APPENDIX-1

Zero Order Polarizations of Degenerate Double $\Lambda$ System

The zero order coherences under the perturbative approach are obtained as follows:

\[ \rho_{23}^{(0)} = -i\alpha_2[(\kappa_4\kappa_6 + \alpha_1^2)(\rho_{22}^{(0)} - \rho_{33}^{(0)}) - \alpha_2^2(\rho_{22}^{(0)} - \rho_{44}^{(0)})]/C_1, \]  
(A1.1)

\[ \rho_{42}^{(0)} = i\alpha_1[(\kappa_4\kappa_6 + \alpha_1^2)(\rho_{22}^{(0)} - \rho_{44}^{(0)}) - \alpha_2^2(\rho_{22}^{(0)} - \rho_{33}^{(0)})]/C_1, \]  
(A1.2)

\[ \rho_{43}^{(0)} = \alpha_1\alpha_2[\kappa_5(\rho_{22}^{(0)} - \rho_{33}^{(0)}) + \kappa_4^*(\rho_{22}^{(0)} - \rho_{44}^{(0)})]/C_1, \]  
(A1.3)

where

\[ C_1 = \kappa_4^*\kappa_6 + \alpha_1^2\kappa_5 + \alpha_2^2\kappa_4^*. \]  
(A1.4)

Coefficients $\kappa_i$, ($i=1\ldots6$) are as defined in Eq. (4.3). Similarly $\rho_{ii}^{(0)}$ are given as,

\[ \rho_{22}^{(0)} = \Gamma_1[\alpha_4^2\zeta_1^2 + (\gamma_3 + \alpha_2^2\gamma_2)(\gamma_4 + \alpha_2^2\gamma_3) - \gamma_3\gamma_4 - \alpha_1^2\alpha_2^2\zeta_1\Gamma_1]/C_2, \]  
(A1.5)

\[ \rho_{33}^{(0)} = \Gamma_1[\alpha_1^2(\zeta_3 - \alpha_2^2\zeta_1)\Gamma_3 + \alpha_2^2[\gamma_2(\zeta_3 - \gamma_2) + \alpha_1^2(\zeta_3 - \alpha_2^2\zeta_1)]]/C_2, \]  
(A1.6)

\[ \rho_{44}^{(0)} = \Gamma_1[\alpha_1^2(\zeta_4 - \alpha_2^2\zeta_1)\Gamma_4 + \alpha_2^2[\gamma_3(\zeta_4 - \gamma_3) + \alpha_1^2(\zeta_4 - \alpha_2^2\zeta_1)]]/C_2, \]  
(A1.7)

\[ \rho_{11}^{(0)} = 1 - \rho_{22}^{(0)} - \rho_{33}^{(0)} - \rho_{44}^{(0)}, \]  
(A1.8)

where

\[ \zeta_1 = \text{Re}(C_1)/|C_1|^2, \quad \zeta_2 = \text{Re}[(\kappa_4\kappa_6 + \alpha_2^2)/C_1], \quad \zeta_3 = \text{Re}[(\kappa_4^*\kappa_6 + \alpha_2^2)/C_1], \]  
(A1.9)

\[ C_2 = (\Gamma_1 + \Gamma_2)[\gamma_3\gamma_4 + \alpha_1^2\gamma_3\gamma_4 + \alpha_2^2\gamma_4\gamma_3 + \alpha_1^2\alpha_2^2(\zeta_3 - \zeta_1) - \Gamma_1\Gamma_3 + \alpha_2^2(\zeta_3 - \alpha_2^2\zeta_1)(\Gamma_1 + \gamma_1) + \alpha_1^2(\zeta_3 - \alpha_2^2\zeta_1)(\Gamma_1 + \gamma_1)]/C_2, \]  
(A1.10)

These coherences and populations are used in the analysis of Sec. 4.5 and 5.3.
Absorption and Dispersion in Degenerate Double $\Lambda$ System

Absorption and dispersion in a DDL system are related to real and imaginary components of $\tilde{P}$ (cf. Eq. (4.5)). Using Eq. (4.5) – (4.8) we obtain,

\[ \tilde{P} = (p_1 + ip_2)/(u_1 + iu_2), \quad (A2.1) \]

\[ p_1 = \alpha_1^2 \gamma_{31} + \alpha_2^2 \gamma_{41} + (\Gamma_{21} + \Gamma_{12})[\gamma_{31}(\gamma_4 + \Gamma_{12}) + \gamma_{41}(\gamma_3 + \Gamma_{12})] \]
\[ - (\delta_1 \gamma_{31} + \delta_2 \gamma_{41})(\delta_1 - \Delta_1) - \frac{\alpha_1 \alpha_2}{\beta_1 \beta_2}(\beta_1^2 \gamma_{31} + \beta_2^2 \gamma_{41}), \quad (A2.2) \]

\[ p_2 = (\Gamma_{12} + \Gamma_{21})(\delta_1 \gamma_{31} + \delta_2 \gamma_{41}) + [\gamma_{31}(\gamma_4 + \Gamma_{12}) + \gamma_{41}(\gamma_3 + \Gamma_{12})](\delta_1 - \Delta_1), \quad (A2.3) \]

\[ u_1 = \alpha_1^2 \delta_1 + \alpha_2^2 \delta_1 + (\delta_1 - \Delta_1)[\gamma_3 + \Gamma_{12}](\gamma_4 + \Gamma_{12}) - \delta \delta_2 \]
\[ + (\Gamma_{12} + \Gamma_{21})(\gamma_3 + \Gamma_{12})\delta_1 + (\gamma_4 + \Gamma_{12})\delta_2, \quad (A2.4) \]

\[ u_1 = (\delta_1 - \Delta_1)[\gamma_3 + \Gamma_{12}]\delta_1 + (\gamma_4 + \Gamma_{12})\delta_2 - \alpha_1^2(\gamma_3 + \Gamma_{12}) - \alpha_2^2(\gamma_4 + \Gamma_{12}) \]
\[ - (\Gamma_{12} + \Gamma_{21})(\gamma_3 + \Gamma_{12})(\gamma_4 + \Gamma_{12}) - \delta_1 \delta_2. \quad (A2.5) \]

