

# Derivation for the Theory of Dicke Narrowing in Electromagnetically induced Transparency

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## 1 Theory

### 1.1 Theorization of the System

#### 1.1.1 EOM of the System

As derived in a different section, the preliminary c dot solutions of the system writes:

$$\begin{aligned}\dot{c}_1 &= i\chi_{31}^*(t)c_3 \\ \dot{c}_2 &= -i\omega_{21}c_2 + i\chi_{32}^*(t)c_3 \\ \dot{c}_3 &= -i\omega_{31}c_3 + i\chi_{31}(t)c_1 + i\chi_{32}(t)c_2.\end{aligned}$$

This gives the coherence terms:

$$\dot{\rho}_{21} = -i\chi_{31}(t)\rho_{23} + i\chi_{32}^*(t)\rho_{31} - i(\omega_{21} - i\gamma_{21})\rho_{21}, \quad (1a)$$

$$\dot{\rho}_{31} = -i\chi_{31}(t)(\rho_{33} - \rho_{11}) + i\chi_{32}(t)\rho_{21} - i(\omega_{31} - i\gamma_{31})\rho_{31}, \quad (1b)$$

$$\dot{\rho}_{32} = -i\chi_{32}(t)(\rho_{33} - \rho_{22}) + i\chi_{31}(t)\rho_{12} - i(\omega_{32} - i\gamma_{32})\rho_{32}. \quad (1c)$$

To simplify population terms, apply a complex equivalency for complex number  $z$ :

$$\begin{aligned}\text{Im}(z) &= \text{Re} [(-i)z] = \text{Re} (iz^*), \\ \text{Re}(z) &= \text{Im} (iz) = \text{Im} (iz^*),\end{aligned}$$

which can be easily proved by replacing  $z$  by  $a + ib$  for real numbers  $a$  and  $b$ , and recall that any complex number can be expressed in this form. Now the population terms writes

$$\dot{\rho}_{33} = 2\text{Im} [\chi_{31}^*(t)\rho_{31} + \chi_{32}^*(t)\rho_{32}] - \gamma_{33}\rho_{33}. \quad (1d)$$

Consider the case of weak probe, namely  $\chi_{31} \ll \chi_{32}$ , and avoid saturation by assuming  $\chi_{32} < \text{any} \gamma_{nn'}$ . The perturbation solution of Eqs. 8 can easily be done.

To the zeroth order in  $\chi_{31}$ , with the initial state  $\rho(0) = |1\rangle\langle 1|$ , we find

$$\rho_{11}^{(0)} = 1, \quad (2)$$

and all other matrix elements vanish. To the first order in  $\chi_{31}$ ,

$$\dot{\rho}_{21}^{(1)} = i\chi_{32}^*(t)\rho_{31} - i(\omega_{21} - i\gamma_{21})\rho_{21}, \quad (3a)$$

$$\dot{\rho}_{31}^{(1)} = i\chi_{31}(t) + i\chi_{32}(t)\rho_{21} - i(\omega_{31} - i\gamma_1)\rho_{31}. \quad (3b)$$

And all the other matrix elements vanish. Note that the  $\gamma_{31}$  from Eq. (1b) was replaced by  $\gamma_1$  in Eq. (3b). The definition, along with the physical significance, of these gamma terms are discussed in a separate section.

## 1.2 Dicke Narrowing

### 1.2.1 $S(\omega)$

The time average absorption spectrum is given by

$$S(\omega) \equiv \text{Im} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \frac{\rho_{31}(t)}{\chi_{31}(t)} = \text{Im} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \frac{\rho_{31}(t)}{\chi_{31}} e^{-i\mathbf{q}_1 \cdot \mathbf{r}(t) + i\omega_1 t}. \quad (1.2.1)$$

A first-order linear differential equation takes the form:

$$y' + p(x)y + g(x) = 0.$$

And the solution is given by:

$$y = Ce^{\int p(x)dx} + e^{\int p(x)dx} \int g(x)e^{-\int p(x)dx} dx,$$

where the notations follow the conventions in educational mathematics with C here being a constant of integration.

Eq.(3) is a set of first-order differential equations. Assume initially there is no coherence, i.e., the coherence terms vanish at  $t=0$  (thus the corresponding integration constant  $c$  vanishes). Eq.(3a) has the solution:

$$\rho_{21} = i \int_0^t \chi_{32}^*(t_1)\rho_{31}(t_1)e^{(-i\omega_{21}-\gamma_{21})(t-t_1)} dt_1.$$

Substitute it back to Eq.(3b),

$$\dot{\rho}_{31} = i\chi_{31}(t) - \chi_{32}(t) \int_0^t \chi_{32}^*(t_1)\rho_{31}(t_1)e^{(-i\omega_{21}-\gamma_{21})(t-t_1)} dt_1 - i(\omega_{31} - i\gamma_1)\rho_{31},$$

which is still a first-order linear differential equation. Following the same process, the preliminary  $\rho_{31}$  solution is

$$\rho_{31} = \int_0^t e^{(-i\omega_{31}-\gamma_1)(t-t_2)} \left[ i\chi_{31}(t_2) - \chi_{32}(t_2) \int_0^{t_2} \chi_{32}^*(t_1)e^{(-i\omega_{21}-\gamma_{21})(t_2-t_1)} \rho_{31}(t_1) dt_1 \right] dt_2. \quad (1.2.2)$$

It invokes much complexity to analytically solve the equation. Instead, analogous to the Runge-Kutta2 method in computational mathematics, we introduce an intermediate step solution to  $\rho_{31}$

and perform another iteration of the formula, i.e., we put the zeroth order of  $\rho_{31}$  ( $=0$ , see the contexts of Eq. (2)) in the brackets of Eq. (1.2.2). Now,

$$\begin{aligned}\rho_{31}^{(mid)} &= \int_0^t e^{(-i\omega_{31}-\gamma_1)(t-t_2)} [i\chi_{31}(t_2) - \chi_{32}(t_2) \int_0^{t_2} \chi_{32}^*(t_1) e^{(-i\omega_{21}-\gamma_{21})(t_2-t_1)} \rho_{31}(t_1)^{(0)} dt_1] dt_2 \\ &= \int_0^t e^{(-i\omega_{31}-\gamma_1)(t-t_2)} i\chi_{31}(t_2) dt_2.\end{aligned}\quad (1.2.3)$$

