Supplementary material.

for

Electrical Anharmonicity in Hydrogen Bonded Systems. Complete interpretation of the IR spectra of $Cl-\vec{H}$ stretching band in the gaseous $(CH_3)_2O$...HCl complex".

by Najeh Rekik, Jamal Suleiman, Paul Blaise, Marek J. Wojcik, Henryk T. Flakus and Takahito Nakajima.

1 Appendix 1: Expression of the Franck-Condon Factors

$\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} = \langle(0)|\left\{A(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\}|(0)\rangle$

$= \langle(0)|\, e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle = e^{\frac{1}{2}|\Phi(t)|^{2}\Phi(t)^{*}(1+\langle n\rangle)}\,\langle(0)|\, e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}}e^{-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle$

$e^{\frac{1}{2}|\Phi(t)|^{2}(1+\langle n\rangle)}\,\langle(0)|\, e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}}e^{-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle =$

$e^{\frac{1}{2}|\Phi(t)|^{2}(1+\langle n\rangle)}\sum\sum\frac{(-1)^{l}\Phi(t)^{k}\Phi(t)^{*l}\,(1+\langle n\rangle)^{\frac{1}{2}k+l}}{k!l!}\,\langle(0)|\,\mathbf{a}^{\dagger k}\mathbf{a}^{l}\,|(0)\rangle$

l and k must be zero, then

$$\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} = e^{\frac{1}{2}|\Phi(t)|^{2}(1+\langle n\rangle)} \tag{1}$$

also,

$$\left\{\Gamma_{0,0}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} = e^{\frac{1}{2}|\Phi(t)|^{2}\langle n\rangle} \tag{2}$$

$\left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} = e^{\frac{1}{2}|\Phi(t)|^{2}\langle n\rangle}\sum\sum\frac{(-1)^{l}\Phi(t)^{k}\Phi(t)^{*l}\,\langle n\rangle^{\frac{1}{2}k+l}}{k!l!}\,\langle(0)|\,a^{\dagger k}a^{l}\,|(1)\rangle$

k must be zero and l may be1 so that:

$$\left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} = (-1)e^{\frac{1}{2}|\Phi(t)|^{2}\langle n\rangle}\Phi(t)^{*}\,\langle n\rangle^{\frac{1}{2}} \tag{3}$$

$\left\{\Gamma_{1,0}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} = e^{\frac{1}{2}|\Phi(t)|^{2}\langle n\rangle}\sum\sum\frac{(-1)^{l}\Phi(t)^{k}\Phi(t)^{*l}\,\langle n\rangle^{\frac{1}{2}k+l}}{k!l!}\,\langle(1)|\,a^{\dagger k}a^{l}\,|(0)\rangle$

l must be zero and k must be 1

$$\left\{\Gamma_{1,0}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} = \Phi(t)\,\langle n\rangle^{\frac{1}{2}}\,e^{\frac{1}{2}|\Phi(t)|^{2}\langle n\rangle} \tag{4}$$

$\left\{\Gamma_{1,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} = e^{\frac{1}{2}|\Phi(t)|^{2}(1+\langle n\rangle)}\sum\sum\frac{(-1)^{l}\Phi(t)^{k}\Phi(t)^{*l}\,(1+\langle n\rangle)^{\frac{1}{2}k+l}}{k!l!}\,\langle(1)|\,a^{\dagger k}a^{l}\,|(0)\rangle$

k must be 1 and l must be 0,

$$\left\{\Gamma_{1,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} = (\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,e^{\frac{1}{2}|\Phi(t)|^{2}(1+\langle n\rangle)} \tag{5}$$

$\left\{\Gamma_{0,1}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} = e^{\frac{1}{2}|\Phi(t)|^{2}(1+\langle n\rangle)}\sum\sum\frac{(-1)^{l}\Phi(t)^{k}\Phi(t)^{*l}\,(1+\langle n\rangle)^{\frac{1}{2}k+l}}{k!l!}\,\langle(0)|\,a^{\dagger k}a^{l}\,|(1)\rangle$

k=0 and l=1

$$\left\{\Gamma_{0,1}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} = e^{\frac{1}{2}|\Phi(t)|^{2}(1+\langle n\rangle)}(-1)\Phi(t)^{*}\,(1+\langle n\rangle)^{\frac{1}{2}} \tag{6}$$

$$\left\{\Gamma_{0,1}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} = e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle} \sum\sum \frac{(-1)^l \Phi(t)^k \Phi(t)^{*l} \langle n\rangle^{\frac{1}{2}k+l}}{k!l!} \langle(0)| a^{\dagger k} a^l |(1)\rangle$$

