\left[e^{-i\hat{H}t/\hbar}\left|l\right\rangle + \int_{-\infty}^{t} dt' e^{-i\hat{H}(t-t')/\hbar}\frac{\hat{U}(t')}{i\hbar}e^{-i\hat{H}(t')/\hbar}\left|l\right\rangle\right] \tag{130}$$

$$= N\left[e^{-i\omega_l t}\left|l\right\rangle + \int_{-\infty}^{t} dt' e^{-i\omega_{l'}(t-t')}\frac{\hat{U}e^{-i\omega t'}}{i\hbar}e^{-i\omega_l t'}\left|l\right\rangle\right] \tag{131}$$

$$= N\left[e^{-i\omega_l t}\left|l\right\rangle + \int_{-\infty}^{t} dt' e^{-i\omega_{l'}(t-t')}\sum_{l'}\left|l'\right\rangle\left\langle l'\right|\frac{\hat{U}}{i\hbar}e^{-i\omega_l t'}\left|l\right\rangle e^{-i\omega t'}\right] \tag{132}$$

$$= N\left[e^{-i\omega_l t}\left|l\right\rangle + \frac{1}{i\hbar}\sum_{l'}\int_{-\infty}^{t} dt'\left|l'\right\rangle e^{-i\omega_{l'}(t-t')}\left\langle l'\right|\hat{U}\left|l\right\rangle e^{-i\omega_l t'}e^{-i\omega t'}\right] \tag{133}$$

$$= N\left[e^{-i\omega_l t}\left|l\right\rangle + \sum_{l'}\frac{\left|l'\right\rangle\left\langle l'\right|\hat{U}\left|l\right\rangle}{i\hbar}\int_{-\infty}^{t} dt' e^{-i\omega_{l'}(t-t')}e^{-i\omega_l t'}e^{-i\omega t'}\right] \tag{134}$$

$$= N\left[e^{-i\omega_l t}\left|l\right\rangle + \sum_{l'}\frac{\left|l'\right\rangle\left\langle l'\right|\hat{U}\left|l\right\rangle}{i\hbar}e^{-i\omega_{l'}t}\int_{-\infty}^{t} dt' e^{i\omega_{l'}t'}e^{-i\omega_l t'}e^{-i\omega t'}\right] \tag{135}$$

$$= N\left[e^{-i\omega_l t}\left|l\right\rangle + \sum_{l'}\frac{\left|l'\right\rangle\left\langle l'\right|\hat{U}\left|l\right\rangle}{i\hbar}e^{-i\omega_{l'}t}\int_{-\infty}^{t} dt' e^{-i(-\omega_{l'}+\omega_l+\omega)t'}\right] \tag{136}$$

When the range of the integral is from $-\infty$ to t, one can evaluate $\int_{-\infty}^{t} dt' e^{-i(-\omega_{l'}+\omega_l+\omega)t'}$ as follows:

$$\int_{-\infty}^{t} dt' e^{-i(-\omega_{l'}+\omega_l+\omega)t'} = \lim_{\eta\to 0^+}\int_{-\infty}^{t} dt' e^{-i(-\omega_{l'}+\omega_l+\omega+i\eta)t'} = \lim_{\eta\to 0^+}\int_{-\infty}^{t} dt' e^{i(\omega_{l'}-\omega_l-\omega-i\eta)t'} \tag{137}$$

$$= \lim_{\eta\to 0^+}\left.\frac{e^{i(\omega_{l'}-\omega_l-\omega-i\eta)t'}}{i(\omega_{l'}-\omega_l-\omega-i\eta)}\right|_{-\infty}^{t} = \frac{e^{i(\omega_{l'}-\omega_l-\omega-i\eta)t}}{i(\omega_{l'}-\omega_l-\omega-i\eta)} - \frac{e^{i(\omega_{l'}-\omega_l-\omega-i\eta)(-\infty)}}{i(\omega_{l'}-\omega_l-\omega-i\eta)} \tag{138}$$

$$= \frac{e^{i(\omega_{l'}-\omega_l-\omega-i\eta)t}}{i(\omega_{l'}-\omega_l-\omega-i\eta)} - \frac{e^{i(\omega_{l'}-\omega_l-\omega)(-\infty)+i^2\eta\infty}}{i(\omega_{l'}-\omega_l-\omega-i\eta)} \tag{139}$$

$$= \frac{e^{i(\omega_{l'}-\omega_l-\omega-i\eta)t}}{i(\omega_{l'}-\omega_l-\omega-i\eta)} - \frac{e^{i(\omega_{l'}-\omega_l-\omega)(-\infty)}e^{-\eta\infty}}{i(\omega_{l'}-\omega_l-\omega-i\eta)} = \frac{e^{i(\omega_{l'}-\omega_l-\omega-i\eta)t}}{i(\omega_{l'}-\omega_l-\omega-i\eta)} \tag{140}$$

where $e^{i(\omega_{l'}-\omega_l-\omega)(-\infty)}$ is a sinusoidal wave and $e^{-\eta\infty}$ is close to 0 at a small η. Therefore,

$$\left|\tilde{l}(t)\right\rangle \approx N\left[e^{-i\omega_l t}\left|l\right\rangle + \sum_{l'}\frac{\left|l'\right\rangle\left\langle l'\right|\hat{U}\left|l\right\rangle}{i\hbar}e^{-i\omega_{l'}t}\lim_{\eta\to 0^+}\int_{-\infty}^{t} dt e^{-i(\omega_l-\omega_{l'}+\omega+i\eta)t'}\right] \tag{141}$$

$$= N\left[e^{-i\omega_l t}\left|l\right\rangle + \sum_{l'}\frac{\left|l'\right\rangle\left\langle l'\right|\hat{U}\left|l\right\rangle}{i\hbar}e^{-i\omega_{l'}t}\frac{e^{i(\omega_{l'}-\omega_l-\omega-i\eta)t}}{i(\omega_{l'}-\omega_l-\omega-i\eta)}\right] \tag{142}$$

$$= N\left[e^{-i\omega_l t}\left|l\right\rangle + \sum_{l'}\frac{\left|l'\right\rangle\left\langle l'\right|\hat{U}\left|l\right\rangle}{i\hbar}\frac{e^{-i\omega_{l'}t}e^{i\omega_{l'}t}e^{-i\omega_l t}e^{-i(\omega+i\eta)t}}{i(\omega_{l'}-\omega_l-\omega-i\eta)}\right] \tag{143}$$

$$= N\left[\left|l\right\rangle + \sum_{l'}\frac{\left|l'\right\rangle\left\langle l'\right|\hat{U}\left|l\right\rangle}{i\hbar}\frac{e^{-i(\omega+i\eta)t}}{i(\omega_{l'}-\omega_l-\omega-i\eta)}\right]e^{-i\omega_l t} \tag{144}$$

$$= N\left[\left|l\right\rangle + \sum_{l'}\frac{\left|l'\right\rangle\left\langle l'\right|\hat{U}\left|l\right\rangle}{\hbar}\frac{e^{-i(\omega+i\eta)t}}{(-\omega_{l'}+\omega_l+\omega+i\eta)}\right]e^{-i\omega_l t} \tag{145}$$

$$\left|\tilde{l}(t)\right\rangle \approx N\left[|l\rangle + \sum_{l'} \frac{|l'\rangle \langle l'| \hat{U} |l\rangle e^{-i(\omega+i\eta)t}}{\hbar(\omega_l - \omega_{l'} + \omega + i\eta)}\right] e^{-i\omega_l t} \tag{146}$$

$$= N\left[|l\rangle + \sum_{l'} \frac{|l'\rangle \langle l'| \hat{U} |l\rangle e^{-i\omega' t}}{\hbar(\omega_l - \omega_{l'} + \omega')}\right] e^{-i\omega_l t} \tag{147}$$

where N is the normalization factor and $\omega' = \omega + i\eta$. And, $\hat{U}^*(t) = \hat{U}^* e^{i\omega^* t}$. One can find similar $\left|\tilde{l}(t)\right\rangle$ as below.