The final iteration is done in the same way by inputting the  $\rho_{31}(t_1)$  in Eq. (1.2.2) by Eq. (1.2.3). Now, with  $t_i$ 's adjusted in numerical order and  $\chi$ 's expressed with explicit time dependence terms, the approximated  $\rho_{31}$  becomes

$$\begin{aligned}\rho_{31} &= i\chi_{31} \int_0^t dt_1 e^{(-i\omega_{31}-\gamma_1)(t-t_1)} e^{i\mathbf{q}_1 \cdot \mathbf{r}(t_1) - i\omega_1 t_1} \\ &\quad + (-i|\chi_{32}|^2 \chi_{31}) \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 e^{(-i\omega_{31}-\gamma_1)(t-t_1) + i\mathbf{q}_2 \cdot \mathbf{r}(t_1) - i\omega_2 t_1} \\ &\quad \times e^{(-i\omega_{21}-\gamma_{21})(t_1-t_2) - i\mathbf{q}_2 \cdot \mathbf{r}(t_2) + i\omega_2 t_2} e^{(-i\omega_{31}-\gamma_1)(t_2-t_3) + i\mathbf{q}_1 \cdot \mathbf{r}(t_3) - i\omega_1 t_3}.\end{aligned}$$

Apply Eq.(1.2.1) and split the two addend terms on the right hand side above to have  $S(\omega) = S_1 + S_2$ , i.e.,

$$S_1 = \text{Im} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \frac{1}{\chi_{31}} e^{-i\mathbf{q}_1 \cdot \mathbf{r}(t) + i\omega_1 t} [i\chi_{31} \int_0^t dt_1 e^{(-i\omega_{31}-\gamma_1)(t-t_1)} e^{i\mathbf{q}_1 \cdot \mathbf{r}(t_1) - i\omega_1 t_1}], \quad (1.2.4a)$$

$$\begin{aligned}S_2 &= \text{Im} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \frac{1}{\chi_{31}} e^{-i\mathbf{q}_1 \cdot \mathbf{r}(t) + i\omega_1 t} \\ &\quad \times [(-i|\chi_{32}|^2 \chi_{31}) \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 e^{(-i\omega_{31}-\gamma_1)(t-t_1) + i\mathbf{q}_2 \cdot \mathbf{r}(t_1) - i\omega_2 t_1} \\ &\quad \times e^{(-i\omega_{21}-\gamma_{21})(t_1-t_2) - i\mathbf{q}_2 \cdot \mathbf{r}(t_2) + i\omega_2 t_2} e^{(-i\omega_{31}-\gamma_1)(t_2-t_3) + i\mathbf{q}_1 \cdot \mathbf{r}(t_3) - i\omega_1 t_3}].\end{aligned}\quad (1.2.4b)$$

### 1.2.2 $S_1$

#### i> A Preliminary Expression

From Eq.(1.2.4a), we have

$$\begin{aligned}S_1 &= \text{Im} \lim_{T \rightarrow \infty} i \int_0^T \frac{dt}{T} \int_0^t dt_1 e^{-i\mathbf{q}_1 \cdot \mathbf{r}(t) + i\omega_1 t} e^{(-i\omega_{31}-\gamma_1)(t-t_1)} e^{i\mathbf{q}_1 \cdot \mathbf{r}(t_1) - i\omega_1 t_1} \\ &= -\text{Re} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \int_0^t dt_1 e^{i\mathbf{q}_1 \cdot \mathbf{r}(t) - i\omega_1 t} e^{(i\omega_{31}-\gamma_1)(t-t_1)} e^{-i\mathbf{q}_1 \cdot \mathbf{r}(t_1) + i\omega_1 t_1},\end{aligned}\quad (1.2.5)$$

in which one of the complex identities was applied. Let  $\tau = t - t_1$  and set the  $\tau$  integral upper limit to  $\infty$ . In this basis, Eq.(1.2.5) becomes

$$\begin{aligned}
S_1 &= \text{Re} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \int_0^\infty d\tau e^{i\mathbf{q}_1 \cdot \mathbf{r}(t) - i\omega_1 t} e^{(i\omega_{31} - \gamma_1)\tau} e^{-i\mathbf{q}_1 \cdot \mathbf{r}(t-\tau) + i\omega_1(t-\tau)} \\
&= \text{Re} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \int_0^\infty d\tau e^{-i(\Delta_1 - i\gamma_1)\tau} e^{i\mathbf{q}_1 [\mathbf{r}(t) - \mathbf{r}(t-\tau)]} \\
&= \text{Re} \int_0^\infty d\tau e^{-i(\Delta_1 - i\gamma_1)\tau} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} e^{i\mathbf{q}_1 [\mathbf{r}(t) - \mathbf{r}(t-\tau)]} \\
&= \text{Re} \int_0^\infty d\tau e^{-i(\Delta_1 - i\gamma_1)\tau} \overline{e^{i\Phi_1(t, \tau)}}, \tag{1.2.6}
\end{aligned}$$

where  $\Delta_1 \equiv \omega_1 - \omega_{31}$  and  $\Phi_n(t, \tau) \equiv \mathbf{q}_n [\mathbf{r}(t) - \mathbf{r}(t - \tau)]$ .

## ii> The $\overline{e^{i\Phi}}$ Term

Limit our discussion to the situation where the atoms have sufficiently long reached thermal equilibrium before meeting the light fields. We have

$$\begin{aligned}
\overline{e^{i\Phi_n(t, \tau)}} &= \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} e^{i\Phi_n(t, \tau)} \\
&= \lim_{T \rightarrow \infty} \left[ \int_{\Delta t}^T \frac{dt}{T} e^{i\Phi_n(t, \tau)} + \int_0^{\Delta t} \frac{dt}{T} e^{i\Phi_n(t, \tau)} \right],
\end{aligned}$$

For any  $\Delta t \neq \infty$ . Since  $\Phi$  is real, the magnitude of  $e^{i\Phi}$  would not introduce infinity for all  $t$ , allowing the second term to vanish. Then,

$$\begin{aligned}
\overline{e^{i\Phi_n(t, \tau)}} &= \lim_{T \rightarrow \infty} \int_{\Delta t}^T \frac{dt}{T} e^{i\Phi_n(t, \tau)} \\
&= \lim_{T \rightarrow \infty} \int_{\Delta t}^{T+\Delta t} \frac{dt}{T} e^{i\Phi_n(t, \tau)} \\
&= \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} e^{i\Phi_n(t+\Delta t, \tau)} \\
&= \overline{e^{i\Phi_n(t+\Delta t, \tau)}}.
\end{aligned}$$

An ensemble of atoms in a box in thermal equilibrium is a case of an ergodic system. In an ergodic system, the sufficiently long temporal average of a statistical measurement on an individual equals the average of the ensemble at any specific time. Let a superscript of apostrophe by the overbar indicate the ensemble average at  $t = 0$ , we have

$$\overline{e^{i\Phi_n(t+\Delta t, \tau)}} = \overline{e^{i\Phi_n(\Delta t, \tau)}}'.$$