Therefore absorption and dispersion can be obtained as follows:

\[ A = [(\delta_1 - \Delta_1)^2 \{(\gamma_4 + \Gamma_{12})\gamma_{41}[(\gamma_3 + \Gamma_{12})^2 + \delta_2^2] + (\gamma_3 + \Gamma_{12})\gamma_{31}[(\gamma_4 + \Gamma_{12})^2 + \delta_1^2]\}
\[ + (\delta_1 - \Delta_1)
\[ \left\{[(\gamma_3 + \Gamma_{12})\delta_1 + (\gamma_4 + \Gamma_{12})\delta_2] \frac{\alpha_1 \alpha_2}{\beta_1 \beta_2}(\beta_1^2 \gamma_{31} + \beta_2^2 \gamma_{41})
\[ - 2[\gamma_{31}(\gamma_3 + \Gamma_{12})\alpha_1 \delta_1 + \gamma_{41}(\gamma_4 + \Gamma_{12})\alpha_2 \delta_2]
\[ + (\Gamma_{12} + \Gamma_{21})^2 \{[(\gamma_4 + \Gamma_{12})\gamma_{41}[(\gamma_3 + \Gamma_{12})^2 + \delta_2^2] + (\gamma_3 + \Gamma_{12})\gamma_{31}[(\gamma_4 + \Gamma_{12})^2 + \delta_1^2]\}
\[ + (\Gamma_{12} + \Gamma_{21})^2
\[ \left\{\alpha_1^2 \gamma_{41}[(\gamma_3 + \Gamma_{12})^2 + \delta_2^2] + \alpha_2^2 \gamma_{31}[(\gamma_4 + \Gamma_{12})^2 + \delta_1^2] + 2(\gamma_3 + \Gamma_{12})(\gamma_4 + \Gamma_{12}) \right\}
\[ + (\Gamma_{12} + \Gamma_{21})^2
\[ \left\{\alpha_1^2 \gamma_{31} + \alpha_2^2 \gamma_{41}\right\} - [(\gamma_3 + \Gamma_{12})(\gamma_4 + \Gamma_{12}) - \delta_1 \delta_2] \frac{\alpha_1 \alpha_2}{\beta_1 \beta_2}(\beta_1^2 \gamma_{31} + \beta_2^2 \gamma_{41})
\[ + [\alpha_1^2(\gamma_3 + \Gamma_{12}) + \alpha_2^2(\gamma_4 + \Gamma_{12})] \left\{\alpha_1^2 \gamma_{31} + \alpha_2^2 \gamma_{41} - \frac{\alpha_1 \alpha_2}{\beta_1 \beta_2}(\beta_1^2 \gamma_{31} + \beta_2^2 \gamma_{41})\right\}\}/(u_1^2 + u_2^2), \quad (A2.6) \]
\[
\eta = [(\delta_1 - \Delta_1)^2 \{\alpha_2^2 \gamma_{41}[(\gamma_3 + \Gamma_{12})^2 + \delta_1^2] + \delta_2 \gamma_{31}[(\gamma_4 + \Gamma_{12})^2 + \delta_1^2]\} \\
- (\delta_1 - \Delta_1)^2 \left\{ \frac{\alpha_2^2 \gamma_{41}[(\gamma_3 + \Gamma_{12})^2 + \delta_1^2]}{\beta_1 \beta_2} \alpha_2 \gamma_{31}[(\gamma_4 + \Gamma_{12})^2 + \delta_1^2] \right\} \\
+ [(\gamma_3 + \Gamma_{12})(\gamma_4 + \Gamma_{12}) - \delta_1 \delta_2] \frac{\alpha_2 \alpha_2}{\beta_1 \beta_2} \left( \beta_1^2 \gamma_{31} + \beta_2^2 \gamma_{41} \right) \\
+ (\Gamma_{12} + \Gamma_{21})^2 \{\delta_1 \gamma_{41}[(\gamma_3 + \Gamma_{12})^2 + \delta_1^2] + \delta_2 \gamma_{31}[(\gamma_4 + \Gamma_{12})^2 + \delta_1^2]\} \\
+ (\Gamma_{12} + \Gamma_{21}) \left\{ 2[\alpha_2^2 \delta_1 \gamma_{41}(\gamma_3 + \Gamma_{12}) + \alpha_2^2 \delta_2 \gamma_{31}(\gamma_4 + \Gamma_{12})] \\
- [(\gamma_3 + \Gamma_{12})\delta_1 + (\gamma_4 + \Gamma_{12})\delta_2] \frac{\alpha_2 \alpha_2}{\beta_1 \beta_2} \left( \beta_1^2 \gamma_{31} + \beta_2^2 \gamma_{41} \right) \right\} \right) \\
+ (\alpha_2^2 \delta_2 + \alpha_2^2 \delta_1) \left\{ \frac{\alpha_2^2 \gamma_{31} + \alpha_2^2 \gamma_{41}}{\beta_1 \beta_2} - \frac{\alpha_2 \alpha_2}{\beta_1 \beta_2} \left( \beta_1^2 \gamma_{31} + \beta_2^2 \gamma_{41} \right) \right\} \right\} \right) / (u_1^2 + u_2^2),
\]