$$\left|\tilde{l}(t)\right\rangle \approx N\left[e^{-i\hat{H}t/\hbar}|l\rangle + \int_{-\infty}^{t} dt' e^{-i\hat{H}(t-t')/\hbar} \frac{\hat{U}^*(t')}{i\hbar} e^{-i\hat{H}(t')/\hbar}|l\rangle\right] \tag{148}$$

$$= N\left[e^{-i\omega_l t}|l\rangle + \int_{-\infty}^{t} dt' e^{-i\omega_{l'}(t-t')} \frac{\hat{U}^* e^{i\omega^* t'}}{i\hbar} e^{-i\omega_l t'}|l\rangle\right] \tag{149}$$

$$= N\left[e^{-i\omega_l t}|l\rangle + \int_{-\infty}^{t} dt' \sum_{l'} |l'\rangle \langle l'| \frac{\hat{U}^*}{i\hbar} |l\rangle e^{-i\omega_{l'}(t-t')} e^{-i\omega_l t'} e^{i\omega^* t'}\right] \tag{150}$$

$$= N\left[e^{-i\omega_l t}|l\rangle + \sum_{l'} \frac{|l'\rangle \langle l'| \hat{U}^* |l\rangle}{i\hbar} \int_{-\infty}^{t} dt' e^{-i\omega_{l'}(t-t')} e^{-i\omega_l t'} e^{i\omega^* t'}\right] \tag{151}$$

$$= N\left[e^{-i\omega_l t}|l\rangle + \sum_{l'} \frac{|l'\rangle \langle l'| \hat{U}^* |l\rangle}{i\hbar} e^{-i\omega_{l'} t} \int_{-\infty}^{t} dt' e^{i\omega_{l'} t'} e^{-i\omega_l t'} e^{i\omega^* t'}\right] \tag{152}$$

$$= N\left[e^{-i\omega_l t}|l\rangle + \sum_{l'} \frac{|l'\rangle \langle l'| \hat{U}^* |l\rangle}{i\hbar} e^{-i\omega_{l'} t} \lim_{\eta \to 0^-} \int_{-\infty}^{t} dt' e^{i(\omega_{l'} - \omega_l + \omega - i\eta)t'}\right] \tag{153}$$

$$= N\left[e^{-i\omega_l t}|l\rangle + \sum_{l'} \frac{|l'\rangle \langle l'| \hat{U}^* |l\rangle}{i\hbar} e^{-i\omega_{l'} t} \frac{e^{i(\omega_{l'} - \omega_l + \omega - i\eta)t}}{i(\omega_{l'} - \omega_l + \omega - i\eta)}\right] \tag{154}$$

$$= N\left[e^{-i\omega_l t}|l\rangle + \sum_{l'} \frac{|l'\rangle \langle l'| \hat{U}^* |l\rangle}{i\hbar} \frac{e^{-i\omega_l t} e^{i(\omega - i\eta)t}}{i(\omega_{l'} - \omega_l + \omega - i\eta)}\right] \tag{155}$$

$$= N\left[|l\rangle + \sum_{l'} \frac{|l'\rangle \langle l'| \hat{U}^* |l\rangle e^{i(\omega - i\eta)t}}{\hbar(\omega_l - \omega_{l'} - (\omega - i\eta))}\right] e^{-i\omega_l t} \tag{156}$$

$$= N\left[|l\rangle + \sum_{l'} \frac{|l'\rangle \langle l'| \hat{U}^* |l\rangle e^{i\omega'^* t}}{\hbar(\omega_l - \omega_{l'} - \omega'^*)}\right] e^{-i\omega_l t} \tag{157}$$

where $\omega'^* = \omega - i\eta$.

3.3.2 Conductivity tensor

One can treat the light as a spatially uniform oscillating field with

$$\vec{A} = \frac{c\vec{E}}{i\omega} e^{-i\omega t} + c.c. \tag{158}$$

And, the current operator is

$$\hat{j} = -\frac{e}{m}\left[\hat{P} + \frac{e}{c}\hat{A}\right]. \tag{159}$$

To linear order in applied fileds, the Hamiltonian is changed by addition of a term

$$\hat{U}(t) = \frac{e}{mi\omega}[\vec{E}\cdot\hat{P}]e^{-i\omega t} - \frac{e}{mi\omega^*}[\vec{E}\cdot\hat{P}]e^{i\omega^* t} = \hat{U}e^{-i\omega t} + \hat{U}^* e^{i\omega^* t} \tag{160}$$

Therefore, the contribution to the current of state $\left|\tilde{l}\right\rangle$ is

$$\begin{aligned}
\vec{J} = \left\langle \tilde{l}\right| \hat{j} \left|\tilde{l}\right\rangle &= -\frac{e}{m}\left\langle \tilde{l}\right| \hat{P} + \frac{e}{c}\hat{A}\left|\tilde{l}\right\rangle & (161)\\
&= -\frac{e}{m}\left\langle \tilde{l}\right| \hat{P} + \frac{e\vec{E}}{i\omega}e^{-i\omega t} - \frac{e\vec{E}}{i\omega^*}e^{i\omega^* t}\left|\tilde{l}\right\rangle & (162)\\
&= -\frac{e}{m}\left\langle \tilde{l}\right| \hat{P}\left|\tilde{l}\right\rangle - \frac{e}{m}\left\langle \tilde{l}\right| \frac{e\vec{E}}{i\omega}e^{-i\omega t}\left|\tilde{l}\right\rangle + \frac{e}{m}\left\langle \tilde{l}\right| \frac{e\vec{E}}{i\omega^*}e^{i\omega^* t}\left|\tilde{l}\right\rangle & (163)\\
&= -\frac{e}{m}\left[\langle l| + \sum_{l'\neq l}\frac{\langle l|\hat{U}|l'\rangle\langle l'| e^{i\omega^* t}}{\hbar(\omega_l-\omega_{l'}+\omega^*)}\right]e^{i\omega_l^* t}\hat{P}\left[\sum_{l'\neq l}\frac{|l'\rangle\langle l'|\hat{U}|l\rangle e^{-i\omega t}}{\hbar(\omega_l-\omega_{l'}+\omega)} + |l\rangle\right]e^{-i\omega_l t} & (164)\\
&\quad -\frac{e}{m}\frac{e\vec{E}}{i\omega}e^{-i\omega t}\left\langle \tilde{l}\middle|\tilde{l}\right\rangle + \frac{e}{m}\frac{e\vec{E}}{i\omega^*}e^{i\omega^* t}\left\langle \tilde{l}\middle|\tilde{l}\right\rangle & (165)\\
&= -\frac{e}{m}\langle l|\hat{P}|l\rangle - \left[\frac{e^2\vec{E}}{mi\omega}e^{-i\omega t} - \frac{e^2\vec{E}}{mi\omega^*}e^{i\omega^* t}\right]\left\langle \tilde{l}\middle|\tilde{l}\right\rangle & (166)\\
&\quad -\frac{e}{m}\sum_{l'\neq l}\frac{\langle l|\hat{U}^*|l'\rangle\langle l'|\hat{P}|l\rangle e^{i\omega^* t}}{\hbar(\omega_l-\omega_{l'}+\omega^*)} - \frac{e}{m}\sum_{l'\neq l}\frac{\langle l|\hat{P}|l'\rangle\langle l'|\hat{U}|l\rangle e^{-i\omega t}}{\hbar(\omega_l-\omega_{l'}+\omega)} & (167)\\
&\quad -\frac{e}{m}\sum_{l'\neq l}\frac{\langle l|\hat{U}|l'\rangle\langle l'|\hat{P}|l\rangle e^{-i\omega t}}{\hbar(\omega_l-\omega_{l'}-\omega)} - \frac{e}{m}\sum_{l'\neq l}\frac{\langle l|\hat{P}|l'\rangle\langle l'|\hat{U}^*|l\rangle e^{i\omega^* t}}{\hbar(\omega_l-\omega_{l'}-\omega^*)} & (168)\\
&= -\frac{e}{m}\langle l|\hat{P}|l\rangle - \left[\frac{e^2\vec{E}}{mi\omega}e^{-i\omega t} - \frac{e^2\vec{E}}{mi\omega^*}e^{i\omega^* t}\right]\left\langle \tilde{l}\middle|\tilde{l}\right\rangle & (169)\\
&\quad +\frac{e^2}{m^2 i\omega^*}\sum_{l'\neq l}\frac{\langle l|\vec{E}\cdot\hat{P}|l'\rangle\langle l'|\hat{P}|l\rangle e^{i\omega^* t}}{\hbar(\omega_l-\omega_{l'}+\omega^*)} - \frac{e^2}{m^2 i\omega}\sum_{l'\neq l}\frac{\langle l|\hat{P}|l'\rangle\langle l'|\vec{E}\cdot\hat{P}|l\rangle e^{-i\omega t}}{\hbar(\omega_l-\omega_{l'}+\omega)} & (170)\\
&\quad -\frac{e^2}{m^2 i\omega}\sum_{l'\neq l}\frac{\langle l|\vec{E}\cdot\hat{P}|l'\rangle\langle l'|\hat{P}|l\rangle e^{-i\omega t}}{\hbar(\omega_l-\omega_{l'}-\omega)} + \frac{e^2}{m^2 i\omega^*}\sum_{l'\neq l}\frac{\langle l|\hat{P}|l'\rangle\langle l'|\vec{E}\cdot\hat{P}|l\rangle e^{i\omega^* t}}{\hbar(\omega_l-\omega_{l'}-\omega^*)} & (171)\\
&= -\frac{e}{m}\langle l|\hat{P}|l\rangle - \left[\frac{e^2\vec{E}}{mi\omega}e^{-i\omega t} - \frac{e^2\vec{E}}{mi\omega^*}e^{i\omega^* t}\right]\left\langle \tilde{l}\middle|\tilde{l}\right\rangle & (172)\\
&\quad -\frac{e^2}{i\hbar m^2}\sum_{l'\neq l}\langle l|\hat{P}|l'\rangle\langle l'|\vec{E}\cdot\hat{P}|l\rangle\left[\frac{e^{-i\omega t}}{\omega(\omega_l-\omega_{l'}+\omega)} - \frac{e^{i\omega^* t}}{\omega^*(\omega_l-\omega_{l'}-\omega^*)}\right] & (173)\\
&\quad -\frac{e^2}{i\hbar m^2}\sum_{l'\neq l}\langle l'|\vec{E}\cdot\hat{P}|l\rangle\langle l|\hat{P}|l'\rangle\left[\frac{e^{-i\omega t}}{\omega(\omega_l-\omega_{l'}-\omega)} - \frac{e^{i\omega^* t}}{\omega^*(\omega_l-\omega_{l'}+\omega^*)}\right] & (174)
\end{aligned}$$

