

HW5

November 25, 2016

1 Thermal state of radiation

1.1 Part 1

- (1) For EM modes in a cavity (omitting zero-point energy)

$$E_i = \hbar\omega_i n_i$$

$$H = \sum_i \hbar\omega_i a_i^\dagger a_i$$

Consider the eigenbasis of ρ , which is (number basis) $\{|\phi_{E_i}\rangle, i \in \text{modes}\}$. According to canonical ensemble theory

$$\rho = \sum_i \frac{e^{-\beta E_i}}{Z} |\phi_{E_i}\rangle \langle \phi_{E_i}| = \sum_i \frac{e^{-\beta \hbar\omega_i a_i^\dagger a_i}}{Z} |\phi_{E_i}\rangle \langle \phi_{E_i}| = \frac{e^{-\beta \sum_i \hbar\omega_i a_i^\dagger a_i}}{Z}$$

The expectation value of the offdiagonal operator $a_i a_j^\dagger (i \neq j)$ is

$$\begin{aligned} \text{Tr}(\rho a_i a_j^\dagger) &= \sum_m |\phi_{E_m}\rangle \frac{e^{-\beta \sum_i \hbar\omega_i a_i^\dagger a_i}}{Z} a_i a_j^\dagger \langle \phi_{E_m}| = \sum_{mn} |\phi_{E_m}\rangle \frac{e^{-\beta \sum_i \hbar\omega_i a_i^\dagger a_i}}{Z} \langle \phi_{E_n}| \phi_{E_n}\rangle a_i a_j^\dagger \langle \phi_{E_m}| = \\ &= \sum_{mn} \delta_{mn} \frac{e^{-\beta E_m}}{Z} |\phi_{E_m}\rangle \langle \phi_{E_n}| \phi_{E_n}\rangle a_i a_j^\dagger \langle \phi_{E_m}| = \sum_m \frac{e^{-\beta E_m}}{Z} |\phi_{E_m}\rangle a_i a_j^\dagger \langle \phi_{E_m}| = 0 \quad (i \neq j) \end{aligned}$$

1.2 Part 2

- (1) Similarly, for any operator A with zero diagonal elements, ie, $|\phi_{E_m}\rangle A \langle \phi_{E_m}| = 0$ for all m

$$\begin{aligned} \text{Tr}(\rho A) &= \sum_m |\phi_{E_m}\rangle \frac{e^{-\beta \sum_i \hbar\omega_i a_i^\dagger a_i}}{Z} A \langle \phi_{E_m}| = \sum_{mn} |\phi_{E_m}\rangle \frac{e^{-\beta \sum_i \hbar\omega_i a_i^\dagger a_i}}{Z} \langle \phi_{E_n}| \phi_{E_n}\rangle A \langle \phi_{E_m}| = \\ &= \sum_{mn} \delta_{mn} \frac{e^{-\beta E_m}}{Z} |\phi_{E_m}\rangle \langle \phi_{E_n}| \phi_{E_n}\rangle A \langle \phi_{E_m}| = \sum_m \frac{e^{-\beta E_m}}{Z} |\phi_{E_m}\rangle A \langle \phi_{E_m}| = 0 \end{aligned}$$

2 Four wave mixing

2.1 Part 1

- (1) With no damping, the equation of motion is given by

$$H(t) = H_0 - \mu \cdot (\varepsilon_1(t) + \varepsilon_2(t))$$

$$|\psi(t)\rangle = a(t)e^{-i\frac{E_1}{\hbar}t} |1\rangle + b(t)e^{-i\frac{E_2}{\hbar}t} |2\rangle$$

Or writting explicitly

$$i\hbar \frac{da}{dt} = -\mu_{12}(\varepsilon_1(t) + \varepsilon_2(t))e^{-i\Omega t}b$$

$$i\hbar \frac{db}{dt} = -\mu_{21}(\varepsilon_1(t) + \varepsilon_2(t))e^{i\Omega t}a$$

Assume $\mu_{12} = \mu_{21} = \mu$ is real, and using

$$\varepsilon_1(t) = \frac{1}{2}(\varepsilon_1^- e^{-i\omega_1 t} + \varepsilon_1^+ e^{i\omega_1 t})$$

$$\varepsilon_2(t) = \frac{1}{2}(\varepsilon_2^- e^{-i\omega_2 t} + \varepsilon_2^+ e^{i\omega_2 t})$$

We have (rotating wave approximation)

$$i\hbar \frac{da}{dt} = -\mu_{12} \frac{1}{2}(\varepsilon_1^+ e^{i\omega_1 t} + \varepsilon_2^+ e^{i\omega_2 t})e^{-i\Omega t}b$$

$$i\hbar \frac{db}{dt} = -\mu_{21} \frac{1}{2}(\varepsilon_1^- e^{-i\omega_1 t} + \varepsilon_2^- e^{-i\omega_2 t})e^{i\Omega t}a$$

$$\text{Define } \omega_2 - \Omega = \omega_1 = \delta$$

$$\frac{da}{dt} = i\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+ e^{-i\delta t} + \varepsilon_2^+ e^{i\delta t})b$$

$$\frac{db}{dt} = i\frac{\mu_{21}}{2\hbar}(\varepsilon_1^- e^{i\delta t} + \varepsilon_2^- e^{-i\delta t})a$$