(A2.7)

\[
u_1^2 + u_2^2 = \{(\delta_1 - \Delta_1)[(\gamma_3 + \Gamma_{12})\delta_1 + (\gamma_4 + \Gamma_{12})\delta_2] - \alpha_2^2 (\gamma_3 + \Gamma_{12}) - \alpha_2^2 (\gamma_4 + \Gamma_{12}) \\
- (\Gamma_{12} + \Gamma_{21})[(\gamma_3 + \Gamma_{12})(\gamma_4 + \Gamma_{12}) - \delta_1 \delta_2] \right\}^2 \\
\left[ (\gamma_3 + \Gamma_{12})(\gamma_4 + \Gamma_{12}) - \delta_1 \delta_2 \right] + (\Gamma_{12} + \Gamma_{21})[(\gamma_3 + \Gamma_{12})\delta_1 + (\gamma_4 + \Gamma_{12})\delta_2] \right\}^2.
\]

(A2.8)

From equations (A2.6) – (A2.8) it is clear that at two-photon resonance condition \( \delta_1 = \Delta_1, \ A \rightarrow 0 \) and \( \eta \rightarrow 0 \). These expressions prove that susceptibility of DDL system remains finite though small at \( \delta_1 = \Delta_1 \), i.e., \( A \) does not go to exact zero at \( \delta_1 = \Delta_1 \) which causes the shift in EIT from exact two-photon resonance (cf. Sec. 4.5).
APPENDIX-3

Low Frequency Coherence in Degenerate Double $\Lambda$ System

Of particular interest in the analysis of AWI is the low frequency coherence $\rho_{21}^{(0)}$.

In the limit of weak excitation, $\rho_{11}^{(0)} \approx 1$ and $\rho_{22}^{(0)} = \rho_{33}^{(0)} = \rho_{44}^{(0)} \approx 0$, we may write

$$\rho_{21}^{(0)} \approx (\phi_1 + i\phi_2) / C,$$  \hspace{1cm} (A3.1)

where

$$\phi_1 = (-\alpha_1 \beta_1 \{[\delta_1^2 + (\gamma_3 + \Gamma_{12})]^2[\alpha_1^2 - \delta_1 (\delta_1 - \Delta_1) + (\Gamma_{12} + \Gamma_{21})(\gamma_4 + \Gamma_{12})]$$

$$- \alpha_2^2 [\delta_1 \delta_2 + (\gamma_3 + \Gamma_{12})(\gamma_4 + \Gamma_{12})])\}$$

$$+ (\alpha_1 \leftrightarrow \alpha_2, \beta_1 \leftrightarrow \beta_2, \Delta_1 \leftrightarrow \Delta_2, \delta_1 \leftrightarrow \delta_2, \gamma_3 \leftrightarrow \gamma_4),$$ \hspace{1cm} (A3.2)

$$\phi_2 = (\alpha_1 \beta_1 \{[\delta_1^2 + (\gamma_3 + \Gamma_{12})]^2[\delta_1^2 (\gamma_4 + \Gamma_{12}) + \delta_1 (\Gamma_{12} + \Gamma_{12})]$$

$$+ \alpha_2^2 [\delta_1 (\gamma_3 + \Gamma_{12}) - \delta_2 (\gamma_4 + \Gamma_{12})])\}$$

$$+ (\alpha_1 \leftrightarrow \alpha_2, \beta_1 \leftrightarrow \beta_2, \delta_1 \leftrightarrow \delta_2, \Delta_1 \leftrightarrow \Delta_2, \gamma_3 \leftrightarrow \gamma_4),$$ \hspace{1cm} (A3.3)

$$C = \{[\delta_1^2 - \Delta_1]([\gamma_3 + \Gamma_{12}] \delta_1 + (\gamma_4 + \Gamma_{12} \delta_2) - \alpha_1^2 (\gamma_3 + \Gamma_{12}) - \alpha_2^2 (\gamma_4 + \Gamma_{12})$$

$$- (\Gamma_{12} + \Gamma_{21})[(\gamma_3 + \Gamma_{12})(\gamma_4 + \Gamma_{12}) - \delta_1 \delta_2]\}^2 + \{\alpha_1^2 \delta_2 + \alpha_2^2 \delta_1 + (\delta_1 - \Delta_1)$$

$$[(\gamma_3 + \Gamma_{12})(\gamma_4 + \Gamma_{12}) - \delta_1 \delta_2] + (\Gamma_{12} + \Gamma_{21})[(\gamma_3 + \Gamma_{12}) \delta_1 + (\gamma_4 + \Gamma_{12}) \delta_2]\}^2.$$ \hspace{1cm} (A3.4)