Because of $\vec{J} = \sigma\vec{E}$, the conductivity tensor $\sigma_{\alpha\beta}$ can be obtained as

$$\sigma_{\alpha\beta}(\omega) = \frac{-e^2}{im\omega V}\sum_l\left[f_l\delta_{\alpha\beta} + \sum_{l'}\frac{f_l}{\hbar m}\left(\frac{\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{\omega_l-\omega_{l'}+\omega} + \frac{\langle l|\hat{P}_\beta|l'\rangle\langle l'|\hat{P}_\alpha|l\rangle}{\omega_l^*-\omega_{l'}^*-\omega}\right)\right] \tag{175}$$

where f is the occupation numbers at state l and V is the volume. This equation can be simplified if ω_l are real. After exchanging l and l' in the second term, as below,

$$\sigma_{\alpha\beta}(\omega) = \frac{-e^2}{im\omega V}\left[\sum_l f_l\delta_{\alpha\beta} + \sum_l\sum_{l'}\frac{f_l}{\hbar m}\left(\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_l-\omega_{l'}+\omega} + \frac{\langle l|\,\hat{P}_\beta\,|l'\rangle\,\langle l'|\,\hat{P}_\alpha\,|l\rangle}{\omega_l^*-\omega_{l'}^*-\omega}\right)\right] \tag{176}$$

$$= \frac{-e^2}{im\omega V}\left[\sum_l f_l\delta_{\alpha\beta} + \sum_l\sum_{l'}\frac{f_l}{\hbar m}\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_l-\omega_{l'}+\omega} + \sum_l\sum_{l'}\frac{f_l}{\hbar m}\frac{\langle l|\,\hat{P}_\beta\,|l'\rangle\,\langle l'|\,\hat{P}_\alpha\,|l\rangle}{\omega_l^*-\omega_{l'}^*-\omega}\right] \tag{177}$$

$$= \frac{-e^2}{im\omega V}\left[\sum_l f_l\delta_{\alpha\beta} + \sum_l\sum_{l'}\frac{f_l}{\hbar m}\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_l-\omega_{l'}+\omega} + \sum_{l'}\sum_{l}\frac{f_{l'}}{\hbar m}\frac{\langle l'|\,\hat{P}_\beta\,|l\rangle\,\langle l|\,\hat{P}_\alpha\,|l'\rangle}{\omega_{l'}^*-\omega_{l}^*-\omega}\right] \tag{178}$$

$$= \frac{-e^2}{im\omega V}\left[\sum_l f_l\delta_{\alpha\beta} + \sum_l\sum_{l'}\frac{f_l}{\hbar m}\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_l-\omega_{l'}+\omega} - \sum_{l'}\sum_{l}\frac{f_{l'}}{\hbar m}\frac{\langle l'|\,\hat{P}_\beta\,|l\rangle\,\langle l|\,\hat{P}_\alpha\,|l'\rangle}{-\omega_{l'}^*+\omega_{l}^*+\omega}\right] \tag{179}$$

$$= \frac{-e^2}{im\omega V}\left[\sum_l f_l\delta_{\alpha\beta} + \sum_l\sum_{l'}\frac{f_l}{\hbar m}\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_l-\omega_{l'}+\omega} - \sum_{l'}\sum_{l}\frac{f_{l'}}{\hbar m}\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_{l}^*-\omega_{l'}^*+\omega}\right] \tag{180}$$

$$= \frac{-e^2}{im\omega V}\left[\sum_l f_l\delta_{\alpha\beta} + \sum_l\sum_{l'}\frac{f_l-f_{l'}}{\hbar m}\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_l-\omega_{l'}+\omega}\right] \tag{181}$$

and adding a small imaginary part $i\eta$ to complex ω, one can get the Eq. (182).

$$\sigma_{\alpha\beta}(\omega) = \frac{-e^2}{im\omega V}\left[\sum_l f_l\delta_{\alpha\beta} + \sum_l\sum_{l'}\frac{f_l-f_{l'}}{\hbar m}\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_l-\omega_{l'}+\omega+i\eta}\right] \tag{182}$$

$$= \frac{ie^2}{m\omega V}\left[\sum_l f_l\delta_{\alpha\beta} + \sum_l\sum_{l'}\frac{f_l-f_{l'}}{\hbar m}\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_l-\omega_{l'}+\omega+i\eta}\right] \tag{183}$$

In atomic unit, $\hbar = 1$, $m = 1$, and $e = 1$. Also, conductivity tensor can be written as

$$\sigma_{\alpha\beta}(\omega) = \frac{i}{\omega V}\left[\sum_l f_l\delta_{\alpha\beta} + \sum_l\sum_{l'}(f_l-f_{l'})\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\omega_l-\omega_{l'}+\omega+i\eta}\right] \tag{184}$$

Following the f sum rule (i.e. (185)) and the Eq. (187),

$$\frac{1}{\Omega}\sum_l\sum_{l'}(f_\mu-f_\nu)\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\epsilon_{\mu\nu}} = -m\rho\delta_{\alpha\beta} = -\frac{m\sum_l f_l\delta_{\alpha\beta}}{V} \tag{185}$$

$$\sum_l f_l\delta_{\alpha\beta} = -\sum_l\sum_{l'}\frac{(f_\mu-f_\nu)}{m}\frac{\langle l|\,\hat{P}_\alpha\,|l'\rangle\,\langle l'|\,\hat{P}_\beta\,|l\rangle}{\epsilon_{\mu\nu}} \tag{186}$$

$$\frac{1}{\epsilon_{\mu\nu}\pm\omega} = \frac{1}{\epsilon_{\mu\nu}}(1\mp\frac{\omega}{\epsilon_{\mu\nu}\pm\omega}) \tag{187}$$

Prove the Eq. (187)*:*

(1) For $1/(\epsilon+\omega)$:

$$\frac{1}{\epsilon}\left(1-\frac{\omega}{\epsilon+\omega}\right) = \frac{1}{\epsilon}\left(\frac{\epsilon+\omega}{\epsilon+\omega}-\frac{\omega}{\epsilon\pm\omega}\right) = \frac{1}{\epsilon}\left(\frac{\epsilon}{\epsilon+\omega}\right) = \frac{1}{\epsilon+\omega} \tag{188}$$