Since  $\Delta t$  is arbitrary, for any  $t_1, t_2 \neq \infty$  we have

$$\begin{aligned}
\overline{e^{i\Phi_n(t_1, \tau)}}' &= \overline{e^{i\Phi_n(t_2, \tau)}}', \\
\implies \overline{e^{i\Phi_n(0, \tau)}}' &= \overline{e^{i\Phi_n(\Delta t + \tau, \tau)}}'.
\end{aligned}$$

Further,

$$\begin{aligned}
\Phi_n(\Delta t + \tau, \tau) &= \mathbf{q}_n [\mathbf{r}(\Delta t + \tau) - \mathbf{r}(\Delta t)] \\
&= \mathbf{q}_n \int_{\Delta t}^{\tau + \Delta t} \frac{d\mathbf{r}(t_1)}{dt_1} dt_1 \\
&= \mathbf{q}_n \int_0^\tau \frac{d\mathbf{r}(\Delta t + t_1)}{dt_1} dt_1 \\
&= \mathbf{q}_n \int_0^\tau \mathbf{v}(\Delta t + t_1) dt_1 \\
&= \sum_{\alpha} q_n^{\alpha} \int_0^\tau v^{\alpha}(\Delta t + t_1) dt_1,
\end{aligned} \tag{1.2.7}$$

where the index notation is used here as  $\alpha = 1, 2, 3$  for the value of 1 stands for x, 2 for y, and 3 for z.

Follow the cumulant expansion and have

$$\overline{e^{i\Phi}}' \approx e^{-\overline{\Phi^2}'/2}. \tag{1.2.8}$$

Next, we seek solutions to  $\overline{\Phi^2}'$  and have, for any  $t_1, t_2 \neq \infty$ ,

$$\begin{aligned}
e^{-\overline{\Phi_n(t_1, \tau)^2}'/2} &= \overline{e^{i\Phi_n(t_2, \tau)}}' \\
&= \overline{e^{i\Phi_n(t_2, \tau)}}' \\
&= e^{-\overline{\Phi_n(t_2, \tau)^2}'/2}, \\
\implies \overline{\Phi_n(t_1, \tau)^2}' &= \overline{\Phi_n(t_2, \tau)^2}', \\
\implies \overline{\Phi_n(\Delta t + \tau, \tau)^2}' &= \overline{\Phi_n(0, \tau)^2}'.
\end{aligned}$$

Then

$$\begin{aligned}
\overline{\Phi_n(0, \tau)^2}' &= \overline{\Phi_n(\Delta t + \tau, \tau)^2}' = \overline{\sum_{\alpha} q_n^{\alpha} \int_0^\tau v^{\alpha}(\Delta t + t_1) dt_1 \sum_{\beta} q_n^{\beta} \int_0^\tau v^{\beta}(\Delta t + t_2) dt_2}' \\
&= \sum_{\alpha, \beta} q_n^{\alpha} q_n^{\beta} \int_0^\tau dt_1 \int_0^\tau dt_2 \overline{v^{\alpha}(\Delta t + t_1) v^{\beta}(\Delta t + t_2)}'
\end{aligned}$$

for the same notation on  $\alpha$  applied to  $\beta$ . The quality of being arbitrary of  $\Delta t$  also allows to claim that  $\overline{v^{\alpha}(t_1) v^{\beta}(t_2)} = \overline{v^{\alpha}(t_3) v^{\beta}(t_4)}$  for  $t_1 - t_2 = t_3 - t_4$ . Hence

$$\begin{aligned}
\overline{\Phi_n(0, \tau)^2}' &= \sum_{\alpha, \beta} q_n^{\alpha} q_n^{\beta} \int_0^\tau dt_1 \int_0^\tau dt_2 \overline{v^{\alpha}(t_1) v^{\beta}(t_2)}' \\
&= \sum_{\alpha, \beta} q_n^{\alpha} q_n^{\beta} \int_0^\tau dt_1 \int_0^\tau dt_2 \overline{v^{\alpha}(t_1 - t_2) v^{\beta}(0)}' \\
&= \sum_{\alpha, \beta} q_n^{\alpha} q_n^{\beta} \lim_{n \rightarrow \infty} \sum_{i=0}^n \sum_{j=0}^n \delta t_1 \delta t_2 \overline{v^{\alpha}(i \delta t_1 - j \delta t_2) v^{\beta}(0)}'
\end{aligned}$$

for  $n\delta t_1 = n\delta t_2 = \tau$ , in which the integral identity  $\int_a^b f(x)dx = \lim_{n \rightarrow \infty} \sum_{i=0}^n \Delta x f(a + i\Delta x)$  for  $n\Delta x = b$  was applied. Since  $t_1$  and  $t_2$  are being integrated over the same boundaries, we can have  $\delta t_1 = \delta t_2 = \delta t$ . Then,

$$\begin{aligned}\overline{\Phi_n(0, \tau)^2}' &= \sum_{\alpha, \beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \sum_{i=0}^n \sum_{j=0}^n (\delta t)^2 \overline{v^\alpha(i\delta t - j\delta t) v^\beta(0)}' \\ &= \sum_{\alpha, \beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \sum_{i=0}^n \sum_{j=0}^n (\delta t)^2 \overline{v^\alpha((i-j)\delta t) v^\beta(0)}' .\end{aligned}$$

To proceed, note that  $(i+m) - (j+m) = i-j$  for any integer  $m$ . Hence, there can be  $n - |i-j|$  same values of  $\overline{v^\alpha((i-j)\delta t) v^\beta(0)}$  in the expression above. Let  $k = i-j$ , we have

$$\begin{aligned}\overline{\Phi_n(0, \tau)^2}' &= \sum_{\alpha, \beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \sum_{k=-n}^n (\delta t)^2 (n - |k|) \overline{v^\alpha(k\delta t) v^\beta(0)}' \\ &= \sum_{\alpha, \beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \left[ \sum_{k=0}^n (\delta t)^2 (n - k) \overline{v^\alpha(k\delta t) v^\beta(0)}' + \sum_{k=0}^n (\delta t)^2 (n - k) \overline{v^\alpha(-k\delta t) v^\beta(0)}' \right. \\ &\quad \left. - (\delta t)^2 n \overline{v^\alpha(0) v^\beta(0)}' \right],\end{aligned}$$

where the last term is to balance out the duplicated  $k = 0$  part in the two previous summations over  $k$ . This term will be dropped due to smallness as it approaches zero. Then

$$\begin{aligned}\overline{\Phi_n(0, \tau)^2}' &= \sum_{\alpha < \beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \left[ \sum_{k=0}^n (\delta t)^2 (n - k) \overline{v^\alpha(k\delta t) v^\beta(0)}' + \sum_{k=0}^n (\delta t)^2 (n - k) \overline{v^\alpha(-k\delta t) v^\beta(0)}' \right. \\ &\quad \left. + \sum_{k=0}^n (\delta t)^2 (n - k) \overline{v^\beta(k\delta t) v^\alpha(0)}' + \sum_{k=0}^n (\delta t)^2 (n - k) \overline{v^\beta(-k\delta t) v^\alpha(0)}' \right] \\ &\quad + \sum_{\alpha = \beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \left[ \sum_{k=0}^n (\delta t)^2 (n - k) \overline{v^\alpha(k\delta t) v^\beta(0)}' + \sum_{k=0}^n (\delta t)^2 (n - k) \overline{v^\alpha(-k\delta t) v^\beta(0)}' \right].\end{aligned}$$