The time-averaged electric dipole is

$$N\langle\mu\rangle = Na^*be^{-i\Omega t}\mu_{12} + Nab^*e^{i\Omega t}\mu_{21} = P_{12}\mu_{12} + P_{21}\mu_{21}$$

Therefore

$$\frac{dP_{12}}{dt} = N\frac{da^*}{dt}be^{-i\Omega t} + Na^*\frac{db}{dt}e^{-i\Omega t} - i\Omega Na^*be^{-i\Omega t} = -iN\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{i\delta t} + \varepsilon_2^-e^{-i\delta t})b^*be^{-i\Omega t} + iN\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{i\delta t} + \varepsilon_2^-e^{-i\delta t})a^*ae^{-i\Omega t} - iN\Omega a^*be^{-i\Omega t} = -i\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{-i\omega_1 t} + \varepsilon_2^-e^{-i\omega_2 t})N(b^*b - a^*a) - i\Omega P_{12}$$

$$\frac{dP_{21}}{dt} = \dots = i\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+e^{i\omega_1 t} + \varepsilon_2^+e^{i\omega_2 t})N(b^*b - a^*a) + i\Omega P_{21}$$

Define $Na^*a = P_{11}$, $Nb^*b = P_{22}$, then

$$\frac{dP_{12}}{dt} = -i\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{-i\omega_1 t} + \varepsilon_2^-e^{-i\omega_2 t})(P_{22} - P_{11}) - i\Omega P_{12}$$

$$\frac{dP_{21}}{dt} = i\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+e^{i\omega_1 t} + \varepsilon_2^+e^{i\omega_2 t})(P_{22} - P_{11}) + i\Omega P_{21}$$

$$\frac{dP_{11}}{dt} = N\frac{da^*}{dt}a + Na^*\frac{da}{dt} = -iN\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{i\delta t} + \varepsilon_2^-e^{-i\delta t})b^*a + iN\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+e^{-i\delta t} + \varepsilon_2^+e^{i\delta t})a^*b = -i\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{-i\omega_1 t} + \varepsilon_2^-e^{-i\omega_2 t})P_{21} + i\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+e^{i\omega_1 t} + \varepsilon_2^+e^{i\omega_2 t})P_{12}$$

$$\frac{dP_{22}}{dt} = \dots = i\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{-i\omega_1 t} + \varepsilon_2^-e^{-i\omega_2 t})P_{21} - i\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+e^{i\omega_1 t} + \varepsilon_2^+e^{i\omega_2 t})P_{12}$$

Define $P_{22} - P_{11} = \Delta N$, the last two equations are equivalent to

$$\frac{d\Delta N}{dt} = 2i\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{-i\omega_1 t} + \varepsilon_2^-e^{-i\omega_2 t})P_{21} - 2i\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+e^{i\omega_1 t} + \varepsilon_2^+e^{i\omega_2 t})P_{12}$$

Add damping terms T_2 and T_1

$$\frac{dP_{12}}{dt} = -i\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{-i\omega_1 t} + \varepsilon_2^-e^{-i\omega_2 t})\Delta N - i\Omega P_{12} - \frac{1}{T_2}P_{12}$$

$$\frac{dP_{21}}{dt} = i\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+e^{i\omega_1 t} + \varepsilon_2^+e^{i\omega_2 t})\Delta N + i\Omega P_{21} - \frac{1}{T_2}P_{21}$$

$$\frac{d\Delta N}{dt} = 2i\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{-i\omega_1 t} + \varepsilon_2^-e^{-i\omega_2 t})P_{21} - 2i\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+e^{i\omega_1 t} + \varepsilon_2^+e^{i\omega_2 t})P_{12} - \frac{\Delta N - \overline{\Delta N}}{T_1}$$

2.2 Part 2

- (1) Since T_2 is the fastest time scale, the above rate equations can be solved by first assuming ΔN is constant and solving the first two equations, and then plugging the solutions to the third one and solving ΔN .