(2) For $1/(\epsilon - \omega)$:

$$\frac{1}{\epsilon}\left(1+\frac{\omega}{\epsilon-\omega}\right)=\frac{1}{\epsilon}\left(\frac{\epsilon-\omega}{\epsilon-\omega}+\frac{\omega}{\epsilon\pm\omega}\right)=\frac{1}{\epsilon}\left(\frac{\epsilon}{\epsilon-\omega}\right)=\frac{1}{\epsilon-\omega} \tag{189}$$

Therefore, one can rewrite the Eq. (182).

$$\begin{aligned}
\sigma_{\alpha\beta}(\omega) &= \frac{ie^2}{m\omega V}\left[\sum_l f_l\delta_{\alpha\beta}+\sum_l\sum_{l'}\frac{f_l-f_{l'}}{\hbar m}\frac{\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{\omega_l-\omega_{l'}+\omega+i\eta}\right] && (190)\\
&= \frac{ie^2}{m\omega V}\left[-\sum_l\sum_{l'}\frac{f_l-f_{l'}}{\hbar m}\frac{\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{\omega_l-\omega_{l'}}+\sum_l\sum_{l'}\frac{f_l-f_{l'}}{\hbar m}\frac{\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{\omega_l-\omega_{l'}+\omega+i\eta}\right] && (191)\\
&= \frac{i}{\omega V}\left[-\sum_l\sum_{l'}(f_l-f_{l'})\frac{\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{\omega_l-\omega_{l'}}+\sum_l\sum_{l'}(f_l-f_{l'})\frac{\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{\omega_l-\omega_{l'}+\omega+i\eta}\right] && (192)\\
&= \frac{i}{\omega V}\left[-\sum_l\sum_{l'}(f_l-f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle\left(\frac{1}{\omega_l-\omega_{l'}}-\frac{1}{\omega_l-\omega_{l'}+\omega+i\eta}\right)\right] && (193)
\end{aligned}$$

One can assume $\omega_l - \omega_{l'} = \epsilon_{ll'}$ and $\omega + i\eta = \omega'$. Then, one can get

$$\begin{aligned}
\sigma_{\alpha\beta}(\omega) &= \frac{i}{\omega V}\left[-\sum_l\sum_{l'}(f_l-f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle\left(\frac{1}{\epsilon_{ll'}}-\frac{1}{\epsilon_{ll'}+\omega'}\right)\right] && (194)\\
&= \frac{-i}{\omega V}\left[\sum_l\sum_{l'}(f_l-f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle\left(\frac{1}{\epsilon_{ll'}}-\frac{1}{\epsilon_{ll'}}+\frac{\omega'}{\epsilon_{ll'}(\epsilon_{ll'}+\omega')}\right)\right] && (195)\\
&= \frac{-i}{\omega V}\left[\sum_l\sum_{l'}(f_l-f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle\frac{\omega'}{\epsilon_{ll'}(\epsilon_{ll'}+\omega')}\right] && (196)\\
&= \frac{-i}{\omega V}\left[\sum_l\sum_{l'}(f_l-f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle\frac{\omega+i\eta}{(\omega_l-\omega_{l'})(\omega_l-\omega_{l'}+\omega+i\eta)}\right] && (197)\\
&= \frac{-i}{V}\frac{\omega+i\eta}{\omega}\left[\sum_l\sum_{l'}(f_l-f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle\frac{1}{(\omega_l-\omega_{l'})(\omega_l-\omega_{l'}+\omega+i\eta)}\right] && (198)\\
&= \frac{-i}{V}\left[\sum_l\sum_{l'}\frac{(f_l-f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{(\omega_l-\omega_{l'})(\omega_l-\omega_{l'}+\omega+i\eta)}\right] && (199)\\
\sigma_{\alpha\beta}(\omega) &= N\frac{-ie^2}{\hbar m_e^2 V}\left[\sum_l\sum_{l'}\frac{(f_l-f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{(\omega_l-\omega_{l'})(\omega_l-\omega_{l'}+\omega+i\eta)}\right]. && (200)
\end{aligned}$$

where $N = 4.59984823346488111 \times 10^6(\Omega \cdot m)^{-1}$ and the unit of $\langle l|\hat{P}_\alpha|l'\rangle$ is $\hbar/a_0$. Unit of conductivity tensor is

$$\frac{C^2}{J\cdot s\cdot kg^2\cdot m^3}\frac{kg^2\cdot m^2\cdot s^{-2}}{s^{-1}\cdot s^{-1}}=\frac{C^2}{J\cdot s\cdot m}=\frac{1}{\Omega m}=\frac{\mho}{m}=\frac{S}{m} \tag{201}$$

where $e = 1.60217656535 \times 10^{-19}(C)$, $\hbar = 1.05457162853 \times 10^{-34}(eV \cdot s)$, $m_e = 9.109382914 \times 10^{-31}(kg)$, $\hbar/a_0 = 1.992851882 \times 10^{-24}(kg \cdot m \cdot s^{-1})$,$1Bohr = 0.529177249 \times 10^{-10}(meter)$, $\omega = 2.41888432650516 \times 10^{-17}(s^{-1})$, Ω is Ohm, $\mho$ $(= 1/\Omega)$ is Mho, and S is Siemens.

Based on Dirac-delta function

$$\frac{1}{a \pm ib} = P\left[\frac{1}{a}\right] \mp i\pi\delta(a), \tag{202}$$

where a=$\omega_l - \omega_{l'} + \omega$ and b=η , the conductivity tensor $\sigma_{\alpha\beta}(\omega) = \sigma^1_{\alpha\beta}(\omega) + \sigma^2_{\alpha\beta}(\omega)$ can be rewritten as

$$\sigma^1_{\alpha\beta}(\omega) = \frac{-\pi}{V}\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{(\omega_l - \omega_{l'})}\delta(\omega_l - \omega_{l'} + \omega)\right] \tag{203}$$

$$\sigma^2_{\alpha\beta}(\omega) = \frac{-i}{V}P\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{(\omega_l - \omega_{l'})(\omega_l - \omega_{l'} + \omega)}\right] \tag{204}$$

One can treat this conductivity tensor by replacing Dirac-delta function with a Lorentian function.

$$\delta(a) = \lim_{b\to 0}\frac{1}{\pi}\frac{b}{(a - a_0)^2 + b^2} \tag{205}$$

where a_0 is the centered point of the Lorentian function. In order to avoid divergence at $\omega = 0$, therefore, the first part of conductivity tensor becomes

$$\sigma^1_{\alpha\beta}(\omega) = \frac{-\pi}{V}\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{1}{\pi}\frac{\eta}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{206}$$

$$= \frac{-1}{V}\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\eta}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{207}$$

$$= \frac{-1}{V}\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})(p^{Re}_\alpha + ip^{Im}_\alpha)(p^{Re}_\beta + ip^{Im}_\beta)}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\eta}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{208}$$

$$= \frac{-1}{V}\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})\left[p^{Re}_\alpha p^{Re}_\beta - p^{Im}_\alpha p^{Im}_\beta\right] + i\left[p^{Re}_\alpha p^{Im}_\beta + p^{Re}_\beta p^{Im}_\alpha\right]}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\eta}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{209}$$

$$= \frac{-1}{V}\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})\left[p^{Re}_\alpha p^{Re}_\beta - p^{Im}_\alpha p^{Im}_\beta\right]}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\eta}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{210}$$

$$- i\frac{1}{V}\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})\left[p^{Re}_\alpha p^{Im}_\beta + p^{Re}_\beta p^{Im}_\alpha\right]}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\eta}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right], \tag{211}$$

and the second part of conductivity tensor becomes

$$\sigma^2_{\alpha\beta}(\omega) = \frac{-i}{V}P\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{(\omega_l - \omega_{l'})(\omega_l - \omega_{l'} + \omega)}\right] \tag{212}$$

$$= \frac{-i}{V}\left[\sum_l \sum_{l'} \frac{(f_l - f_{l'})\langle l|\hat{P}_\alpha|l'\rangle\langle l'|\hat{P}_\beta|l\rangle}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\omega_l - \omega_{l'} + \omega}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{213}$$