Using Eqs. (5.3) and (5.5) we have,

$$\zeta = \left(\frac{\alpha_1 \gamma_3}{\beta_1 \gamma_4} \frac{(\gamma_4 + \Gamma_{12})}{(\gamma_4 + \Gamma_{12})^2 + \delta_1^2} + \frac{\alpha_2 \gamma_3}{\beta_2 \gamma_4} \frac{(\gamma_3 + \Gamma_{12})}{(\gamma_3 + \Gamma_{12})^2 + \delta_2^2}\right)$$

$$- i \left(\frac{\alpha_1 \gamma_3}{\beta_1 \gamma_4} \frac{\delta_1}{(\gamma_4 + \Gamma_{12})^2 + \delta_1^2} + \frac{\alpha_2 \gamma_3}{\beta_2 \gamma_4} \frac{\delta_2}{(\gamma_3 + \Gamma_{12})^2 + \delta_2^2}\right),$$ \hspace{1cm} (A3.5)

$$\text{Re}(\chi_2) = \text{Re}(\rho_{21}^{(0)}) \text{Re}(\zeta) - \text{Im}(\rho_{21}^{(0)}) \text{Im}(\zeta).$$ \hspace{1cm} (A3.6)

At two-photon resonance condition $\delta_1 = \Delta_1$ and for $\delta_1 >> \alpha_1, \alpha_2, \gamma_3, \gamma_4, \Gamma_{12}$ we obtain
\[
\text{Re}(\varphi) = \frac{\alpha_1}{\beta_1} \gamma_{41} \left( \frac{\gamma_4 + \Gamma_{12}}{\delta_1^2} \right) + \frac{\alpha_2}{\beta_2} \gamma_{31} \left( \frac{\gamma_3 + \Gamma_{12}}{\delta_2^2} \right) \rightarrow 0, \quad (A3.7)
\]

\[
\text{Im}(\varphi) = -\frac{\alpha_1}{\beta_1} \gamma_{41} \frac{1}{\delta_1} - \frac{\alpha_2}{\beta_2} \gamma_{31} \frac{1}{\delta_2}. \quad (A3.8)
\]

Thus \( A(\delta_1 = \Delta_1) \approx -\text{Im}(\rho_{21}^{(1)}) \text{Im}(\varphi). \)

We assume \( \Gamma_{34} = \Gamma_{43}, \ \Gamma_{12} = \Gamma_{21} \) and \( \gamma_3 = \gamma_4 = \gamma_d. \) AWI is maximized when \( \Delta_1 = S/2, \) which also implies that \( \Delta_2 = -S/2. \) Under these approximations and at the two-photon resonance condition we can obtain \( \text{Im}(\varphi) \) and \( \text{Im}(\rho_{21}^{(1)}) \) as

\[
\text{Im}(\varphi) = \gamma_d (\alpha_2 \beta_1 - \alpha_1 \beta_2) / S \beta_2, \quad (A3.9)
\]

\[
\text{Im}(\rho_{21}^{(1)}) = \frac{2S^3 (\Gamma_{12} + \Gamma_2) (\alpha_1 \beta_1 - \alpha_2 \beta_2) + 16S \alpha_1 \alpha_2 (\gamma_d + \Gamma_{12}) (\alpha_2 \beta_1 - \alpha_1 \beta_2)}{[S^2 (\Gamma_{12} + \Gamma_2) + 4(\gamma_d + \Gamma_{12} + \Gamma_{34}) (\alpha_1^2 + \alpha_2^2)]^2 + S^2 (\alpha_1^2 - \alpha_2^2)^2}. \quad (A3.10)
\]

These expressions are used in the analysis of Sec. 5.3 to obtain the expression for probe absorption at the two-photon resonance condition (cf. Eq. 5.9).
APPENDIX-4

Probability Amplitudes for Quantum Jump Approach

Probability amplitudes $C_i$ relevant to the analysis of Sec. 5.5 are obtained under the assumptions $\beta_1, \beta_2 \ll \alpha_1, \alpha_2$ and $\gamma_3, \gamma_4 \gg \Gamma_1, \Gamma_2, \beta_1, \beta_2$ so that $\gamma_3, \gamma_4$ represent the fastest time scales in the system. Thus for $t \gg 1/\gamma_3, 1/\gamma_4$ it is possible to eliminate fast oscillating variables $C_{i3}$ and $C_{i4}$ as compared to slow variables $C_{i1}$ and $C_{i2}$. Under this adiabatic elimination, we obtain

$$\dot{C}_{i3}(\tau) = \dot{C}_{i4}(\tau) = 0. \quad (A4.1)$$

$$C_{i3} = i(\beta_2 C_{i1} + \alpha_2 C_{i2})/(\gamma_3 + i\delta_2), \quad (A4.2)$$

$$C_{i4} = i(\beta_1 C_{i1} + \alpha_1 C_{i2})/(\gamma_4 + i\delta_1). \quad (A4.3)$$