Given $\Delta N(t)$

$$P_{12}(t) = P_{12}(0)e^{(-i\Omega - \frac{1}{T_2})t} - i\frac{\mu_{21}}{2\hbar}\int_0^t dt' (\varepsilon_1^-e^{-i\omega_1 t'} + \varepsilon_2^-e^{-i\omega_2 t'})\Delta N(t')e^{(-i\Omega - \frac{1}{T_2})(t-t')} \simeq -i\frac{\mu_{21}}{2\hbar}\Delta N(t)\int_0^t dt' (\varepsilon_1^-e^{-i\omega_1 t'} + \varepsilon_2^-e^{-i\omega_2 t'})e^{(-i\Omega - \frac{1}{T_2})(t-t')}$$

where

$$\int_0^t dt' \varepsilon_1^-e^{-i\omega_1 t'}e^{(-i\Omega - \frac{1}{T_2})(t-t')} + \int_0^t dt' \varepsilon_2^-e^{-i\omega_2 t'}e^{(-i\Omega - \frac{1}{T_2})(t-t')} = e^{-i\omega_1 t}\int_0^t dt' \varepsilon_1^-e^{(-i(\Omega - \omega_1) - \frac{1}{T_2})(t-t')} + e^{-i\omega_2 t}\int_0^t dt' \varepsilon_2^-e^{(-i(\Omega - \omega_2) - \frac{1}{T_2})(t-t')} \simeq e^{-i\omega_1 t}\int_0^\infty dt' \varepsilon_1^-e^{(-i(\Omega - \omega_1) - \frac{1}{T_2})t'} + e^{-i\omega_2 t}\int_0^\infty dt' \varepsilon_2^-e^{(-i(\Omega - \omega_2) - \frac{1}{T_2})t'} = \varepsilon_1^-e^{-i\omega_1 t}\frac{1}{i(\Omega - \omega_1) + \frac{1}{T_2}} + \varepsilon_2^-e^{-i\omega_2 t}\frac{1}{i(\Omega - \omega_2) + \frac{1}{T_2}}$$

Then

$$P_{12}(t) = -i\frac{\mu_{21}}{2\hbar}\Delta N(t)\{\varepsilon_1^-e^{-i\omega_1 t}\frac{1}{i(\Omega - \omega_1) + \frac{1}{T_2}} + \varepsilon_2^-e^{-i\omega_2 t}\frac{1}{i(\Omega - \omega_2) + \frac{1}{T_2}}\}$$

$$P^-(t) = P_{12}(t)\mu_{12} = -i\frac{\mu_{21}\mu_{12}}{2\hbar}\Delta N(t)\{\varepsilon_1^-e^{-i\omega_1 t}\frac{1}{i(\Omega - \omega_1) + \frac{1}{T_2}} + \varepsilon_2^-e^{-i\omega_2 t}\frac{1}{i(\Omega - \omega_2) + \frac{1}{T_2}}\} =$$

$$\epsilon_0\chi(-\omega_1, \Delta N(t))\frac{1}{2}\varepsilon_1^-e^{-i\omega_1 t} + \epsilon_0\chi(-\omega_2, \Delta N(t))\frac{1}{2}\varepsilon_2^-e^{-i\omega_2 t}$$

(Since $\varepsilon^- = \frac{1}{2}(\varepsilon_1^-e^{-i\omega_1 t} + \varepsilon_2^-e^{-i\omega_2 t})$, there should be a $\frac{1}{2}$)

where

$$\chi(-\omega_i, \Delta N(t)) = -i\frac{\mu_{21}\mu_{12}}{\epsilon_0\hbar}\Delta N(t)\frac{1}{i(\Omega - \omega_i) + \frac{1}{T_2}}$$

2.3 Part 3

2.3.1 Inversion density rate equation

$$\text{Since } \frac{d\Delta N}{dt} = 2i\frac{\mu_{21}}{2\hbar}(\varepsilon_1^-e^{-i\omega_1 t} + \varepsilon_2^-e^{-i\omega_2 t})P_{21} - 2i\frac{\mu_{12}}{2\hbar}(\varepsilon_1^+e^{i\omega_1 t} + \varepsilon_2^+e^{i\omega_2 t})P_{12} - \frac{\Delta N - \overline{\Delta N}}{T_1}$$

$$P_{12}(t)\mu_{12} = \epsilon_0\Delta N\chi(-\omega_1, 1)\frac{1}{2}\varepsilon_1^-e^{-i\omega_1 t} + \epsilon_0\Delta N\chi(-\omega_2, 1)\frac{1}{2}\varepsilon_2^-e^{-i\omega_2 t}$$

Then

$$\frac{d\Delta N}{dt} = -\frac{i\epsilon}{2\hbar}\Delta N(\varepsilon_1^+e^{i\omega_1 t} + \varepsilon_2^+e^{i\omega_2 t})(\chi(-\omega_1, 1)\varepsilon_1^-e^{-i\omega_1 t} + \chi(-\omega_2, 1)\varepsilon_2^-e^{-i\omega_2 t}) - \frac{\Delta N - \overline{\Delta N}}{2T_1} + c.c.$$