$$\sigma^2_{\alpha\beta}(\omega) = \frac{-i}{V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left(\left[p^{Re}_\alpha p^{Re}_\beta - p^{Im}_\alpha p^{Im}_\beta\right] + i\left[p^{Re}_\alpha p^{Im}_\beta + p^{Re}_\beta p^{Im}_\alpha\right]\right)}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\omega_l - \omega_{l'} + \omega}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{214}$$

$$= \frac{-i}{V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left(\left[p^{Re}_\alpha p^{Re}_\beta - p^{Im}_\alpha p^{Im}_\beta\right]\right)}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\omega_l - \omega_{l'} + \omega}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{215}$$

$$+ \frac{-i}{V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left(i\left[p^{Re}_\alpha p^{Im}_\beta + p^{Re}_\beta p^{Im}_\alpha\right]\right)}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\omega_l - \omega_{l'} + \omega}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{216}$$

$$= \frac{1}{V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left(\left[p^{Re}_\alpha p^{Im}_\beta + p^{Re}_\beta p^{Im}_\alpha\right]\right)}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\omega_l - \omega_{l'} + \omega}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{217}$$

$$- \frac{i}{V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left(\left[p^{Re}_\alpha p^{Re}_\beta - p^{Im}_\alpha p^{Im}_\beta\right]\right)}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\omega_l - \omega_{l'} + \omega}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right]. \tag{218}$$

where $\langle l|\,\hat{P}_\alpha\,|l'\rangle = p^{Re}_\alpha + ip^{Im}_\alpha$, $\langle l'|\,\hat{P}_\beta\,|l\rangle = p^{Re}_\beta + ip^{Im}_\beta$. Therefore, the real part and imaginery part of conductivity tensor $\sigma_{\alpha\beta}(\omega) = \sigma^{Re}_{\alpha\beta}(\omega) + i\sigma^{Im}_{\alpha\beta}(\omega)$ can be expressed as

$$\sigma^{Re}_{\alpha\beta}(\omega) = \frac{-1}{V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left[p^{Re}_\alpha p^{Re}_\beta - p^{Im}_\alpha p^{Im}_\beta\right]}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\eta}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{219}$$

$$+ \frac{1}{V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left(\left[p^{Re}_\alpha p^{Im}_\beta + p^{Re}_\beta p^{Im}_\alpha\right]\right)}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\omega_l - \omega_{l'} + \omega}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{220}$$

$$\sigma^{Im}_{\alpha\beta}(\omega) = -\frac{1}{V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left[p^{Re}_\alpha p^{Im}_\beta + p^{Re}_\beta p^{Im}_\alpha\right]}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\eta}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{221}$$

$$- \frac{1}{V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left(\left[p^{Re}_\alpha p^{Re}_\beta - p^{Im}_\alpha p^{Im}_\beta\right]\right)}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\omega_l - \omega_{l'} + \omega}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{222}$$

Based on the equation of dielectric function , $\epsilon_{\alpha\beta}(\omega) = \delta_{\alpha\beta} + \frac{i}{\epsilon_0\omega}\sigma_{\alpha\beta}(\omega)$, dielectric function can be evaluated as

$$\epsilon_{\alpha\beta}(\omega) = \epsilon^{Re}_{\alpha\beta}(\omega) + \epsilon^{Im}_{\alpha\beta}(\omega) \tag{223}$$

$$= \delta_{\alpha\beta} + \frac{i}{\epsilon_0\omega}\left[\sigma^{Re}_{\alpha\beta}(\omega) + i\sigma^{Im}_{\alpha\beta}(\omega)\right]. \tag{224}$$

Therefore,

$$\epsilon^{Re}_{\alpha\beta}(\omega) = \delta_{\alpha\beta} - \frac{1}{\epsilon_0\omega}\sigma^{Im}_{\alpha\beta}(\omega) \tag{225}$$

$$= \delta_{\alpha\beta} \tag{226}$$

$$+ \frac{1}{\epsilon_0\omega V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left[p^{Re}_\alpha p^{Im}_\beta + p^{Re}_\beta p^{Im}_\alpha\right]}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\eta}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{227}$$

$$+ \frac{1}{\epsilon_0\omega V}\left[\sum_l\sum_{l'}\frac{(f_l - f_{l'})\left(\left[p^{Re}_\alpha p^{Re}_\beta - p^{Im}_\alpha p^{Im}_\beta\right]\right)}{(\omega_l - \omega_{l'})}\lim_{\eta\to 0}\frac{\omega_l - \omega_{l'} + \omega}{(\omega_l - \omega_{l'} + \omega)^2 + \eta^2}\right] \tag{228}$$

3.3.3 Momentum matrix elements

The first thing to calculate conductivity tensor is how to evaluate the momentum matrix elements.

$$\langle\psi_{k,\mu}(\mathbf{r})|\overrightarrow{p}|\psi_{k,\nu}(\mathbf{r})\rangle = -i\hbar\,\langle\psi_{k,\mu}(\mathbf{r})|\nabla|\psi_{k,\nu}(\mathbf{r})\rangle \tag{229}$$

$$= -i\hbar\frac{1}{\sqrt{N}}\sum_{m}\sum_{i\alpha} c^{*k}_{i,\alpha,\mu}e^{ik\cdot R_m}\frac{1}{\sqrt{N}}\sum_{n}\sum_{j\beta} c^{k}_{j,\beta,\nu}e^{-ik\cdot R_n}\,\langle\phi_{i\alpha}(\mathbf{r})|\nabla|\phi_{j\beta}(\mathbf{r})\rangle \tag{230}$$

$$= \frac{-i\hbar}{N}\sum_{m}\sum_{i\alpha} c^{*k}_{i,\alpha,\mu}e^{ik\cdot R_m}\sum_{n}\sum_{j\beta} c^{k}_{j,\beta,\nu}e^{-ik\cdot R_n}\,\langle\phi_{i\alpha}(\mathbf{r})|\nabla|\phi_{j\beta}(\mathbf{r})\rangle \tag{231}$$

$$= \frac{-i\hbar}{N}\sum_{mn}\sum_{i\alpha} c^{*k}_{i,\alpha,\mu}\sum_{j\beta} c^{k}_{j,\beta,\nu}e^{-ik\cdot(R_n-R_m)}\,\langle\phi_{i\alpha}(\mathbf{r})|\nabla|\phi_{j\beta}(\mathbf{r})\rangle \tag{232}$$

The occupied and unoccupied states in one do-loop can be partitioned into processors and each processor deals with parts of states to calculate MME on simultaneously.

$$\langle\psi_{k,\mu}(\mathbf{r})|\overrightarrow{p}|\psi_{k,\nu}(\mathbf{r})\rangle = \frac{-i\hbar}{N}\sum_{m,n}\sum_{i,\alpha} c^{*k}_{i,\alpha,\mu}\sum_{j,\beta} c^{k}_{j,\beta,\nu}e^{-ik\cdot(R_n-R_m)}\,\langle\phi_{i,\alpha}(\mathbf{r})|\nabla|\phi_{j,\beta}(\mathbf{r})\rangle \tag{233}$$

$$\langle\psi_{k,\mu}(\mathbf{r})|\overrightarrow{p}|\psi_{k,\nu}(\mathbf{r})\rangle\;\langle\psi_{k,\nu}(\mathbf{r})|\overrightarrow{p}|\psi_{k,\mu}(\mathbf{r})\rangle = \langle\psi_{k,\mu}(\mathbf{r})|\overrightarrow{p}|\psi_{k,\nu}(\mathbf{r})\rangle\;\langle\psi_{k,\mu}(\mathbf{r})|\overrightarrow{p}|\psi_{k,\nu}(\mathbf{r})\rangle^* = \tag{234}$$

$$\frac{-i\hbar}{N}\sum_{m,n}\sum_{i,\alpha} c^{*k}_{i,\alpha,\mu}\sum_{j,\beta} c^{k}_{j,\beta,\nu}e^{-ik\cdot(R_n-R_m)}\,\langle\phi_{i,\alpha}(\mathbf{r})|\nabla|\phi_{j,\beta}(\mathbf{r})\rangle \tag{235}$$

$$\cdot\frac{i\hbar}{N}\sum_{m',n'}\sum_{i',\alpha'} c^{*k}_{i',\alpha',\mu}\sum_{j',\beta'} c^{k}_{j',\beta',\nu}e^{ik\cdot(R_{n'}-R_{m'})}\,\langle\phi_{i',\alpha'}(\mathbf{r})|\nabla|\phi_{j',\beta'}(\mathbf{r})\rangle \tag{236}$$

where k is an index of k-points, μ and ν are indexes of occupied and unoccupied states, m and n are indexes of unit cells, i and j are indexes of atom i and the first neighboring atom j, and α and β are orbital indexes of atom i and j.