k=0 and l=1

$$\left\{\Gamma_{0,1}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} = e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle}(-1)\Phi(t)^* \langle n\rangle^{\frac{1}{2}} \tag{7}$$

$$\left\{\Gamma_{1,1}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} = e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle} \sum\sum \frac{(-1)^l \Phi(t)^k \Phi(t)^{*l} \langle n\rangle^{\frac{1}{2}k+l}}{k!l!} \langle(1)| a^{\dagger k} a^l |(1)\rangle$$

k=1 and l=1

$$\left\{\Gamma_{1,1}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} = e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle}(-1)|\Phi(t)|^2 \langle n\rangle \tag{8}$$

$$\left\{\Gamma_{1,1}(\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}})\right\} = e^{\frac{1}{2}|\Phi(t)|^2(1+\langle n\rangle)} \sum\sum \frac{(-1)^l \Phi(t)^k \Phi(t)^{*l} (1+\langle n\rangle)^{\frac{1}{2}k+l}}{k!l!} \langle(1)| a^{\dagger k} a^l |(1)\rangle$$

k=1 and l=1

$$\left\{\Gamma_{1,1}(\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}})\right\} = e^{\frac{1}{2}|\Phi(t)|^2(1+\langle n\rangle)}(-1)|\Phi(t)|^2 (1+\langle n\rangle) \tag{9}$$

$$\left\{\Gamma_{m,n}(\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}})\right\} = \begin{array}{ccc} & |(0)\rangle & |(1)\rangle \\ \langle(0)| & e^{\frac{1}{2}|\Phi(t)|^2(1+\langle n\rangle)} & -e^{\frac{1}{2}|\Phi(t)|^2(1+\langle n\rangle)}(1+\langle n\rangle)^{\frac{1}{2}}\Phi(t)^* \\ \langle(1)| & (1+\langle n\rangle)^{\frac{1}{2}} e^{\frac{1}{2}|\Phi(t)|^2(1+\langle n\rangle)}\Phi(t) & -e^{\frac{1}{2}|\Phi(t)|^2(1+\langle n\rangle)}(1+\langle n\rangle)|\Phi(t)|^2 \end{array} \tag{10}$$

$$\left\{\Gamma_{m,n}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} = \begin{array}{ccc} & |(0)\rangle & |(1)\rangle \\ \langle(0)| & e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle} & -e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle}\langle n\rangle^{\frac{1}{2}}\Phi(t)^* \\ \langle(1)| & \langle n\rangle^{\frac{1}{2}} e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle}\Phi(t) & -e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle}\langle n\rangle|\Phi(t)|^2 \end{array}$$

Then the ACF becomes:

$$G_0(t) = f(t)q^{\circ\circ 2}e^{\frac{1}{2}|\Phi(t)|^2(1+2\langle n\rangle)} \tag{11}$$

$$\begin{aligned} G_1(t) = & -Kf(t)q^{\circ\circ 2}e^{-i\omega^{\circ\circ}t}(1+\langle n\rangle)^{\frac{1}{2}}\langle n\rangle^{\frac{1}{2}}\left\{e^{\frac{1}{2}|\Phi(t)|^2(1+\langle n\rangle)}\right\}\left\{e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle}\Phi(t)^*\right\} \\ & -Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}\langle n\rangle\left\{e^{\frac{1}{2}|\Phi(t)|^2(1+\langle n\rangle)}\right\}\left\{e^{\frac{1}{2}|\Phi(t)|^2\langle n\rangle}\Phi(t)^*\right\} \end{aligned}$$

$$G_1(t) = -Kf(t)q^{\circ\circ 2}\left\{e^{\frac{1}{2}|\Phi(t)|^2(1+2\langle n\rangle)}\right\}\{\Phi(t)^*\}\left[(1+\langle n\rangle)^{\frac{1}{2}}\langle n\rangle^{\frac{1}{2}}e^{-i\omega^{\circ\circ}t} + e^{i\omega^{\circ\circ}t}\langle n\rangle\right] \tag{12}$$

$$\begin{aligned} G_2(t) = & Kf(t)q^{\circ\circ 2}\langle n\rangle^{\frac{1}{2}}\left\{\Gamma_{0,0}(\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}})\right\}\left\{\Gamma_{1,0}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} \\ & +Kf(t)q^{\circ\circ 2}(1+\langle n\rangle)^{\frac{1}{2}}\left\{\Gamma_{1,0}(\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}})\right\}\left\{\Gamma_{0,0}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} \end{aligned}$$

$$G_2(t) = Kf(t)q^{\circ\circ 2}e^{\frac{1}{2}|\Phi(t)|^2(1+2\langle n\rangle)}(1+2\langle n\rangle)\Phi(t) \tag{13}$$