Or

$$\frac{d\Delta N}{dt} = -\frac{i\epsilon_0}{2\hbar}\Delta N\{\varepsilon_1^-\varepsilon_1^+\chi(-\omega_1, 1) + \varepsilon_2^-\varepsilon_2^+\chi(-\omega_2, 1) + \varepsilon_1^-\varepsilon_2^+e^{-i(\omega_1 - \omega_2)t}\chi(-\omega_1, 1) + \varepsilon_1^+\varepsilon_2^-e^{i(\omega_1 - \omega_2)t}\chi(-\omega_2, 1)\} - \frac{\Delta N - \overline{\Delta N}}{2T_1} + c.c.$$

2.3.2 Linearize the inversion density rate equation

- (1) Assume $\Delta N = \Delta N_0 + \eta(t)$, where ΔN_0 is the steady state and $\eta(t)$ is a small-signal time-dependent inversion, then

$$\frac{d\Delta N}{dt} = \frac{d\eta}{dt} = -\frac{i\epsilon_0}{2\hbar}(\Delta N_0 + \eta)\{\epsilon_1^-\epsilon_1^+\chi(-\omega_1, 1) + \epsilon_2^-\epsilon_2^+\chi(-\omega_2, 1) + \epsilon_1^-\epsilon_2^+e^{-i(\omega_1-\omega_2)t}\chi(-\omega_1, 1) + \epsilon_1^+\epsilon_2^-e^{i(\omega_1-\omega_2)t}\chi(-\omega_2, 1)\} - \frac{\Delta N_0 - \overline{\Delta N} + \eta}{2T_1} + c.c.$$

Since ΔN_0 is the steady state inversion

$$\frac{d\Delta N_0}{dt} = -\frac{i\epsilon_0}{2\hbar}\Delta N_0\{\epsilon_1^-\epsilon_1^+\chi(-\omega_1, 1) + \epsilon_2^-\epsilon_2^+\chi(-\omega_2, 1)\} - \frac{\Delta N_0 - \overline{\Delta N}}{2T_1} + c.c. = 0$$

Then

$$\frac{d\eta}{dt} = -\frac{i\epsilon_0}{2\hbar}\eta\{\epsilon_1^-\epsilon_1^+\chi(-\omega_1, 1) + \epsilon_2^-\epsilon_2^+\chi(-\omega_2, 1)\} - \frac{i\epsilon_0}{2\hbar}(\Delta N_0 + \eta)\{\epsilon_1^-\epsilon_2^+e^{-i(\omega_1-\omega_2)t}\chi(-\omega_1, 1) + \epsilon_1^+\epsilon_2^-e^{i(\omega_1-\omega_2)t}\chi(-\omega_2, 1)\} - \frac{\eta}{2T_1} + c.c.$$

Since $\Delta N_0 \gg \eta$

$$\frac{d\eta}{dt} \simeq -\frac{i\epsilon_0}{2\hbar}\eta\{\epsilon_1^-\epsilon_1^+\chi(-\omega_1, 1) + \epsilon_2^-\epsilon_2^+\chi(-\omega_2, 1)\} - \frac{i\epsilon_0}{2\hbar}\Delta N_0\{\epsilon_1^-\epsilon_2^+e^{-i(\omega_1-\omega_2)t}\chi(-\omega_1, 1) + \epsilon_1^+\epsilon_2^-e^{i(\omega_1-\omega_2)t}\chi(-\omega_2, 1)\} - \frac{\eta}{2T_1} + c.c.$$

- (2) Let $\eta(t) = \eta^+e^{i(\omega_1-\omega_2)t} + \eta^-e^{-i(\omega_1-\omega_2)t}$, then

$$\frac{d\eta}{dt} = \frac{d\eta^+}{dt}e^{i(\omega_1-\omega_2)t} + \frac{d\eta^-}{dt}e^{-i(\omega_1-\omega_2)t} + i(\omega_1 - \omega_2)(\eta^+e^{i(\omega_1-\omega_2)t} - \eta^-e^{-i(\omega_1-\omega_2)t}) = -\frac{i\epsilon_0}{2\hbar}(\eta^+e^{i(\omega_1-\omega_2)t} + \eta^-e^{-i(\omega_1-\omega_2)t})\{\epsilon_1^-\epsilon_1^+\chi(-\omega_1, 1) + \epsilon_2^-\epsilon_2^+\chi(-\omega_2, 1)\} - \frac{i\epsilon_0}{2\hbar}\Delta N_0\{\epsilon_1^-\epsilon_2^+e^{-i(\omega_1-\omega_2)t}\chi(-\omega_1, 1) + \epsilon_1^+\epsilon_2^-e^{i(\omega_1-\omega_2)t}\chi(-\omega_2, 1)\} - \frac{\eta^+e^{i(\omega_1-\omega_2)t} + \eta^-e^{-i(\omega_1-\omega_2)t}}{2T_1} + c.c.$$