$$\langle\phi_{i\alpha}(\mathbf{r})|\nabla|\phi_{j\beta}(\mathbf{r})\rangle = \int dr^3\phi^*_{i\alpha}(\mathbf{r})\nabla\phi_{j\beta}(\mathbf{r}) \tag{237}$$

$$= \int dr^3\left[\left(\frac{1}{\sqrt{(2\pi)}}\right)^3\int dk^3\tilde{\phi}^*_{i\alpha}(\mathbf{k})e^{-i\mathbf{k}\cdot\mathbf{r}}\right]\nabla\left[\left(\frac{1}{\sqrt{(2\pi)}}\right)^3\int dk'^3\tilde{\phi}_{j\beta}(\mathbf{k'})e^{i\mathbf{k'}\cdot(\mathbf{r}-\tau)}\right] \tag{238}$$

$$= (2\pi)^{-3}\int dr^3\left[\int dk^3\tilde{R}^*_{pl}(k)\tilde{Y}^*_{lm}(\hat{\mathbf{k}})e^{-i\mathbf{k}\cdot\mathbf{r}}\right]\nabla\left[\int dk'^3\tilde{R}_{p'l'}(k')\tilde{Y}_{l'm'}(\hat{\mathbf{k}}')e^{i\mathbf{k'}\cdot(\mathbf{r}-\tau)}\right] \tag{239}$$

$$= (2\pi)^{-3}\int dk^3\int dk'^3e^{-i\mathbf{k'}\cdot\tau}\tilde{R}^*_{pl}(k)\tilde{Y}^*_{lm}(\hat{\mathbf{k}})i\overrightarrow{\mathbf{k'}}\tilde{R}_{p'l'}(k')\tilde{Y}_{l'm'}(\hat{\mathbf{k}}')\int dr^3e^{i(\mathbf{k'}-\mathbf{k})\cdot\mathbf{r}} \tag{240}$$

$$= i(2\pi)^{-3}\int dk^3\int dk'^3e^{-i\mathbf{k'}\cdot\tau}\tilde{R}^*_{pl}(k)Y^*_{lm}(\hat{\mathbf{k}})\overrightarrow{\mathbf{k'}}\tilde{R}_{p'l'}(k')Y_{l'm'}(\hat{\mathbf{k}}')(2\pi)^3\delta(\mathbf{k'}-\mathbf{k}) \tag{241}$$

$$= i\int dk^3e^{-i\mathbf{k}\cdot\tau}\tilde{R}^*_{pl}(k)Y^*_{lm}(\hat{\mathbf{k}})\overrightarrow{\mathbf{k}}\tilde{R}_{p'l'}(k)Y_{l'm'}(\hat{\mathbf{k}}) \tag{242}$$

$$= i\int dk^3\left[4\pi\sum_{L=0}^{\infty}\sum_{M=-L}^{L}(-1)^Lj_L(k|\tau|)Y_{LM}(\hat{\mathbf{k}})Y^*_{LM}(\hat{\tau})\right]\tilde{R}^*_{pl}(k)Y^*_{lm}(\hat{\mathbf{k}})\overrightarrow{\mathbf{k}}\tilde{R}_{p'l'}(k)Y_{l'm'}(\hat{\mathbf{k}}) \tag{243}$$

$$= i4\pi \sum_{L=0}^{\infty} \sum_{M=-L}^{L} (-1)^L Y_{LM}^*(\hat{\tau}) \int dk^3 Y_{lm}^*(\hat{\mathbf{k}}) Y_{l'm'}(\hat{\mathbf{k}}) Y_{LM}(\hat{\mathbf{k}}) \vec{\mathbf{k}} j_L(k|\tau|) \tilde{R}_{pl}^*(k) \tilde{R}_{p'l'}(k) \quad (244)$$

$$= i4\pi \sum_{L=0}^{\infty} \sum_{M=-L}^{L} (-1)^L Y_{LM}^*(\hat{\tau}) \int d\phi \int d\theta \sin\theta \int dk k^2 Y_{lm}^*(\hat{\mathbf{k}}) Y_{l'm'}(\hat{\mathbf{k}}) Y_{LM}(\hat{\mathbf{k}}) \vec{\mathbf{k}}(k,\theta,\phi) j_L(k|\tau|) \tilde{R}_{pl}^*(k) \tilde{R}_{p'l'}(k) \quad (245)$$

For x/y/z direction:

$$\langle \phi_{k,\mu}(\mathbf{r}) | \frac{\partial}{\partial x} | \phi_{k,\nu}(\mathbf{r}) \rangle = 4\pi \sum_{L=0}^{\infty} \sum_{M=-L}^{L} (-1)^L Y_{LM}^*(\hat{\tau}) C_{l(-m),l'm',LM}^{x} \int dk k^3 j_L(k|\tau|) \tilde{R}_{pl}^*(k) \tilde{R}_{p'l'}(k) \quad (246)$$

$$\langle \phi_{k,\mu}(\mathbf{r}) | \frac{\partial}{\partial y} | \phi_{k,\nu}(\mathbf{r}) \rangle = 4\pi \sum_{L=0}^{\infty} \sum_{M=-L}^{L} (-1)^L Y_{LM}^*(\hat{\tau}) C_{l(-m),l'm',LM}^{y} \int dk k^3 j_L(k|\tau|) \tilde{R}_{pl}^*(k) \tilde{R}_{p'l'}(k) \quad (247)$$

$$\langle \phi_{k,\mu}(\mathbf{r}) | \frac{\partial}{\partial z} | \phi_{k,\nu}(\mathbf{r}) \rangle = 4\pi \sum_{L=0}^{\infty} \sum_{M=-L}^{L} (-1)^L Y_{LM}^*(\hat{\tau}) C_{l(-m),l'm',LM}^{z} \int dk k^3 j_L(k|\tau|) \tilde{R}_{pl}^*(k) \tilde{R}_{p'l'}(k) \quad (248)$$

where

$$C_{l(-m),l'm',LM}^{x} = \int_0^{2\pi} \int_0^{\pi} d\phi d\theta \sin\theta \, [Y_{lm}^*(\theta,\phi) Y_{l'm'}(\theta,\phi) Y_{LM}(\theta,\phi)] \sin\theta \cos\phi \quad (249)$$

$$C_{l(-m),l'm',LM}^{y} = \int_0^{2\pi} \int_0^{\pi} d\phi d\theta \sin\theta \, [Y_{lm}^*(\theta,\phi) Y_{l'm'}(\theta,\phi) Y_{LM}(\theta,\phi)] \sin\theta \sin\phi \quad (250)$$

$$C_{l(-m),l'm',LM}^{z} = \int_0^{2\pi} \int_0^{\pi} d\phi d\theta \sin\theta \, [Y_{lm}^*(\theta,\phi) Y_{l'm'}(\theta,\phi) Y_{LM}(\theta,\phi)] \cos\theta \quad (251)$$

Approach 1:
Here, the equations from (249)-(251) can be solved by Mathemetica to get their individual analytical solution - a constant number - at each l_1,m_1,l_2, m_2, L, and M. Furthermore, due to symmtery of two spherical harmonics function, e.g. spherical harmonics functions exchange l and m with each other, the non-zero terms are the same in x and z directions without sign changed and in y direction with sign changed, respectively. And, in both of x and y directions, values of non-zero terms are the same (except sign changed if $m' < m'' + M$).