Since  $\overline{v^\alpha(-k\delta t)v^\beta(0)} = \overline{v^\alpha(0)v^\beta(k\delta t)} = \overline{v^\beta(k\delta t)v^\alpha(0)}$ , with the same applicable to the reversed order of  $\alpha$  and  $\beta$ , we have

$$\begin{aligned}
\overline{\Phi_n(0,\tau)^2}' &= \sum_{\alpha<\beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \left[ \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\alpha(k\delta t)v^\beta(0)}' + \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\beta(k\delta t)v^\alpha(0)}' \right. \\
&\quad \left. + \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\beta(k\delta t)v^\alpha(0)}' + \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\alpha(k\delta t)v^\beta(0)}' \right] \\
&\quad + \sum_{\alpha=\beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \left[ \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\alpha(k\delta t)v^\beta(0)}' + \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\beta(k\delta t)v^\alpha(0)}' \right] \\
&= \sum_{\alpha<\beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \left[ 2 \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\alpha(k\delta t)v^\beta(0)}' + 2 \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\beta(k\delta t)v^\alpha(0)}' \right] \\
&\quad + \sum_{\alpha=\beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \left[ 2 \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\alpha(k\delta t)v^\beta(0)}' \right] \\
&= \sum_{\alpha,\beta} q_n^\alpha q_n^\beta \lim_{n \rightarrow \infty} \left[ 2 \sum_{k=0}^n (\delta t)^2 (n-k) \overline{v^\alpha(k\delta t)v^\beta(0)}' \right].
\end{aligned}$$

Let  $t = k\delta t$ , the equation above reduces to:

$$\overline{\Phi_n(0,\tau)^2}' = 2 \sum_{\alpha,\beta} q_n^\alpha q_n^\beta \int_0^\tau dt (\tau-t) \overline{v^\alpha(t)v^\beta(0)}'. \quad (1.2.9)$$

The speed-speed correlation between the initial speed and the speed after time  $t$  under the velocity relaxation rate or the collision rate  $\gamma$  is given by

$$\overline{v^\alpha(t)v^\beta(0)}' \approx \delta_{\alpha\beta} v_{th}^2 e^{-\gamma|t|}, \quad (1.2.10)$$

where  $v_{th}$  is the average speed of Maxwell-Boltzmann velocity distribution. This gives

$$\begin{aligned}
\overline{\Phi_n(0,\tau)^2}' &= 2 \sum_{\alpha,\beta} q_n^\alpha q_n^\beta \int_0^\tau dt (\tau-t) \delta_{\alpha\beta} v_{th}^2 e^{-\gamma|t|} \\
&= 2q_n^2 v_{th}^2 \int_0^\tau dt (\tau-t) e^{-\gamma t} \\
&= 2q_n^2 v_{th}^2 \int_0^\tau dt \tau e^{-\gamma t} + \int_0^\tau (-t) dt e^{-\gamma t} \\
&= 2q_n^2 v_{th}^2 \left[ -\frac{\tau}{\gamma} e^{-\gamma t} \right]_0^\tau + \left[ \frac{t}{\gamma} e^{-\gamma t} \right]_0^\tau - \int_0^\tau \frac{1}{\gamma} e^{-\gamma t} \\
&= 2q_n^2 v_{th}^2 \left( -\frac{\tau}{\gamma} e^{-\gamma \tau} + \frac{\tau}{\gamma} + \frac{\tau}{\gamma} e^{-\gamma \tau} + \frac{1}{\gamma^2} e^{-\gamma \tau} - \frac{1}{\gamma^2} \right) \\
&= \frac{2q_n^2 v_{th}^2}{\gamma^2} (\gamma \tau + e^{-\gamma \tau} - 1) \\
&= \frac{2q_n^2 v_{th}^2}{\gamma^2} H(\gamma \tau),
\end{aligned}$$

where  $H(x) \equiv x + e^{-x} - 1$ .

### iii> Solution to $S_1$

The above subsection gives  $\overline{\Phi_1(0, \tau)^2}' = \frac{2q_1^2 v_{th}^2}{\gamma^2} H(\gamma\tau)$ . Substitute back in Eq.(1.2.8) and in Eq.(1.2.6),

$$\begin{aligned} S_1 &= \text{Re} \int d\tau e^{-i(\Delta_1 - i\gamma_1)\tau} e^{-\frac{q_1^2 v_{th}^2}{\gamma^2} H(\gamma\tau)} \\ &= \int d\tau e^{-\gamma_1\tau - \frac{\Gamma_D^2}{\gamma^2} H(\gamma\tau)} \cos(\Delta_1\tau), \end{aligned} \quad (1.2.11)$$

where  $\Gamma_D \equiv q_1 v$ .

One can end the discussion for  $S_1$  here for the fact that  $S_1$  is only the description of the one-photon absorption case. While for the two-photon EIT case, the following sections which take some important math results from above are what would determine the EIT spectrum.