Or writing separately

$$\frac{d\eta^+}{dt} + i(\omega_1 - \omega_2)\eta^+ = -\frac{i\epsilon_0}{2\hbar}\eta^+\{\epsilon_1^-\epsilon_1^+\chi(-\omega_1, 1) + \epsilon_2^-\epsilon_2^+\chi(-\omega_2, 1) - \epsilon_1^-\epsilon_1^+\chi^*(-\omega_1, 1) - \epsilon_2^-\epsilon_2^+\chi^*(-\omega_2, 1)\} - \frac{i\epsilon_0}{2\hbar}\Delta N_0\{\epsilon_1^+\epsilon_2^-\chi(-\omega_2, 1) - \epsilon_1^+\epsilon_2^-\chi^*(-\omega_1, 1)\} - \frac{\eta^+}{T_1}$$

Using

$$\chi(-\omega_i, 1) = \chi_R(-\omega_i, 1) + i\chi_I(-\omega_i, 1)$$

$$\text{Since } \omega_2 - \Omega = \Omega - \omega_1 = \delta$$

$$\chi(-\omega_1, 1) = -i\frac{\mu_{21}\mu_{12}}{\epsilon_0\hbar} \frac{1}{-i\delta + \frac{1}{T_2}} = -\chi^*(-\omega_2, 1)$$

$$\chi_R(-\omega_1, 1) = -\chi_R(-\omega_2, 1)$$

$$\chi_I(-\omega_1, 1) = \chi_I(-\omega_2, 1)$$

Therefore

$$\frac{d\eta^+}{dt} + i(\omega_1 - \omega_2)\eta^+ = \frac{\epsilon_0}{\hbar}\eta^+\{(\epsilon_1^-\epsilon_1^+ + \epsilon_2^-\epsilon_2^+)\chi_I(-\omega_2, 1)\} - \frac{i\epsilon_0}{\hbar}\Delta N_0\epsilon_1^+\epsilon_2^-\chi(-\omega_2, 1) - \frac{\eta^+}{T_1}$$

$$\frac{d\eta^+}{dt} + i(\omega_1 - \omega_2)\eta^+ + \frac{\eta^+}{T_1} = -\frac{i\epsilon_0}{\hbar}\Delta N_0\epsilon_1^+\epsilon_2^-\chi(-\omega_2, 1)$$

$$\text{where } \frac{1}{T_1} = \frac{1}{T_1} - \frac{\epsilon_0}{\hbar}\{(\epsilon_1^-\epsilon_1^+ + \epsilon_2^-\epsilon_2^+)\chi_I(-\omega_2, 1)\}$$

Steady state solution

$$\eta^+ = -\frac{\frac{i\epsilon_0}{\hbar}\epsilon_1^+\epsilon_2^-\chi(-\omega_2, 1)}{i(\omega_1-\omega_2) + \frac{1}{T_1}}\Delta N_0 = \beta^+\epsilon_1^+\epsilon_2^-\Delta N_0$$

where

$$\beta^+ = -\frac{\frac{i\epsilon_0}{\hbar}\chi(-\omega_2, 1)}{i(\omega_1-\omega_2) + \frac{1}{T_1}}$$

Similarly

$$\eta^- = -\frac{\frac{i\epsilon_0}{\hbar}\epsilon_1^-\epsilon_2^+\chi(-\omega_1, 1)}{-i(\omega_1-\omega_2) + \frac{1}{T_1}}\Delta N_0 = \beta^-\epsilon_1^-\epsilon_2^+\Delta N_0$$

where

$$\beta^- = -\frac{\frac{i\epsilon_0}{\hbar}\chi(-\omega_1, 1)}{-i(\omega_1-\omega_2) + \frac{1}{T_1}}$$

2.4 Part 4

- (1) Polarization

$$\begin{aligned} P(t) &= P_{12}(t)\mu_{12} + P_{21}(t)\mu_{21} = \epsilon_0\Delta N\chi(-\omega_1, 1)\frac{1}{2}\epsilon_1^-e^{-i\omega_1 t} + \epsilon_0\Delta N\chi(-\omega_2, 1)\frac{1}{2}\epsilon_2^-e^{-i\omega_2 t} + c.c. \\ &= (\Delta N_0 + \eta^+e^{i(\omega_1-\omega_2)t} + \eta^-e^{-i(\omega_1-\omega_2)t})(\epsilon_0\chi(-\omega_1, 1)\frac{1}{2}\epsilon_1^-e^{-i\omega_1 t} + \epsilon_0\chi(-\omega_2, 1)\frac{1}{2}\epsilon_2^-e^{-i\omega_2 t}) + c.c. \\ &= \Delta N_0(1 + \beta^+\epsilon_1^+\epsilon_2^-e^{i(\omega_1-\omega_2)t} + \beta^-\epsilon_1^-\epsilon_2^+e^{-i(\omega_1-\omega_2)t})(\epsilon_0\chi(-\omega_1, 1)\frac{1}{2}\epsilon_1^-e^{-i\omega_1 t} + \epsilon_0\chi(-\omega_2, 1)\frac{1}{2}\epsilon_2^-e^{-i\omega_2 t}) + c.c. \\ &= \Delta N_0(\epsilon_0\chi(-\omega_1, 1)\frac{1}{2}\epsilon_1^-e^{-i\omega_1 t} + \epsilon_0\chi(-\omega_2, 1)\frac{1}{2}\epsilon_2^-e^{-i\omega_2 t} + \epsilon_0\chi(-\omega_1, 1)\frac{1}{2}\beta^+\epsilon_1^+\epsilon_1^-\epsilon_2^-e^{-i\omega_2 t} + \end{aligned}$$