Using the above equations in Eq. (5.22) we get the following coupled equations:

$$\dot{C}_{i1}(\tau) = -q_1 C_{i1}(\tau) - q_2 C_{i2}(\tau), \quad (A4.4)$$

$$\dot{C}_{i2}(\tau) = -q_2 C_{i1}(\tau) - q_3 C_{i2}(\tau), \quad (A4.5)$$

$$q_1 = \Gamma_1 + \frac{\beta_1^2 \gamma_4}{\gamma_4^2 + \delta_1^2} + \frac{\beta_2^2 \gamma_3}{\gamma_3^2 + \delta_2^2} - i\left[\frac{\beta_1^2 \delta_3}{\gamma_4^2 + \delta_1^2} + \frac{\beta_2^2 \delta_2}{\gamma_3^2 + \delta_2^2}\right], \quad (A4.6)$$

$$q_2 = \frac{\alpha_1 \beta_1 \gamma_4}{\gamma_4^2 + \delta_1^2} + \frac{\alpha_2 \beta_2 \gamma_3}{\gamma_3^2 + \delta_2^2} - i\left[\frac{\alpha_1 \beta_1 \delta_3}{\gamma_4^2 + \delta_1^2} + \frac{\alpha_2 \beta_2 \delta_2}{\gamma_3^2 + \delta_2^2}\right], \quad (A4.7)$$

$$q_3 = \Gamma_2 + \frac{\alpha_1^2 \gamma_4}{\gamma_4^2 + \delta_1^2} + \frac{\alpha_2^2 \gamma_3}{\gamma_3^2 + \delta_2^2} - i\left[\Delta_1 - \delta_1 + \frac{\alpha_1^2 \delta_1}{\gamma_4^2 + \delta_1^2} + \frac{\alpha_2^2 \delta_2}{\gamma_3^2 + \delta_2^2}\right]. \quad (A4.8)$$

Eq. (A4.2) – (A4.5) can be easily solved under the initial condition $C_i(0) = \delta_i$. 

199
APPENDIX-5

Steady State Populations in Tripod System

Considering all the non-radiative decays to be equal i.e. $\Gamma_{ij} = \Gamma$, we obtain the populations in tripod system as,

$$\rho_{11}^{(0)} = \Gamma \left[ 3\Gamma (\gamma_4 + h_1 + 2h_2 + h_3) + 2(h_1h_3 - h_2^2) + (h_1 - h_2)\gamma_4 \right] / \mathcal{Z},$$  \hspace{1cm} (A5.1)

$$\rho_{22}^{(0)} = \Gamma \left[ 3\Gamma (\gamma_4 + h_1 + 2h_2 + h_3) + 2(h_1h_3 - h_2^2) + (h_1 - h_2)\gamma_4 \right] / \mathcal{Z},$$  \hspace{1cm} (A5.2)

$$\rho_{44}^{(0)} = \Gamma \left[ 3\Gamma (h_1 + 2h_2 + h_3) + 2(h_1h_3 - h_2^2) \right] / \mathcal{Z},$$  \hspace{1cm} (A5.3)

$$\rho_{33}^{(0)} = 1 - \rho_{11}^{(0)} - \rho_{22}^{(0)} - \rho_{44}^{(0)},$$  \hspace{1cm} (A5.4)

where

$$h_1 = \alpha_1^2 \text{Re} \left( \frac{b_1 b_5^* + \alpha_1^2}{\mathcal{Z}_1} \right), \quad h_2 = -\alpha_2^2 \alpha_1^2 \text{Re} \left( \frac{1}{\mathcal{Z}_1} \right), \quad h_3 = \alpha_2^2 \text{Re} \left( \frac{b_4^* b_3^* + \alpha_2^2}{\mathcal{Z}_1} \right),$$  \hspace{1cm} (A5.5)

$$\mathcal{Z} = 3\Gamma^2 (4h_1 + 8h_2 + 4h_3 + 3\gamma_4) + 2\Gamma \gamma_4 (h_1 + h_3 - h_2) + (8\Gamma + \gamma_{43})(h_1h_3 - h_2^2),$$  \hspace{1cm} (A5.6)

$$\mathcal{Z} = b_4^* b_3^* b_5 + \alpha_1^2 b_5^* + \alpha_2^2 b_3,$$  \hspace{1cm} (A5.7)

where the coefficients $b_i, (i=1..6)$ are defined in Eq. (6.2). These population terms are used in the analysis of Sec. 6.2.1.
APPENDIX-6

Steady State Populations in N System

For Model A

\[ \rho_{11}^{(0)} = \Gamma_{21}[(\Gamma_{34} - \Gamma_{3} \Gamma_{4}) + x_3(\Gamma_{34} + \Gamma_{4}) + (\Gamma_{3} x_4 + \Gamma_{4} x_2) + x_3^2 - x_2 x_4]/x_1, \]  
\[ \rho_{22}^{(0)} = [(x_3^2 - x_2 x_4)(\Gamma_{12} + \gamma_4 + \gamma_3) + \Gamma_{12}(\Gamma_{34} - \Gamma_{3} \Gamma_{4}) + x_3(\Gamma_{4} \gamma_{32} + \Gamma_{3} \gamma_{42} + \Gamma_{4} \gamma_{32} + \Gamma_{43} \gamma_{34})] + x_2(\Gamma_{4} (\Gamma_{12} + \gamma_3) + \gamma_{42} \Gamma_{34}) + x_4(\Gamma_{3} (\Gamma_{12} + \gamma_4) + \gamma_{32} \Gamma_{43})]/x_1, \]  
\[ \rho_{33}^{(0)} = 2\Gamma_{21}[x_3^2 - x_2 x_4 + \Gamma_{3} (x_2 + x_3) + \Gamma_{34} (x_3 + x_4)]/x_1, \]  
\[ \rho_{44}^{(0)} = 2\Gamma_{21}[x_3^2 - x_2 x_4 + \Gamma_{3} (x_3 + x_4) + \Gamma_{34} (x_2 + x_3)]/x_1, \]