### 1.2.3 $S_2$

#### i> Preliminary Solution to $S_2$

Similarly, from Eq.(1.2.4b)

$$\begin{aligned} S_2 &= \text{Im}(-i) |\chi_{32}|^2 \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \\ &\quad \times e^{-i\mathbf{q}_1 \cdot \mathbf{r}(t) + i\omega_1 t} e^{(-i\omega_{31} - \gamma_1)(t - t_1) + i\mathbf{q}_2 \cdot \mathbf{r}(t_1) - i\omega_2 t_1} \\ &\quad \times e^{(-i\omega_{21} - \gamma_{21})(t_1 - t_2) - i\mathbf{q}_2 \cdot \mathbf{r}(t_2) + i\omega_2 t_2} e^{(-i\omega_{31} - \gamma_1)(t_2 - t_3) + i\mathbf{q}_1 \cdot \mathbf{r}(t_3) - i\omega_1 t_3} \\ &= -|\chi_{32}|^2 \text{Re} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \\ &\quad \times \exp\{[-i\mathbf{q}_1 \cdot \mathbf{r}(t) + i\omega_1 t + (-i\omega_{31} - \gamma_1)(t - t_1) + i\mathbf{q}_2 \cdot \mathbf{r}(t_1) - i\omega_2 t_1 \\ &\quad + (-i\omega_{21} - \gamma_{21})(t_1 - t_2) - i\mathbf{q}_2 \cdot \mathbf{r}(t_2) + i\omega_2 t_2 + (-i\omega_{31} - \gamma_1)(t_2 - t_3) \\ &\quad + i\mathbf{q}_1 \cdot \mathbf{r}(t_3) - i\omega_1 t_3] + i\omega_1 t_1 - i\omega_1 t_1 + i\omega_1 t_2 - i\omega_1 t_2\} \\ &= -|\chi_{32}|^2 \text{Re} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \\ &\quad \times e^{(i\Delta_1 - \gamma_1)(t - t_1 + t_2 - t_3)} e^{(i\Delta_R - \gamma_{21})(t_1 - t_2)} e^{i\mathbf{q}_2 \cdot [\mathbf{r}(t_1) - \mathbf{r}(t_2)] - i\mathbf{q}_1 \cdot [\mathbf{r}(t) - \mathbf{r}(t_3)]}, \end{aligned}$$

where  $\Delta_R \equiv \omega_1 - \omega_{21} - \omega_2$ .

#### ii> 3-D Changing of Variables

Next, to facilitate the calculation, change the coordinate basis from  $\{t_1, t_2, t_3\}$  to  $\{\tau, \tau_1, \tau_2\}$  such that

$$\begin{aligned} \tau &= t_1 - t_2, \\ \tau_1 &= t - t_1 + t_2 - t_3, \\ \text{and } \tau_2 &= t - t_1. \end{aligned} \quad (1.2.12)$$



It can be easily shown, e.g., converting to matrix and find the Reduced Row Echelon form to be an identity, that the newly-defined parameters form a linearly independent set, which qualifies them to form a basis. The integrand becomes

$$e^{(i\Delta_1 - \gamma_1)\tau_1} e^{(i\Delta_R - \gamma_{21})\tau} e^{i\mathbf{q}_2 \cdot [\mathbf{r}(t - \tau_2) - \mathbf{r}((t - \tau_2) - \tau)] - i\mathbf{q}_1 \cdot [\mathbf{r}(t) - \mathbf{r}(t - (\tau + \tau_1))]}.$$

To determine the integration "volume" elements, we first calculate the Jacobian  $J \equiv \frac{\partial(\tau, \tau_1, \tau_2)}{\partial(t_1, t_2, t_3)}$  of the transformation. We have

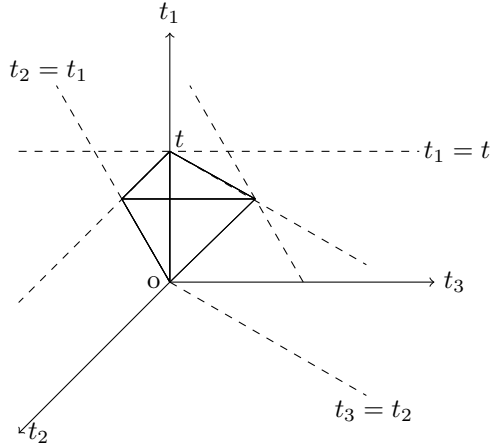
$$\begin{aligned} J &= \det \begin{bmatrix} \partial\tau/\partial t_1 & \partial\tau/\partial t_2 & \partial\tau/\partial t_3 \\ \partial\tau_1/\partial t_1 & \partial\tau_1/\partial t_2 & \partial\tau_1/\partial t_3 \\ \partial\tau_2/\partial t_1 & \partial\tau_2/\partial t_2 & \partial\tau_2/\partial t_3 \end{bmatrix} \\ &= \begin{vmatrix} 1 & -1 & 0 \\ -1 & 1 & -1 \\ -1 & 0 & 0 \end{vmatrix} \\ &= -1, \end{aligned}$$

which gives  $|J| = 1$ . Hence, the volume element of the new coordinates is  $|J|d\tau d\tau_1 d\tau_2 = d\tau d\tau_1 d\tau_2$ .

To form a corresponding set of integration limits, we first find the original integration domain represented in t-space:

$$\begin{aligned} 0 &\leq t_1 \leq t \\ 0 &\leq t_2 \leq t_1 \\ 0 &\leq t_3 \leq t_2. \end{aligned} \tag{1.2.12b}$$

This integration domain in the original coordinate system is drawn below. Note that this domain is equivalent to the volume integration of  $\int_0^t \int_0^{t_1} \int_0^{t_2} dt_1 dt_2 dt_3$ , whose multiplication with the integrand is the original integral.



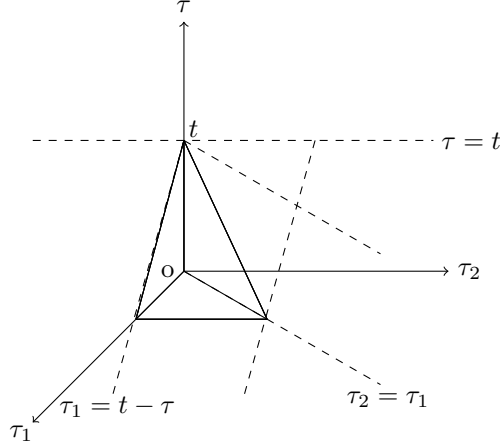
The new coordinate variables are a linear transformation from the old ones and, as aforementioned, are mutually linearly independent, which is also a required condition, allowing one to build a mutually orthogonal set of axes measuring the same object.