Approach 2:
Based on

$$Y_{1,0}(\theta,\phi) = \frac{1}{2}\sqrt{\frac{3}{\pi}} \cos\theta \quad (252)$$

$$Y_{1,\pm1}(\theta,\phi) = \mp\frac{1}{2}\sqrt{\frac{3}{2\pi}} \sin\theta e^{\pm i\phi} = \mp\frac{1}{2}\sqrt{\frac{3}{2\pi}} \sin\theta \, (\cos\phi \pm i \sin\phi) \, , \quad (253)$$

one can easily get the relations between spherical harmonics functions and corresponding $\sin\theta\cos\phi$, $\sin\theta\sin\phi$, and $\cos\theta$, i.e.

$$\sin\theta\cos\phi = \sqrt{\frac{2\pi}{3}}\left[Y_{1,1}(\theta,\phi) + Y_{1,-1}(\theta,\phi)\right] \tag{254}$$

$$\sin\theta\sin\phi = i\sqrt{\frac{2\pi}{3}}\left[Y_{1,1}(\theta,\phi) - Y_{1,-1}(\theta,\phi)\right] \tag{255}$$

$$\cos\theta = 2\sqrt{\frac{\pi}{3}}Y_{1,0}(\theta,\phi) \tag{256}$$

After these three equations are put back to (249) (251), Gaunt coefficients can be expressed as below.

$$C^x_{l(-m),l'm',LM} = \sqrt{\frac{2\pi}{3}}\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta \left[Y^*_{lm}(\theta,\phi)Y_{l'm'}(\theta,\phi)Y_{LM}(\theta,\phi)\right]\left[Y_{1,1}(\theta,\phi) + Y_{1,-1}(\theta,\phi)\right] \tag{257}$$

$$C^y_{l(-m),l'm',LM} = i\sqrt{\frac{2\pi}{3}}\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta \left[Y^*_{lm}(\theta,\phi)Y_{l'm'}(\theta,\phi)Y_{LM}(\theta,\phi)\right]\left[Y_{1,1}(\theta,\phi) - Y_{1,-1}(\theta,\phi)\right] \tag{258}$$

$$C^z_{l(-m),l'm',LM} = 2\sqrt{\frac{\pi}{3}}\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta \left[Y^*_{lm}(\theta,\phi)Y_{l'm'}(\theta,\phi)Y_{LM}(\theta,\phi)\right]Y_{1,0}(\theta,\phi) \tag{259}$$

According to

$$Y^*_{l,m}(\theta,\phi) = (-1)^m Y_{l,-m}(\theta,\phi) \tag{260}$$

$$Y_{l_1,m_1}(\theta,\phi)Y_{l_2,m_2}(\theta,\phi) = \sqrt{\frac{(2l_1+1)(2l_2+1)}{4\pi}}\sum_{L,M}\sqrt{2L+1}\begin{pmatrix} l_1 & l_2 & L \\ m_1 & m_2 & -M\end{pmatrix}\begin{pmatrix} l_1 & l_2 & L \\ 0 & 0 & 0\end{pmatrix}Y_{L,M}(\theta,\phi)(-1)^M \tag{261}$$

$$Y_{l_1,m_1}(\theta,\phi)Y_{l_2,m_2}(\theta,\phi) = \sqrt{\frac{(2l_1+1)(2l_2+1)}{4\pi}}\sum_{L,M}\sqrt{2L+1}\begin{pmatrix} l_1 & l_2 & L \\ m_1 & m_2 & M\end{pmatrix}\begin{pmatrix} l_1 & l_2 & L \\ 0 & 0 & 0\end{pmatrix}Y^*_{L,M}(\theta,\phi) \tag{262}$$

and

$$\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta Y_{l_1,m_1}(\theta,\phi)Y_{l_2,m_2}(\theta,\phi)Y_{l_3,m_3}(\theta,\phi)Y_{l_4,m_4}(\theta,\phi) \tag{263}$$

$$= \sqrt{\frac{(2l_1+1)(2l_2+1)}{4\pi}}\sum_{L,M}(-1)^M\sqrt{2L+1}\begin{pmatrix} l_1 & l_2 & L \\ m_1 & m_2 & -M\end{pmatrix}\begin{pmatrix} l_1 & l_2 & L \\ 0 & 0 & 0\end{pmatrix} \tag{264}$$

$$\times \int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta Y_{L,M}(\theta,\phi)Y_{l_3,m_3}(\theta,\phi)Y_{l_4,m_4}(\theta,\phi) \tag{265}$$

$$= \sqrt{\frac{(2l_1+1)(2l_2+1)}{4\pi}}\sum_{L,M}(-1)^M\sqrt{2L+1}\begin{pmatrix} l_1 & l_2 & L \\ m_1 & m_2 & -M\end{pmatrix}\begin{pmatrix} l_1 & l_2 & L \\ 0 & 0 & 0\end{pmatrix} \tag{266}$$

$$\times \sqrt{\frac{(2l_3+1)(2l_4+1)}{4\pi}}\sum_{L',M'}\sqrt{2L'+1}\begin{pmatrix} l_3 & l_4 & L' \\ m_3 & m_4 & M'\end{pmatrix}\begin{pmatrix} l_3 & l_4 & L' \\ 0 & 0 & 0\end{pmatrix} \tag{267}$$

$$\times \int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta Y_{L,M}(\theta,\phi)Y^*_{L',M'}(\theta,\phi) \tag{268}$$

$$= \frac{\sqrt{(2l_1+1)(2l_2+1)(2l_3+1)(2l_4+1)}}{4\pi} \tag{269}$$

$$\times \sum_{L,M} (-1)^M (2L+1) \begin{pmatrix} l_1 & l_2 & L \\ m_1 & m_2 & -M \end{pmatrix} \begin{pmatrix} l_1 & l_2 & L \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_3 & l_4 & L \\ m_3 & m_4 & M \end{pmatrix} \begin{pmatrix} l_3 & l_4 & L \\ 0 & 0 & 0 \end{pmatrix} \tag{270}$$

the equations from (257) ot (259) can be reorganized as below.
(1)

$$C^x_{l(-m),l'm',LM} = \sqrt{\frac{2\pi}{3}} \int_0^{2\pi} \int_0^{\pi} d\phi d\theta \sin\theta Y^*_{l,-m}(\theta,\phi) Y_{l',m'}(\theta,\phi) Y_{L,M}(\theta,\phi) \left[Y_{1,1}(\theta,\phi) + Y_{1,-1}(\theta,\phi)\right] \tag{271}$$

$$= \sqrt{\frac{2\pi}{3}} (-1)^m \int_0^{2\pi} \int_0^{\pi} d\phi d\theta \sin\theta Y_{l,-m}(\theta,\phi) Y_{l',m'}(\theta,\phi) Y_{L,M}(\theta,\phi) Y_{1,1}(\theta,\phi) \tag{272}$$

$$+ \sqrt{\frac{2\pi}{3}} (-1)^m \int_0^{2\pi} \int_0^{\pi} d\phi d\theta \sin\theta Y_{l,-m}(\theta,\phi) Y_{l',m'}(\theta,\phi) Y_{L,M}(\theta,\phi) Y_{1,-1}(\theta,\phi) \tag{273}$$

$$= \sqrt{\frac{2\pi}{3}} (-1)^m \frac{\sqrt{(2l+1)(2l'+1)(2L+1)(2\cdot 1+1)}}{4\pi} \tag{274}$$

$$\times \sum_{L',M'} (-1)^{M'} (2L'+1) \begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix} \begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ M & 1 & M' \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{275}$$

$$+ \sqrt{\frac{2\pi}{3}} (-1)^m \frac{\sqrt{(2l+1)(2l'+1)(2L+1)(2\cdot 1+1)}}{4\pi} \tag{276}$$

$$\times \sum_{L',M'} (-1)^{M'} (2L'+1) \begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix} \begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ M & -1 & M' \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{277}$$

$$= \sqrt{\frac{2\pi}{3}} (-1)^m \frac{\sqrt{(2l+1)(2l'+1)(2L+1)(2\cdot 1+1)}}{4\pi} \tag{278}$$

$$\times \sum_{L',M'} (-1)^{M'} (2L'+1) \begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix} \begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ M & 1 & M' \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{279}$$

$$+ \sqrt{\frac{2\pi}{3}} (-1)^m \frac{\sqrt{(2l+1)(2l'+1)(2L+1)(2\cdot 1+1)}}{4\pi} \tag{280}$$

$$\times \sum_{L',M'} (-1)^{M'} (2L'+1) \begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix} \begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ M & -1 & M' \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{281}$$

$$= (-1)^m \sqrt{\frac{(2l+1)(2l'+1)(2L+1)}{8\pi}} \tag{282}$$

$$\times \sum_{L',M'} (-1)^{M'} (2L'+1) \begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix} \begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{283}$$

$$\times \left[\begin{pmatrix} L & 1 & L' \\ M & 1 & M' \end{pmatrix} + \begin{pmatrix} L & 1 & L' \\ M & -1 & M' \end{pmatrix} \right] \tag{284}$$