where

\[ x_1 = (x_3^2 - x_2 x_4)(\Gamma_{12} + 3\Gamma_{21} + \gamma_{32} + \gamma_4) + (\Gamma_{12} + \Gamma_{21})(\Gamma_{34} - \Gamma_{3} \Gamma_{4}) + x_4(\Gamma_{4} (\Gamma_{21} + \gamma_{32}) + \Gamma_{4} (\Gamma_{21} + 2\Gamma_{21} + \gamma_{32}) + \Gamma_{43} (\Gamma_{12} + \gamma_{32}) + \Gamma_{43} (\Gamma_{21} + 2\Gamma_{21} + \gamma_{32}) + \Gamma_{34} (\Gamma_{12} + \Gamma_{21} + \gamma_{32})], \]  
\[ x_2 = -\alpha_2^2 \text{Re}(\frac{a_4 a_{12} + \alpha_1^2}{x_5}), \]  
\[ x_3 = \alpha_2^2 \alpha_2^2 \text{Re}(\frac{1}{x_5}), \]  
\[ x_4 = -\alpha_2^2 \text{Re}(\frac{a_4 a_{12} + \alpha_2^2}{x}). \]

For Model B

\[ \rho_{11}^{(0)} = [\Gamma_3 \Gamma_{21}(\gamma_4 + \gamma_{42} - y_2 - 2y_3 - y_4) + \Gamma_3 (y_2 y_4 - y_3^2) + \Gamma_{21} \Gamma_{43} (\gamma_3 + \gamma_{32}) + y_3 (\Gamma_3 \gamma_{42} + \Gamma_{3} \gamma_{32}) - y_4 (\Gamma_3 \gamma_{41} + \Gamma_{3} \gamma_{31})]/y_1, \]  
\[ \rho_{22}^{(0)} = [\Gamma_3 \Gamma_{12}(\gamma_4 + \gamma_{42} - y_2 - 2y_3 - y_4) + \Gamma_3 (y_2 y_4 - y_3^2) + \Gamma_{12} \Gamma_{43} (\gamma_3 + \gamma_{32}) + y_3 (\Gamma_3 \gamma_{42} + \Gamma_{3} \gamma_{32}) - y_2 (\Gamma_3 \gamma_{41} + \Gamma_{3} \gamma_{31})]/y_1, \]
\[ \rho_{33}^{(0)} = \Gamma_3 \left[ (y_2 y_4 - y_3^2) - \Gamma_{12} (y_3 + y_4) - \Gamma_{21} (y_2 + y_3) \right] / y_1, \]  
(A6.10)

\[ \rho_{44}^{(0)} = \Gamma_3 \left[ (y_2 y_4 - y_3^2) - \Gamma_{12} (y_3 + y_4) - \Gamma_{21} (y_2 + y_3) \right] / y_1, \]  
(A6.11)

where

\[
y_1 = (2 \Gamma_3 + \Gamma_4 + \Gamma_3^2) (y_2 y_4 - y_3^2) + \Gamma_{12} (\gamma_{41} + \gamma_{42} - y_2 - 3 y_3 - 2 y_4) \\
+ \Gamma_{21} (\gamma_{41} + \gamma_{42} - 2 y_2 - 3 y_3 - y_4) + \Gamma_{12} \Gamma_{43} (\gamma_{31} + \gamma_{32} - y_3 - y_4) \\
+ \Gamma_{21} \Gamma_{43} (\gamma_{31} + \gamma_{32} - y_2 - y_3) + (\Gamma_{43} \gamma_{32} + \Gamma_{3} \gamma_{42}) (y_3 - y_2) \\
+ (\Gamma_{3} \gamma_{41} + \Gamma_{43} \gamma_{31}) (y_3 - y_4),
\]
(A6.12)

\[
y_2 = -\alpha_2^2 \text{Re} \left( \frac{a_4 a_{14} + \alpha_3^2}{y_5} \right), \quad y_3 = \alpha_2^2 \alpha_3 \text{Re} \left( \frac{1}{y_5} \right), \quad y_4 = -\alpha_3^2 \text{Re} \left( \frac{a_4 a_4 + \alpha_2^2}{y_5} \right),
\]
(A6.13)

\[
y_5 = a_4 a_{14} + \alpha_2^2 a_4 + \alpha_3^2 a_{14}.
\]
(A6.14)

For Model C

\[
\rho_{11}^{(0)} = [\Gamma_{21} (\Gamma_3 \Gamma_4 - \Gamma_{34} \Gamma_4) + \alpha_1^2 \zeta_2 \Gamma_{21} \Gamma_4 + \alpha_1^2 \alpha_3 \zeta_2 \zeta_3 (\Gamma_{21} + \Gamma_4 - \gamma_{42}) \\
+ \alpha_3^2 \zeta_3 \{ \Gamma_3 (\Gamma_{21} + \Gamma_4 - \gamma_{42}) - \Gamma_{43} (\gamma_{32} + \gamma_{34}) \}] / z_1,
\]
(A6.15)

\[
\rho_{22}^{(0)} = [\Gamma_{12} (\Gamma_3 \Gamma_4 - \Gamma_{34} \Gamma_4) + \alpha_1^2 \zeta_2 \{ \Gamma_4 (\Gamma_{12} + \gamma_{32}) + \Gamma_{43} \gamma_{42} \} \\
+ \alpha_3^2 \zeta_2 \Gamma_3 \Gamma_4 + \alpha_3^2 \alpha_3 \zeta_2 \zeta_3 (\Gamma_{12} + \gamma_{32} + \gamma_{34})] / z_1,
\]
(A6.16)