Next is to find where the new integration domain is situated in the new coordinate system, or,

$\tau$ -space. Note that the domain in  $t$ -space (the geometric structure drawn above) is a tetrahedron (a convex polyhedron). A convex polyhedron under a linear transformation is mapped to another convex polyhedron. A convex polyhedron can be uniquely defined by its vertices (also known as the generators in geometry theory). Hence, We first transform the four vertices of the integration domain in  $t$ -space to  $\tau$ -space per Eq. (1.2.12) in coordinate representations, i.e., from  $(t_1, t_2, t_3) \longrightarrow (\tau, \tau_1, \tau_2)$ , and have

$$\begin{aligned}(t, t, t) &\longrightarrow (0, 0, 0) \\ (0, 0, 0) &\longrightarrow (0, t, t) \\ (t, t, 0) &\longrightarrow (0, t, 0) \\ (t, 0, 0) &\longrightarrow (t, 0, 0).\end{aligned}$$

Since the new polyhedron must be also convex, the lines connecting these new points must also be in the new polyhedron. So we connect them and draw it as below



Note that this structure also builds a tetrahedron. With an easy comparison, this image takes the exact geometric structure as the original one (but with a different orientation in the new space). Since this is a new volume integral, and a volume integral must be a bounded shape, all the points inside this tetrahedron must be in the new integration domain. Since the volume of this tetrahedron is the same as the original one, we know this tetrahedron is exactly the new integration domain in  $\tau$ -space since the magnitude of the Jacobian is unity.

The volume encompassed by the edge lines of the tetrahedron is the new integration domain. One can easily extract from the plot the limits bounding the domain -  $\tau$ :  $0 \rightarrow t$ ,  $\tau_1$ :  $0 \rightarrow t - \tau$ , and  $\tau_2$ :  $0 \rightarrow \tau_1$ . Hence, the new integral limits writes  $\int_0^t \int_0^{t-\tau} \int_0^{\tau_1}$ , leading to the new  $S_2$ :

$$\begin{aligned}S_2 &= -|\chi_{32}|^2 \text{Re} \lim_{T \rightarrow \infty} \int_0^T \frac{dt}{T} \int_0^t d\tau \int_0^{t-\tau} d\tau_1 \int_0^{\tau_1} d\tau_2 \\ &\quad \times e^{-\gamma_1 \tau_1} e^{(i\Delta_R - \gamma_{21})\tau} e^{i\mathbf{q}_2 \cdot [\mathbf{r}(t-\tau_2) - \mathbf{r}((t-\tau_2)-\tau)] - i\mathbf{q}_1 \cdot [\mathbf{r}(t) - \mathbf{r}(t-(\tau+\tau_1))]} \\ &= -|\chi_{32}|^2 \text{Re} \int_0^t d\tau \int_0^{t-\tau} d\tau_1 \int_0^{\tau_1} d\tau_2 \\ &\quad \times e^{-\gamma_1 \tau_1} e^{(i\Delta_R - \gamma_{21})\tau} \overline{e^{i[\Phi_2(t-\tau_2, \tau) - \Phi_1(t, \tau_1 + \tau)]}},\end{aligned}$$

in which  $\Delta_1$  was taken to be zero.

Since both the integrals over  $\tau$  and  $\tau_1$  contain exponentially decay terms, we set the decay term boundaries to infinity and get

$$S_2 \approx -|\chi_{32}|^2 \text{Re} \int_0^\infty d\tau \int_0^\infty d\tau_1 \int_0^{\tau_1} d\tau_2 \\ \times e^{-\gamma_1 \tau_1} e^{(i\Delta_R - \gamma_{21})\tau} \overline{e^{i\Theta}}, \quad (1.2.13)$$

where  $\Theta \equiv \Phi_2(t - \tau_2, \tau) - \Phi_1(t, \tau_1 + \tau)$ .

### iii> The $\overline{e^{i\Theta}}$ Term

Follow the same cumulant expansion as in  $S_1$  and have

$$\overline{e^{i\Theta}} \approx e^{-\overline{\Theta^2}/2}. \quad (1.2.14)$$

Then

$$\begin{aligned} \overline{\Theta^2} &= \overline{\Phi_2(t - \tau_2, \tau)^2 + \Phi_1(t, \tau_1 + \tau)^2 - 2\Phi_2(t - \tau_2, \tau)\Phi_1(t, \tau_1 + \tau)} \\ &= \overline{\Phi_2(t - \tau_2, \tau)^2} + \overline{\Phi_1(t, \tau_1 + \tau)^2} - 2\overline{\Phi_2(t - \tau_2, \tau)\Phi_1(t, \tau_1 + \tau)} \\ &= \overline{\Phi_2(0, \tau)^2}' + \overline{\Phi_1(0, \tau_1 + \tau)^2}' - 2\overline{\Phi_2(-\tau_2, \tau)\Phi_1(0, \tau_1 + \tau)}'. \end{aligned} \quad (1.2.15)$$

From section 1.2.3 ii> we have

$$\overline{\Phi_2(0, \tau)^2}' = \frac{2q_2^2 v_{th}^2}{\gamma^2} H(\gamma\tau), \quad (1.2.16)$$

$$\overline{\Phi_1(0, \tau_1 + \tau)^2}' = \frac{2q_1^2 v_{th}^2}{\gamma^2} H(\gamma(\tau_1 + \tau)). \quad (1.2.17)$$

And for the last term,

$$\begin{aligned}
\overline{\Phi_2(-\tau_2, \tau)\Phi_1(0, \tau_1 + \tau)}' &= \overline{\mathbf{q}_2[\mathbf{r}(-\tau_2) - \mathbf{r}(-\tau_2 - \tau)]\mathbf{q}_1[\mathbf{r}(0) - \mathbf{r}(-\tau_1 - \tau)]}' \\
&= \overline{\sum_{\alpha} q_n^{\alpha} \int_{-\tau_1 - \tau}^0 v^{\alpha}(t_1) dt_1 \sum_{\beta} q_2^{\beta} \int_{-\tau_2 - \tau}^{-\tau_2} v^{\beta}(t_2) dt_2}' \\
&= \sum_{\alpha, \beta} q_n^{\alpha} q_2^{\beta} \int_{-\tau_1 - \tau}^0 dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 \overline{v^{\alpha}(t_1) v^{\beta}(t_2)}' \\
&= \sum_{\alpha, \beta} q_n^{\alpha} q_2^{\beta} \int_{-\tau_1 - \tau}^0 dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 \overline{v^{\alpha}(t_1 - t_2) v^{\beta}(0)}' \\
&= \sum_{\alpha, \beta} \delta_{\alpha\beta} q_n^{\alpha} q_2^{\beta} \int_{-\tau_1 - \tau}^0 dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 v_{th}^2 e^{-\gamma|t_1 - t_2|} \\
&= \mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2 \int_{-\tau_1 - \tau}^0 dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 e^{-\gamma|t_1 - t_2|}.
\end{aligned}$$