$$\epsilon_0 \chi(-\omega_2, 1) \frac{1}{2} \beta^+ \epsilon_1^+ \epsilon_2^- \epsilon_2^- e^{i(\omega_1 - 2\omega_2)t} + \epsilon_0 \chi(-\omega_1, 1) \frac{1}{2} \beta^- \epsilon_1^- \epsilon_1^- \epsilon_2^+ e^{-i(2\omega_1 - \omega_2)t} + \epsilon_0 \chi(-\omega_2, 1) \frac{1}{2} \beta^- \epsilon_1^- \epsilon_2^+ \epsilon_2^- e^{-i\omega_1 t} + c.c.$$

oscillates at four frequencies $\omega_1, \omega_2, 2\omega_1 - \omega_2, 2\omega_2 - \omega_1$

The corresponding third order susceptibilities are

$$\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \Delta N_0) = 4\Delta N_0 \chi(-\omega_1, 1) \beta^+ = -4\Delta N_0 \frac{\frac{i\epsilon_0}{\hbar} \chi(-\omega_2, 1) \chi(-\omega_1, 1)}{i(\omega_1 - \omega_2) + \frac{1}{T_1}}$$

$$\chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0) = 4\Delta N_0 \chi(-\omega_2, 1) \beta^- = -4\Delta N_0 \frac{\frac{i\epsilon_0}{\hbar} \chi(-\omega_2, 1) \chi(-\omega_1, 1)}{-i(\omega_1 - \omega_2) + \frac{1}{T_1}}$$

$$\chi^{(3)}(-\omega_1, -\omega_1, \omega_2, \Delta N_0) = 4\Delta N_0 \chi(-\omega_1, 1) \beta^- = -4\Delta N_0 \frac{\frac{i\epsilon_0}{\hbar} \chi(-\omega_1, 1)^2}{-i(\omega_1 - \omega_2) + \frac{1}{T_1}}$$

$$\chi^{(3)}(-\omega_2, -\omega_2, \omega_1, \Delta N_0) = 4\Delta N_0 \chi(-\omega_2, 1) \beta^+ = -4\Delta N_0 \frac{\frac{i\epsilon_0}{\hbar} \chi(-\omega_2, 1)^2}{i(\omega_1 - \omega_2) + \frac{1}{T_1}}$$

$P(t)$ is defined as

$$P(t) = \epsilon_0 \chi(-\omega_1, \Delta N_0) \frac{1}{2} \epsilon_1^- e^{-i\omega_1 t} + \epsilon_0 \chi(-\omega_2, \Delta N_0) \frac{1}{2} \epsilon_2^- e^{-i\omega_2 t} + \epsilon_0 \chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \Delta N_0) \frac{1}{2} \epsilon_1^- e^{-i\omega_1 t} \frac{1}{2} \epsilon_1^+ e^{i\omega_1 t} \frac{1}{2} \epsilon_2^- e^{-i\omega_2 t} + \epsilon_0 \chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0) \frac{1}{2} \epsilon_2^- e^{-i\omega_2 t} \frac{1}{2} \epsilon_2^+ e^{i\omega_2 t} \frac{1}{2} \epsilon_1^- e^{-i\omega_1 t} + \epsilon_0 \chi^{(3)}(-\omega_1, -\omega_1, \omega_2, \Delta N_0) \frac{1}{2} \epsilon_1^- e^{-i\omega_1 t} \frac{1}{2} \epsilon_1^- e^{-i\omega_1 t} \frac{1}{2} \epsilon_2^+ e^{i\omega_2 t} + \epsilon_0 \chi^{(3)}(-\omega_2, -\omega_2, \omega_1, \Delta N_0) \frac{1}{2} \epsilon_2^- e^{-i\omega_2 t} \frac{1}{2} \epsilon_2^- e^{-i\omega_2 t} \frac{1}{2} \epsilon_1^+ e^{i\omega_1 t} + c.c.$$