(2)

$$C^i_{l(-m),l'm',LM} = i\sqrt{\frac{2\pi}{3}}\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta Y^*_{l,-m}(\theta,\phi)Y_{l',m'}(\theta,\phi)Y_{L,M}(\theta,\phi)\left[Y_{1,1}(\theta,\phi) - Y_{1,-1}(\theta,\phi)\right] \tag{285}$$

$$= i(-1)^m\sqrt{\frac{(2l+1)(2l'+1)(2L+1)}{8\pi}} \tag{286}$$

$$\times \sum_{L',M'}(-1)^{M'}(2L'+1)\begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix}\begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} L & 1 & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{287}$$

$$\times \left[\begin{pmatrix} L & 1 & L' \\ M & 1 & M' \end{pmatrix} - \begin{pmatrix} L & 1 & L' \\ M & -1 & M' \end{pmatrix}\right] \tag{288}$$

(3)

$$C^z_{l(-m),l'm',LM} = 2\sqrt{\frac{\pi}{3}}(-1)^m\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta(-1)^m Y_{l,-m}(\theta,\phi)Y_{l',m'}(\theta,\phi)Y_{L,M}(\theta,\phi)Y_{1,0}(\theta,\phi) \tag{289}$$

$$= 2\sqrt{\frac{\pi}{3}}(-1)^m\frac{\sqrt{(2l+1)(2l'+1)(2L+1)(2\cdot 1+1)}}{4\pi} \tag{290}$$

$$\times \sum_{L',M'}(-1)^{M'}(2L'+1)\begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix}\begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} L & 1 & L' \\ M & -1 & M' \end{pmatrix}\begin{pmatrix} L & 1 & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{291}$$

$$= (-1)^m\sqrt{\frac{(2l+1)(2l'+1)(2L+1)}{4\pi}} \tag{292}$$

$$\times \sum_{L',M'}(-1)^{M'}(2L'+1)\begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix}\begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} L & 1 & L' \\ M & 0 & M' \end{pmatrix}\begin{pmatrix} L & 1 & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{293}$$

or

$$C^z_{l(-m),l'm',LM} = 2\sqrt{\frac{\pi}{3}}\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta(-1)^m Y_{l,-m}(\theta,\phi)Y_{l',m'}(\theta,\phi)Y_{L,M}(\theta,\phi)Y_{1,0}(\theta,\phi) \tag{294}$$

$$= 2\sqrt{\frac{\pi}{3}}(-1)^m\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta\left[Y_{l,-m}(\theta,\phi)Y_{l',m'}(\theta,\phi)\right]Y_{L,M}(\theta,\phi)Y_{1,0}(\theta,\phi) \tag{295}$$

$$= \sqrt{\frac{(2l+1)(2l'+1)}{4\pi}}\sum_{L',M'}(-1)^{M'}\sqrt{2L'+1}\begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix}\begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{296}$$

$$\times 2\sqrt{\frac{\pi}{3}}(-1)^m\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta Y_{L',M'}(\theta,\phi)\left[Y_{L,M}(\theta,\phi)Y_{1,0}(\theta,\phi)\right] \tag{297}$$

$$= \sqrt{\frac{(2l+1)(2l'+1)}{4\pi}}\sum_{L',M'}(-1)^{M'}\sqrt{2L'+1}\begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix}\begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{298}$$

$$\times \sqrt{\frac{(2L+1)(2\cdot 1+1)}{4\pi}}\sum_{L'',M''}\sqrt{2L''+1}\begin{pmatrix} L & 1 & L'' \\ M & 0 & M'' \end{pmatrix}\begin{pmatrix} L & 1 & L'' \\ 0 & 0 & 0 \end{pmatrix} \tag{299}$$

$$\times 2\sqrt{\frac{\pi}{3}}(-1)^m\int_0^{2\pi}\int_0^{\pi} d\phi d\theta \sin\theta Y_{L',M'}(\theta,\phi)Y^*_{L'',M''}(\theta,\phi) \tag{300}$$

or

$$= (-1)^m \sqrt{\frac{(2l+1)(2l'+1)}{4\pi}} \sum_{L',M'} (-1)^{M'} \sqrt{2L'+1} \begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix} \begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{301}$$

$$\times \sqrt{2L+1} \sum_{L'',M''} \sqrt{2L''+1} \begin{pmatrix} L & 1 & L'' \\ M & 0 & M'' \end{pmatrix} \begin{pmatrix} L & 1 & L'' \\ 0 & 0 & 0 \end{pmatrix} \delta_{L',L''}\delta_{M',M''} \tag{302}$$

$$= (-1)^m \sqrt{\frac{(2l+1)(2l'+1)(2L+1)}{4\pi}} \tag{303}$$

$$\times \sum_{L',M'} (-1)^{M'} (2L'+1) \begin{pmatrix} l & l' & L' \\ -m & m' & -M' \end{pmatrix} \begin{pmatrix} l & l' & L' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ M & 0 & M' \end{pmatrix} \begin{pmatrix} L & 1 & L' \\ 0 & 0 & 0 \end{pmatrix} \tag{304}$$

Furthermore,
Prove $\langle \psi_{k,\mu} | \vec{p} | \psi_{k,\nu} \rangle = \langle \psi_{k,\nu} | \vec{p} | \psi_{k,\mu} \rangle^*$:

$$\langle \psi_{k,\nu} | \vec{p} | \psi_{k,\mu} \rangle^* = \left(-i\hbar \int \psi_{k,\nu}^* \nabla \psi_{k,\mu} d\tau \right)^* = i\hbar \int \psi_{k,\nu} \nabla \psi_{k,\mu}^* d\tau \tag{305}$$

$$= i\hbar \int \nabla \left(\psi_{k,\nu} \psi_{k,\mu}^* \right) d\tau - i\hbar \int \left(\nabla \psi_{k,\nu} \right) \psi_{k,\mu}^* d\tau = i\hbar \left[\psi_{k,\nu} \psi_{k,\mu}^* \right]_a^{a+R} - i\hbar \int \left(\nabla \psi_{k,\nu} \right) \psi_{k,\mu}^* d\tau \tag{306}$$

$$= i\hbar \left[\psi_{k,\nu}(a+R) \psi_{k,\mu}^*(a+R) - \psi_{k,\nu}(a) \psi_{k,\mu}^*(a) \right] - i\hbar \int \left(\nabla \psi_{k,\nu} \right) \psi_{k,\mu}^* d\tau \tag{307}$$

$$= i\hbar \left[0 \right] - i\hbar \int \left(\nabla \psi_{k,\nu} \right) \psi_{k,\mu}^* d\tau = -i\hbar \int \psi_{k,\mu}^* \left(\nabla \psi_{k,\nu} \right) d\tau = \langle \psi_{k,m} | \vec{p} | \psi_{k,\nu} \rangle \tag{308}$$

where $\nabla \left(\psi_{k,\nu} \psi_{k,\mu}^* \right) = \left(\nabla \psi_{k,\nu} \right) \psi_{k,\mu}^* + \psi_{k,\nu} \left(\nabla \psi_{k,\mu}^* \right)$.

$$\sigma_{\alpha\beta}(\omega) = \sum_{ijkl=s,p,d,\ldots} \sigma_{\alpha\beta}^{ijkl}(\omega) \tag{309}$$

$$\epsilon_{\alpha\beta}(\omega) = \sum_{ijkl=s,p,d,\ldots} \epsilon_{\alpha\beta}^{ijkl}(\omega) \tag{310}$$

$$\sigma_{\alpha\beta}(\omega) = \sum_{i=s,p,d} \sigma_{\alpha\beta}^{i}(\omega) \tag{311}$$

$$\epsilon_{\alpha\beta}(\omega) = \sum_{i=s,p,d} \epsilon_{\alpha\beta}^{i}(\omega) \tag{312}$$

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