To solve the integral, note that terms with  $\tau_2$  is integrated from 0 to  $\tau_1$  as in [Eq.\(1.2.13\)](#). Hence,  $\tau_2 < \tau_1$ . Since  $\tau$ 's are positive, we have  $-\tau_1 - \tau < -\tau_2 - \tau < \tau_2 < 0$ . Then,

$$\begin{aligned}
\overline{\Phi_2(-\tau_2, \tau)\Phi_1(0, \tau_1 + \tau)}' &= \mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2 \left[ \int_{-\tau_2}^0 dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 e^{-\gamma(t_1 - t_2)} + \int_{-\tau_1 - \tau}^{-\tau_2 - \tau} dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 e^{-\gamma(t_2 - t_1)} \right. \\
&\quad \left. + \int_{-\tau_2 - \tau}^{-\tau_2} dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 e^{-\gamma|t_1 - t_2|} \right] \\
&= \mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2 \left\{ \left( \frac{1}{\gamma^2} \right) \left[ -e^{-\gamma\tau_2} + e^{-\gamma(\tau_2 + \tau)} - e^{-\gamma\tau} - 1 \right. \right. \\
&\quad \left. \left. - e^{-\gamma\tau} + e^{-\gamma(\tau_1 + \tau - \tau_2)} - e^{-\gamma(\tau_1 - \tau_2)} - 1 \right] \right. \\
&\quad \left. + \int_{-\tau_2 - \tau}^{-\tau_2} dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 e^{-\gamma|t_1 - t_2|} \right\} \\
&= \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma^2} \left[ (e^{-\gamma\tau} - 1)(e^{-\gamma\tau_1 + \gamma\tau_2} + e^{-\gamma\tau_2} - 2) \right. \\
&\quad \left. + \gamma^2 \int_{-\tau_2 - \tau}^{-\tau_2} dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 e^{-\gamma|t_1 - t_2|} \right] \tag{1.2.18}
\end{aligned}$$

To proceed, we convert the last term to its summation form and have, for  $-\tau_2 - \tau + n\delta t = -\tau_2 \implies n\delta t = \tau$ ,

$$\begin{aligned}
\int_{-\tau_2 - \tau}^{-\tau_2} dt_1 \int_{-\tau_2 - \tau}^{-\tau_2} dt_2 e^{-\gamma|t_1 - t_2|} &= \lim_{n \rightarrow \infty} \sum_{i, j=0}^n (\delta t)^2 e^{-\gamma|[-(\tau_2 - \tau) + i\delta t] - [-(\tau_2 - \tau) + j\delta t]|} \\
&= \lim_{n \rightarrow \infty} \sum_{i, j=0}^n (\delta t)^2 e^{-\gamma\delta t|i - j|}.
\end{aligned}$$

Recall the same reasoning used previously that since  $(i + m) - (j + m) = i - j$  for any integer  $m$ ,

there can be  $n - |i - j|$  same values of  $e^{-\gamma\delta t|i-j|}$  in the expression above. Let  $k = i - j$  and have

$$\begin{aligned} \int_{-\tau_2-\tau}^{-\tau_2} dt_1 \int_{-\tau_2-\tau}^{-\tau_2} dt_2 e^{-\gamma|t_1-t_2|} &= \lim_{n \rightarrow \infty} \sum_{k=-n}^n (\delta t)^2 (n - |k|) e^{-\gamma\delta t|k|} \\ &= 2 \lim_{n \rightarrow \infty} \sum_{k=0}^n (\delta t)^2 (n - |k|) e^{-\gamma\delta t|k|} - n(\delta t)^2 \\ &= 2 \lim_{n \rightarrow \infty} \sum_{k=0}^n (\delta t)^2 (n - k) e^{-\gamma\delta tk}, \end{aligned}$$

in which we dropped the infinitesimally small term  $n(\delta t)^2$  stemming from balancing the duplicate  $k = 0$  term. Let  $k\delta t = t$ , we have

$$\begin{aligned} \int_{-\tau_2-\tau}^{-\tau_2} dt_1 \int_{-\tau_2-\tau}^{-\tau_2} dt_2 e^{-\gamma|t_1-t_2|} &= 2 \int_0^\tau dt (\tau - t) e^{-\gamma t} \\ &= 2 \frac{1}{\gamma^2} (\gamma\tau - 1 + e^{-\gamma\tau}). \end{aligned}$$

Plug this back into [Eq.\(1.2.18\)](#), we have

$$\begin{aligned} \overline{\Phi_2(-\tau_2, \tau) \Phi_1(0, \tau_1 + \tau)}' &= \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma^2} [(e^{-\gamma\tau} - 1)(e^{-\gamma\tau_1 + \gamma\tau_2} + e^{-\gamma\tau_2} - 2) \\ &\quad + 2(\gamma\tau - 1 + e^{-\gamma\tau})]. \end{aligned} \quad (1.2.19)$$

#### iv> Solution to $S_2$

Plug [Eq.\(1.2.19\)](#), [Eq.\(1.2.16\)](#), and [Eq.\(1.2.17\)](#) into [Eq.\(1.2.15\)](#), and further into [Eq.\(1.2.14\)](#) and then [Eq.\(1.2.13\)](#),

$$\begin{aligned} S_2 &= -|\chi_{32}|^2 \text{Re} \int d\tau \int d\tau_1 \int d\tau_2 e^{-\gamma_1 \tau_1} e^{(i\Delta_R - \gamma_{21})\tau} \\ &\quad \times EXP \left\{ -\frac{1}{2} \left[ \frac{2q_2^2 v_{th}^2}{\gamma^2} (\gamma\tau - 1 + e^{-\gamma\tau}) + \frac{2q_1^2 v_{th}^2}{\gamma^2} (\gamma\tau_1 + \gamma\tau - 1 + e^{-\gamma\tau_1} e^{-\gamma\tau}) \right. \right. \\ &\quad \left. \left. - 2 \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma^2} [(e^{-\gamma\tau} - 1)(e^{-\gamma\tau_1 + \gamma\tau_2} + e^{-\gamma\tau_2} - 2) + 2(\gamma\tau - 1 + e^{-\gamma\tau})] \right] \right\}. \end{aligned}$$

Define  $\Gamma_D^{res} = |\mathbf{q}_1 - \mathbf{q}_2| v_{th}$ , then  $(\Gamma_D^{res})^2 = v_{th}^2 (q_1^2 + q_2^2 - 2\mathbf{q}_1 \cdot \mathbf{q}_2) \implies q_2^2 v_{th}^2 = (\Gamma_D^{res})^2 - \Gamma_D^2 + 2\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2$ .