\[
\rho_{33}^{(0)} = [\alpha_3^2 \zeta_2 \Gamma_{21} \Gamma_4 + \alpha_1^2 \zeta_3 \Gamma_{12} \Gamma_{43} + \alpha_1^2 \alpha_3 \zeta_2 \zeta_3 (\Gamma_{21} + \gamma_{41} + \Gamma_{43})] / z_1,
\]
(A6.17)

\[
\rho_{44}^{(0)} = [\alpha_3^2 \zeta_2 \Gamma_{21} \Gamma_3 + \alpha_1^2 \zeta_3 \Gamma_{12} \Gamma_3 + \alpha_1^2 \alpha_3 \zeta_2 \zeta_3 (\Gamma_{12} + \gamma_{32} + \Gamma_{34})] / z_1,
\]
(A6.18)

\[
\rho_{22}^{(j)} = [z_{5}^{(j)} \Gamma_3 \Gamma_4 - \Gamma_{34} \Gamma_3 \Gamma_4 + \alpha_3^2 \zeta_2 \{ 2 \Gamma_{43} + \gamma_{41} \} - z_{6}^{(j)} \alpha_3^2 \zeta_3 (\Gamma_3 + 2 \alpha_1^2 k_2) + z_{4}^{(j)} \\
\{ \gamma_{32} \Gamma_4 + \gamma_{42} \Gamma_{43} - \Gamma_{12} \Gamma_{43} - \Gamma_{12} \Gamma_4 + \alpha_1^2 \zeta_3 (\gamma_{32} - \gamma_{12} + \Gamma_{43}) \} + (z_{5}^{(j)} + z_{6}^{(j)}) \\
\{ \Gamma_{12} \Gamma_3 + \Gamma_{12} \gamma_{43} - \gamma_{32} \Gamma_{43} - \gamma_{42} \gamma_{31} + \alpha_1^2 \zeta_2 \{ \gamma_{32} - 2 \Gamma_3 + \gamma_{12} \} \}] / z_1,
\]
(A6.19)

\[
\rho_{33}^{(j)} = [z_{5}^{(j)} \Gamma_4 - (z_{6}^{(j)} + z_{6}^{(j)}) \Gamma_{43} \Gamma_{12} + \Gamma_{21}) + z_{5}^{(j)} \alpha_3^2 \zeta_2 (\gamma_{42} - \Gamma_4 + \Gamma_{21}) \\
+ z_{6}^{(j)} (\alpha_1^2 \zeta_2 \gamma_{42} + \alpha_1^2 \zeta_2 \Gamma_{21} - \alpha_2^2 \zeta_3 \Gamma_{43} + 2 \alpha_1^2 k_2 \alpha_3^2 \zeta_3) \\
+ z_{4}^{(j)} \alpha_3^2 \zeta_3 (2 \Gamma_{12} + \Gamma_{21} + \gamma_{41} + \Gamma_{43})] / z_1,
\]
(A6.20)
\[ \rho_{44}^{(j)} = \left\{ z_4^{(j)} \Gamma_{34} - (z_6^{(j)} + z_6^{(j)}) \Gamma_{33} \right\} \left( \Gamma_{12} + \Gamma_{21} \right) + z_4^{(j)} \alpha_5^2 z_5 \left( \gamma_{32} + \Gamma_{34} - \Gamma_{12} \right) - z_6^{(j)} \left\{ \alpha_5^2 z_2 \left( \gamma_{32} + \Gamma_{12} + 2 \Gamma_{21} \right) + \alpha_5^2 z_3 \Gamma_{3} + 2 \alpha_5^2 z_2 \alpha_5^2 z_3 \right\} - z_5^{(j)} \alpha_5^2 z_2 \left( \Gamma_{34} - \Gamma_{12} - 2 \Gamma_{21} \right) / z_1, \]  
\[ \rho_{11}^{(j)} = -\left( \rho_{22}^{(j)} + \rho_{33}^{(j)} + \rho_{44}^{(j)} \right). \]  
\[ \text{(A6.21)} \]

where

\[ z_1 = \left( \Gamma_{12} + \Gamma_{21} \right) \left( \Gamma_3 \Gamma_4 - \Gamma_{34} \Gamma_{43} \right) + \alpha_5^2 z_2 \left[ \Gamma_3 \left( \Gamma_{12} + 2 \Gamma_{21} + \gamma_{32} \right) + \Gamma_{34} \left( \Gamma_{21} + \gamma_{42} \right) \right] + \alpha_5^2 z_2 \left[ \Gamma_3 \left( \Gamma_4 + 2 \Gamma_{12} + \gamma_{42} \right) + \Gamma_{43} \left( \Gamma_{12} - \gamma_{32} - \Gamma_{34} \right) \right] + 2 \alpha_5^2 z_2 \alpha_5^2 z_3 \left( \Gamma_{12} + \Gamma_{21} + \Gamma_3 + \Gamma_4 - \gamma_{31} - \gamma_{42} \right), \]  
\[ \text{(A6.23)} \]

\[ z_2 = \frac{\Gamma_3 + \Gamma_{12}}{(\Gamma_3 + \Gamma_{12})^2 + \Delta_1}, \quad z_3 = \frac{\Gamma_4 + \Gamma_{21}}{(\Gamma_4 + \Gamma_{21})^2 + \Delta_3}, \]  
\[ \text{(A6.24)} \]

\[ z_4^{(j)} = \alpha_2 \alpha_3 \text{Re} \left( \rho_{34}^{(j)} / a_2^* \right), \quad z_5^{(j)} = \alpha_2 \alpha_3 \text{Re} \left( \rho_{21}^{(j)} / a_5 \right), \]  
\[ \text{(A6.25)} \]

\[ z_6^{(j)} = -\alpha_2 \text{Im} \left( \rho_{14}^{(j)} \right), \quad (j = 1, 2, 3). \]  
\[ \text{(A6.26)} \]