- (2) Consider the wave equation along the z direction

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2}$$

From above, we have

$$P(t) = \epsilon_0 \chi(-\omega_1, \Delta N_0) \frac{1}{2} \epsilon_1^- e^{-i\omega_1 t} + \epsilon_0 \chi(-\omega_2, \Delta N_0) \frac{1}{2} \epsilon_2^- e^{-i\omega_2 t} + \epsilon_0 \chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \Delta N_0) \frac{1}{8} \epsilon_1^- \epsilon_1^+ \epsilon_2^- e^{-i\omega_2 t} + \epsilon_0 \chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0) \frac{1}{8} \epsilon_2^- \epsilon_2^+ \epsilon_1^- e^{-i\omega_1 t} + \epsilon_0 \chi^{(3)}(-\omega_1, -\omega_1, \omega_2, \Delta N_0) \frac{1}{8} \epsilon_1^- \epsilon_1^- \epsilon_2^+ e^{-i(2\omega_1 - \omega_2)t} + \epsilon_0 \chi^{(3)}(-\omega_2, -\omega_2, \omega_1, \Delta N_0) \frac{1}{8} \epsilon_2^- \epsilon_2^- \epsilon_1^+ e^{i(\omega_1 - 2\omega_2)t} + c.c.$$

$$\begin{aligned} \frac{\partial^2 P}{\partial t^2} &= -\omega_1^2 \epsilon_0 \chi(-\omega_1, \Delta N_0) \frac{1}{2} \epsilon_1^- e^{-i\omega_1 t} - \omega_2^2 \epsilon_0 \chi(-\omega_2, \Delta N_0) \frac{1}{2} \epsilon_2^- e^{-i\omega_2 t} - \\ &\omega_2^2 \epsilon_0 \chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \Delta N_0) \frac{1}{8} \epsilon_1^- \epsilon_1^+ \epsilon_2^- e^{-i\omega_2 t} - \omega_1^2 \epsilon_0 \chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0) \frac{1}{8} \epsilon_2^- \epsilon_2^+ \epsilon_1^- e^{-i\omega_1 t} - \\ &(2\omega_1 - \omega_2)^2 \epsilon_0 \chi^{(3)}(-\omega_1, -\omega_1, \omega_2, \Delta N_0) \frac{1}{8} \epsilon_1^- \epsilon_1^- \epsilon_2^+ e^{-i(2\omega_1 - \omega_2)t} + -(\omega_1 - \omega_2)^2 \epsilon_0 \chi^{(3)}(-\omega_2, -\omega_2, \omega_1, \Delta N_0) \frac{1}{8} \epsilon_2^- \epsilon_2^- \epsilon_1^+ e^{i(\omega_1 - 2\omega_2)t} + c.c. \end{aligned}$$

The plane wave solution has the form

$$E = \frac{1}{2} E_1^- e^{-i(\omega_1 t - k_1 z)} + \frac{1}{2} E_2^- e^{-i(\omega_2 t - k_2 z)} + \frac{1}{2} E_3^- e^{-i((2\omega_1 - \omega_2)t - k_3 z)} + \frac{1}{2} E_4^- e^{-i((2\omega_2 - \omega_1)t - k_4 z)} + c.c.$$

Consider the solution with frequency ω_1 and ω_2 , we have

$$-k_1^2 \frac{1}{2} E_1^- + \frac{\omega_1^2}{c^2} \frac{1}{2} E_1^- = -\frac{\omega_1^2}{c^2} \chi(-\omega_1, \Delta N_0) \frac{1}{2} \epsilon_1^- - \frac{\omega_2^2}{c^2} \chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0) \frac{1}{8} \epsilon_2^- \epsilon_2^+ \epsilon_1^-$$

Then

$$k_1^2 = \frac{\omega_1^2}{c^2} \{1 + \chi(-\omega_1, \Delta N_0) \frac{\epsilon_1^-}{E_1^-} + \chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0) \frac{1}{4} \epsilon_2^- \epsilon_2^+ \frac{\epsilon_1^-}{E_1^-}\}$$

Since $\chi(\cdot)$ and $\chi^{(3)}(\cdot)$ are small compared to 1, then

$$\epsilon_1^- \simeq E_1^-$$

$$k_1 = \frac{\omega_1}{c} \{1 + \frac{1}{2} \chi(-\omega_1, \Delta N_0) + \frac{1}{8} \chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0) \epsilon_2^- \epsilon_2^+\}$$

Similarly

$$k_2 = \frac{\omega_2}{c} \{1 + \frac{1}{2} \chi(-\omega_2, \Delta N_0) + \frac{1}{8} \chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \Delta N_0) \epsilon_1^- \epsilon_1^+\}$$