We have

$$\begin{aligned}
S_2 &= -|\chi_{32}|^2 \text{Re} \int d\tau \int d\tau_1 \int d\tau_2 e^{-\gamma_1 \tau_1} e^{(i\Delta_R - \gamma_{21})\tau} \\
&\quad \times EXP \left\{ -\frac{(\Gamma_D^{res})^2}{\gamma^2} (\gamma\tau - 1 + e^{-\gamma\tau}) - \frac{\Gamma_D^2}{\gamma^2} (\gamma\tau_1 + e^{-\gamma\tau_1} e^{-\gamma\tau} - e^{-\gamma\tau}) - \frac{2\mathbf{q}_1 \cdot \mathbf{q}_2}{\gamma^2} v_{th}^2 (\gamma\tau - 1 + e^{-\gamma\tau}) \right. \\
&\quad \left. + \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma^2} (e^{-\gamma\tau} - 1) (e^{-\gamma\tau_1 + \gamma\tau_2} + e^{-\gamma\tau_2} - 2) + 2 \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma^2} (\gamma\tau - 1 + e^{-\gamma\tau}) \right\} \\
&= -|\chi_{32}|^2 \text{Re} \int_0^\infty d\tau e^{(i\Delta_R - \gamma_{21})\tau} e^{-\frac{(\Gamma_D^{res})^2}{\gamma^2} H(\gamma\tau)} \\
&\quad \times \int_0^\infty d\tau_1 e^{-\gamma_1 \tau_1} e^{-\frac{\Gamma_D^2}{\gamma^2} (\gamma\tau_1 + e^{-\gamma\tau_1} e^{-\gamma\tau} - e^{-\gamma\tau})} \\
&\quad \times \int_0^{\tau_1} d\tau_2 e^{\frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma^2} (e^{-\gamma\tau} - 1) (e^{-\gamma\tau_1} e^{\gamma\tau_2} + e^{-\gamma\tau_2} - 2)}.
\end{aligned}$$

Approximations: for the Doppler regime we take  $e^{-\gamma\tau_1} \approx 1 - \gamma\tau_1$  and for the Dicke regime we take  $e^{-\gamma\tau} \ll 1$ . Then, since  $\tau_2 < \tau_1$ ,  $e^{-\gamma\tau_2} \approx 1 - \gamma\tau_2$ ; the Dicke regime also gives  $H(\gamma\tau) \approx \gamma\tau$ . Then

$$\begin{aligned}
S_2 &\approx -|\chi_{32}|^2 \text{Re} \int_0^\infty d\tau e^{(i\Delta_R - \gamma_{21})\tau} e^{-\frac{(\Gamma_D^{res})^2}{\gamma^2} (\gamma\tau)} \\
&\quad \times \int_0^\infty d\tau_1 e^{-\gamma_1 \tau_1} e^{-\frac{\Gamma_D^2}{\gamma^2} (\gamma\tau_1 + (1 - \gamma\tau_1) e^{-\gamma\tau} - e^{-\gamma\tau})} \\
&\quad \times \int_0^{\tau_1} d\tau_2 e^{-\frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma^2} [(1 - \gamma\tau_1)(1 + \gamma\tau_2) + (1 - \gamma\tau_2) - 2]} \\
&\approx -|\chi_{32}|^2 \int_0^\infty d\tau e^{-\gamma_{21}\tau} e^{-\frac{(\Gamma_D^{res})^2}{\gamma^2} (\gamma\tau)} \cos(\Delta_R \tau) \\
&\quad \times \int_0^\infty d\tau_1 e^{-\gamma_1 \tau_1} e^{-\frac{\Gamma_D^2}{\gamma^2} (\gamma\tau_1)} \int_0^{\tau_1} d\tau_2 e^{\frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma^2} \gamma\tau_1 (1 + \gamma\tau_2)} \\
&= -|\chi_{32}|^2 \frac{\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}}{\Delta_R^2 + [\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}]^2} \int_0^\infty d\tau_1 e^{-(\gamma_1 + \frac{\Gamma_D^2}{\gamma})\tau_1} \times \frac{e^{\frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2 \tau_1}{\gamma}}}{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2 \tau_1} (e^{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2 \tau_1^2} - 1)
\end{aligned}$$

To proceed, perform Taylor expansion on the parenthesized term and have

$$\begin{aligned}
S_2 &= -|\chi_{32}|^2 \frac{\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}}{\Delta_R^2 + [\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}]^2} \int_0^\infty d\tau_1 \frac{e^{-(\gamma_1 + \frac{\Gamma_D^2}{\gamma} - \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma})\tau_1}}{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2 \tau_1} \\
&\quad \times \left( -1 + 1 + \mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2 \tau_1^2 + \frac{(\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2)^2 \tau_1^4}{2} + \dots \right) \\
&= -|\chi_{32}|^2 \frac{\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}}{\Delta_R^2 + [\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}]^2} \left[ \int_0^\infty d\tau_1 e^{-(\gamma_1 + \frac{\Gamma_D^2}{\gamma} - \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma})\tau_1} \tau_1 \right. \\
&\quad \left. + \int_0^\infty d\tau_1 e^{-(\gamma_1 + \frac{\Gamma_D^2}{\gamma} - \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma})\tau_1} \mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2 \tau_1^3 + \dots \right] \\
&= -|\chi_{32}|^2 \frac{\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}}{\Delta_R^2 + [\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}]^2} \left[ \frac{1}{[\gamma_1 + \frac{\Gamma_D^2}{\gamma} - \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma}]^2} + \frac{6\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{[\gamma_1 + \frac{\Gamma_D^2}{\gamma} - \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma}]^4} + \dots \right].
\end{aligned}$$

The second and further terms in the brackets are sufficiently small compared to the first. We approximate to the first order and have

$$\begin{aligned}
S_2 &\approx \frac{\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}}{\Delta_R^2 + [\gamma_{21} + \frac{(\Gamma_D^{res})^2}{\gamma}]^2} \frac{-|\chi_{32}|^2}{[\gamma_1 + \frac{\Gamma_D^2}{\gamma} - \frac{\mathbf{q}_1 \cdot \mathbf{q}_2 v_{th}^2}{\gamma}]^2} \\
&= \frac{-|\chi_{32}|^2}{[\gamma_1 + \mathbf{q}_1 \cdot (\mathbf{q}_1 - \mathbf{q}_2) v_{th}^2 / \gamma]^2} \frac{\gamma_{21} + \eta \Gamma_D^{res}}{\Delta_R^2 + [\gamma_{21} + \eta \Gamma_D^{res}]^2}, \tag{1.2.20}
\end{aligned}$$

where,  $\eta = \frac{\Gamma_D^{res}}{\gamma}$ , and  $\Gamma_D^{res} = |\mathbf{q}_1 - \mathbf{q}_2| v_{th}$ . With all the parameters shown in Eq.(1.2.20) being measurable through experiments, this is the expression predicting the spectrum behavior of EIT in different experimental setups, including the angle between control and probe fields, which serves as the signature of Dicke narrowing.