The populations in the three model schemes are used in the analysis of Sec. 6.3 and Sec 6.4.
**APPENDIX-7**

Non-Zero Elements of Matrix $M_{pq}^q$ in $N$ system

\[ M_{1,3}^{pq} = M_{3,1}^{pq} = M_{5,7}^{pq} = M_{7,5}^{pq} = M_{9,11}^{pq} = M_{11,9}^{pq} = M_{13,15}^{pq} = M_{15,13}^{pq} = -i\alpha_1, \quad (A7.1) \]
\[ M_{1,9}^{pq} = M_{9,1}^{pq} = M_{2,10}^{pq} = M_{10,2}^{pq} = M_{3,11}^{pq} = M_{11,3}^{pq} = M_{4,12}^{pq} = M_{12,4}^{pq} = i\alpha_1, \quad (A7.2) \]
\[ M_{1,4}^{pq} = M_{4,1}^{pq} = M_{5,8}^{pq} = M_{8,5}^{pq} = M_{9,12}^{pq} = M_{12,9}^{pq} = M_{13,16}^{pq} = M_{16,13}^{pq} = -i\alpha_2, \quad (A7.3) \]
\[ M_{1,13}^{pq} = M_{13,1}^{pq} = M_{2,14}^{pq} = M_{14,2}^{pq} = M_{3,15}^{pq} = M_{15,3}^{pq} = M_{4,16}^{pq} = M_{16,4}^{pq} = i\alpha_2, \quad (A7.4) \]
\[ M_{2,4}^{pq} = M_{4,2}^{pq} = M_{6,8}^{pq} = M_{8,6}^{pq} = M_{10,12}^{pq} = M_{12,10}^{pq} = M_{14,16}^{pq} = M_{16,14}^{pq} = -i\alpha_3, \quad (A7.5) \]
\[ M_{5,13}^{pq} = M_{13,5}^{pq} = M_{6,14}^{pq} = M_{14,6}^{pq} = M_{7,15}^{pq} = M_{15,7}^{pq} = M_{8,16}^{pq} = M_{16,8}^{pq} = i\alpha_3, \quad (A7.6) \]
\[ M_{1,6}^{pq} = 2\Gamma_{21}, \quad M_{1,1}^{pq} = 2\gamma_{31}, \quad M_{1,16}^{pq} = 2\gamma_{41}, \quad (A7.7) \]
\[ M_{6,1}^{pq} = 2\Gamma_{12}, \quad M_{6,11}^{pq} = 2\gamma_{32}, \quad M_{6,16}^{pq} = 2\gamma_{42}, \quad (A7.8) \]
\[ M_{11,16}^{pq} = 2\Gamma_{43}, \quad M_{16,11}^{pq} = 2\Gamma_{34}, \quad M_{k,k}^{pq} = -\phi_k. \quad (A7.9) \]

Here $\phi_k$ ($k=1,2,\ldots,16$) are related to terms $a_k$ defined in Eq. (6.12) as follows:

\[ \phi_k = a_k + a(p, q, s), \quad (k = 1, 6, 11, 16), \quad (A7.10) \]
\[ \phi_2 = a_2 + a(p, q - 1, s + 1), \quad (A7.11) \]
\[ \phi_3 = a_3 + a(p - 1, q, s), \quad (A7.12) \]
\[ \phi_4 = a_4 + a(p, q - 1, s), \quad (A7.13) \]
\[ \phi_5 = a_5 + a(p, q + 1, s - 1), \quad (A7.14) \]
\[ \phi_7 = a_7 + a(p - 1, q + 1, s - 1), \quad (A7.15) \]
\[ \phi_8 = a_8 + a(p, q, s - 1), \quad (A7.16) \]
\[ \varphi_9 = a_9 + a(p + 1, q, s), \quad (A7.17) \]

\[ \varphi_{10} = a_{10} + a(p + 1, q - 1, s + 1), \quad (A7.18) \]

\[ \varphi_{12} = a_{12} + a(p + 1, q - 1, s), \quad (A7.19) \]

\[ \varphi_{13} = a_{13} + a(p, q + 1, s), \quad (A7.20) \]

\[ \varphi_{14} = a_2 + a(p, q, s + 1), \quad (A7.21) \]

\[ \varphi_{15} = a_{15} + a(p - 1, q + 1, s), \quad (A7.22) \]

\[ a(p,q,s) = p^2 \gamma_{c_1} + q^2 \gamma_{c_2} + s^2 \gamma_{c_3} + 2pq \gamma_{c_{12}} + 2ps \gamma_{c_{13}} + 2qs \gamma_{c_{23}}. \quad (A7.23) \]

These elements are used for the analysis of three and 2+1-photon resonances in Sec. 7.3.