Therefore, the effective dispersion and absorption of the $\chi^{(3)}$ terms are

$$\begin{aligned} \frac{1}{8} \text{Re}\{\chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0)\} \epsilon_2^- \epsilon_2^+ &= \frac{1}{2} \text{Re}\{\Delta N_0 \chi(-\omega_2, 1) \beta^-\} \epsilon_2^- \epsilon_2^+ \\ \frac{1}{8} \text{Im}\{\chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0)\} \epsilon_2^- \epsilon_2^+ &= \frac{1}{2} \text{Im}\{\Delta N_0 \chi(-\omega_2, 1) \beta^-\} \epsilon_2^- \epsilon_2^+ \\ \frac{1}{8} \text{Re}\{\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \Delta N_0)\} \epsilon_1^- \epsilon_1^+ &= \frac{1}{2} \text{Re}\{\Delta N_0 \chi(-\omega_1, 1) \beta^+\} \epsilon_1^- \epsilon_1^+ \\ \frac{1}{8} \text{Im}\{\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \Delta N_0)\} \epsilon_1^- \epsilon_1^+ &= \frac{1}{2} \text{Im}\{\Delta N_0 \chi(-\omega_1, 1) \beta^+\} \epsilon_1^- \epsilon_1^+ \end{aligned}$$

where

$$\Delta N_0 \chi(-\omega_2, 1) \beta^- = -\Delta N_0 \frac{\frac{i\epsilon_0}{\hbar} \chi(-\omega_2, 1) \chi(-\omega_1, 1)}{-i(\omega_1 - \omega_2) + \frac{1}{T_1}} = \Delta N_0 \frac{\frac{i\epsilon_0}{\hbar} |\chi(-\omega_2, 1)|^2}{-i(\omega_1 - \omega_2) + \frac{1}{T_1}} =$$

$$\Delta N_0 \frac{\epsilon_0}{\hbar} |\chi(-\omega_2, 1)|^2 \frac{-\frac{1}{T_1}}{(\omega_1 - \omega_2)^2 + \frac{1}{T_1^2}} = \Delta N_0 \frac{\epsilon_0^3 \mu_{21}^2 \mu_{12}^2}{\hbar^3} \frac{1}{(\Omega - \omega_2)^2 + \frac{1}{T_2^2}} \frac{-\frac{1}{T_1}}{(\omega_1 - \omega_2)^2 + \frac{1}{T_1^2}} =$$

$$\Delta N_0 \frac{\epsilon_0^3 \mu_{21}^2 \mu_{12}^2}{\hbar^3} \frac{1}{\delta^2 + \frac{1}{T_2^2}} \frac{2\delta + i\frac{1}{T_1}}{4\delta^2 + \frac{1}{T_1^2}}$$

Similarly

$$\Delta N_0 \chi(-\omega_1, 1) \beta^+ = \Delta N_0 \frac{\epsilon_0^3 \mu_{21}^2 \mu_{12}^2}{\hbar^3} \frac{1}{\delta^2 + \frac{1}{T_2^2}} \frac{-2\delta + i\frac{1}{T_1}}{4\delta^2 + \frac{1}{T_1^2}}$$

Then

$$\frac{1}{8}\text{Re}\{\chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0)\}\varepsilon_2^-\varepsilon_2^+ = \frac{1}{2}\Delta N_0 \frac{\epsilon_0^3 \mu_{21}^2 \mu_{12}^2}{\hbar^3} \frac{1}{\delta^2 + \frac{1}{T_2^2}} \frac{2\delta}{4\delta^2 + \frac{1}{T_1^2}} \varepsilon_2^-\varepsilon_2^+$$

$$\frac{1}{8}\text{Im}\{\chi^{(3)}(-\omega_2, \omega_2, -\omega_1, \Delta N_0)\}\varepsilon_2^-\varepsilon_2^+ = \frac{1}{2}\Delta N_0 \frac{\epsilon_0^3 \mu_{21}^2 \mu_{12}^2}{\hbar^3} \frac{1}{\delta^2 + \frac{1}{T_2^2}} \frac{\frac{1}{T_1}}{4\delta^2 + \frac{1}{T_1^2}} \varepsilon_2^-\varepsilon_2^+$$

$$\frac{1}{8}\text{Re}\{\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \Delta N_0)\}\varepsilon_1^-\varepsilon_1^+ = \frac{1}{2}\Delta N_0 \frac{\epsilon_0^3 \mu_{21}^2 \mu_{12}^2}{\hbar^3} \frac{1}{\delta^2 + \frac{1}{T_2^2}} \frac{-2\delta}{4\delta^2 + \frac{1}{T_1^2}} \varepsilon_1^-\varepsilon_1^+$$

$$\frac{1}{8}\text{Im}\{\chi^{(3)}(-\omega_1, \omega_1, -\omega_2, \Delta N_0)\}\varepsilon_1^-\varepsilon_1^+ = \frac{1}{2}\Delta N_0 \frac{\epsilon_0^3 \mu_{21}^2 \mu_{12}^2}{\hbar^3} \frac{1}{\delta^2 + \frac{1}{T_2^2}} \frac{\frac{1}{T_1}}{4\delta^2 + \frac{1}{T_1^2}} \varepsilon_1^-\varepsilon_1^+$$

If $\Delta N_0 > 0$, absorption

If $\Delta N_0 < 0$, gain

If $\Delta N_0 = 0$, transparency