$$G_3(t) = -K^2q^{\circ\circ 2}f(t)e^{\frac{1}{2}|\Phi(t)|^2(1+2\langle n\rangle)}|\Phi(t)|^2\left[e^{i\omega^{\circ\circ}t}\langle n\rangle(1+2\langle n\rangle) + e^{-i\omega^{\circ\circ}t}(1+\langle n\rangle)\left(\langle n\rangle + (1+\langle n\rangle)\langle n\rangle^{\frac{1}{2}}\Phi(t)\right)\right] \tag{14}$$

$$\begin{aligned} G_0(t) &= f(t)q^{\circ\circ 2}e^{\frac{1}{2}|\Phi(t)|^2(1+2\langle n\rangle)} \\ G_1(t) &= -Kf(t)q^{\circ\circ 2}\left\{e^{\frac{1}{2}|\Phi(t)|^2(1+2\langle n\rangle)}\right\}\left[e^{i\omega^{\circ\circ}t}\langle n\rangle + e^{-i\omega^{\circ\circ}t}(1+\langle n\rangle)^{\frac{1}{2}}\langle n\rangle^{\frac{1}{2}}\right]\Phi(t)^* \\ G_2(t) &= Kf(t)q^{\circ\circ 2}e^{\frac{1}{2}|\Phi(t)|^2(1+2\langle n\rangle)}(1+2\langle n\rangle)\Phi(t) \\ G_3(t) &= -K^2q^{\circ\circ 2}f(t)e^{\frac{1}{2}|\Phi(t)|^2(1+2\langle n\rangle)}\left[e^{i\omega^{\circ\circ}t}\langle n\rangle(1+2\langle n\rangle) + e^{-i\omega^{\circ\circ}t}(1+\langle n\rangle)\left(\langle n\rangle + (1+\langle n\rangle)\langle n\rangle^{\frac{1}{2}}\Phi(t)\right)\right]|\Phi(t)|^2 \end{aligned} \tag{15}$$

$$G(t) = G_0(t)\left[\begin{array}{c} 1 + K\left((1+2\langle n\rangle)\Phi(t) - \left[e^{i\omega^{\circ\circ}t}\langle n\rangle + e^{-i\omega^{\circ\circ}t}(1+\langle n\rangle)^{\frac{1}{2}}\langle n\rangle^{\frac{1}{2}}\right]\Phi(t)^*\right) \\ -K^2\left[e^{i\omega^{\circ\circ}t}\langle n\rangle(1+2\langle n\rangle) + e^{-i\omega^{\circ\circ}t}(1+\langle n\rangle)\left(\langle n\rangle + (1+\langle n\rangle)\langle n\rangle^{\frac{1}{2}}\Phi(t)\right)\right]|\Phi(t)|^2 \end{array}\right] \tag{16}$$

Recall that

$$\langle n\rangle = \frac{1}{e^{\frac{\hbar\omega^{\circ\circ}}{k_B T}} - 1} \tag{17}$$

and

$$\Phi(t) = \beta e^{-\frac{\gamma}{2}t}\left(e^{-i\omega^{\circ\circ}t} - 1\right) \tag{18}$$

and

$$\beta = \alpha^{\circ}\frac{2\omega^{\circ\circ 2} + i\gamma\omega^{\circ\circ}}{2\left(\omega^{\circ\circ 2} + \left(\frac{\gamma}{2}\right)^2\right)} \tag{19}$$

$$|\Phi(t)|^2 = \beta e^{-\frac{\gamma}{2}t}\left(e^{-i\omega^{\circ\circ}t} - 1\right)\beta^* e^{-\frac{\gamma}{2}t}\left(e^{i\omega^{\circ\circ}t} - 1\right) = 2\,|\beta|^2\, e^{-\gamma t}\left(1 - \cos(\omega^{\circ\circ}t)\right) \tag{20}$$

2 Appendix 2: Calculation of $G_{11}(t)$ and $G_{12}(t)$

2.1 Calculation of $G_{11}(t)$

$$G_{11}(t) = Kf(t)q^{\circ\circ 2}e^{-i\omega^{\circ\circ}t}tr\left\{e^{-\lambda\mathbf{a}^\dagger\mathbf{a}}e^{\Phi(t)\mathbf{a}^\dagger - \Phi(t)^*\mathbf{a}}\mathbf{a}^\dagger\right\} \tag{21}$$

Theorem:

$$\varepsilon tr\left\{\exp\left\{-\lambda\mathbf{a}^\dagger\mathbf{a}\right\} f\left(\left(\mathbf{a},\mathbf{a}^\dagger\right)\right)\right\} = \langle(0)|\,\langle(0)|\;\; f\left[(1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a} + \langle n\rangle^{\frac{1}{2}}\,\mathbf{b}^\dagger, (1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a}^\dagger + \langle n\rangle^{\frac{1}{2}}\,\mathbf{b}\right]|(0)\rangle\,|(0)\rangle \tag{22}$$

applying the theorem, we get:

$G_{11}(t) =$

$$Kf(t)q^{\circ\circ 2}e^{-i\omega^{\circ\circ}t}\langle(0)|\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}\left((1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a}^\dagger + \langle n\rangle^{\frac{1}{2}}\,\mathbf{b}\right)|(0)\rangle\,|(0)\rangle \tag{23}$$

$G_{11}(t) =$

$$Kf(t)q^{\circ\circ 2}e^{-i\omega^{\circ\circ}t}\langle(0)|\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}\left((1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a}^\dagger + \langle n\rangle^{\frac{1}{2}}\,\mathbf{b}\right)|(0)\rangle\,|(0)\rangle \tag{24}$$

Expression which may be splitted into:

$$G_{11}(t) = G_{111}(t) + G_{112}(t) \tag{25}$$

$$G_{111}(t) = Kf(t)q^{\circ\circ 2}e^{-i\omega^{\circ\circ}t}\,(1+\langle n\rangle)^{\frac{1}{2}}\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle\,\langle(0)|\,e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}\mathbf{a}^\dagger\,|(0)\rangle \tag{26}$$

$$G_{112}(t) = Kf(t)q^{\circ\circ 2}e^{-i\omega^{\circ\circ}t}\,\langle n\rangle^{\frac{1}{2}}\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle\,\langle(0)|\,e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}\mathbf{b}\,|(0)\rangle \tag{27}$$

$G_{112}(t)$ is vanishing, then:

$$G_{11}(t) = G_{111}(t) \tag{28}$$

$$G_{11}(t) = Kf(t)q^{\circ\circ 2}e^{-i\omega^{\circ\circ}t}\,(1+\langle n\rangle)^{\frac{1}{2}}\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle\,\langle(0)|\,e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}\,|(1)\rangle \tag{29}$$

Franck-Condon Factors

since

$$\langle(0)|\, e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle=\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} \tag{30}$$

and

$$\langle(0)|\, e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\,|(1)\rangle=\left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{31}$$

$$G_{11}(t)=Kf(t)q^{\circ\circ 2}e^{-i\omega^{\circ\circ}t}\,(1+\langle n\rangle)^{\frac{1}{2}}\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\}\left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{32}$$

$$G_{12}(t)=Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}\,\langle n\rangle^{\frac{1}{2}}\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\}\left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{33}$$

Then

$$\begin{aligned}G_{1}(t)=&\,Kf(t)q^{\circ\circ 2}e^{-i\omega^{\circ\circ}t}\,(1+\langle n\rangle)^{\frac{1}{2}}\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\}\left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\}\\&+Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}\,\langle n\rangle^{\frac{1}{2}}\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\}\left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\}\end{aligned} \tag{34}$$

2.2 Calculation of $G_{12}(t)$

$$G_{12}(t)=Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}tr\left\{e^{-\lambda\mathbf{a}^{\dagger}\mathbf{a}}e^{\Phi(t)\mathbf{a}^{\dagger}-\Phi(t)^{*}\mathbf{a}}\mathbf{a}\right\} \tag{35}$$

Theorem:

$$\varepsilon tr\left\{\exp\left\{-\lambda\mathbf{a}^{\dagger}\mathbf{a}\right\}f\left(\left(\mathbf{a},\mathbf{a}^{\dagger}\right)\right)\right\}=\langle(0)|\,\langle(0)|\;\;f\left[(1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a}+\langle n\rangle^{\frac{1}{2}}\,\mathbf{b}^{\dagger},(1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a}^{\dagger}+\langle n\rangle^{\frac{1}{2}}\,\mathbf{b}\right]|(0)\rangle\,|(0)\rangle \tag{36}$$

$$\begin{aligned}&G_{12}(t)=Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}\\&\langle(0)|\,\langle(0)|\,e^{\Phi(t)\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\langle n\rangle^{\frac{1}{2}}\mathbf{b}\right)-\Phi(t)^{*}\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}+\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}\right)}\left((1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a}+\langle n\rangle^{\frac{1}{2}}\,\mathbf{b}^{\dagger}\right)|(0)\rangle\,|(0)\rangle\end{aligned} \tag{37}$$

$$\begin{aligned}&G_{12}(t)=Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}\\&\langle(0)|\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\left((1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a}+\langle n\rangle^{\frac{1}{2}}\,\mathbf{b}^{\dagger}\right)|(0)\rangle\,|(0)\rangle\end{aligned} \tag{38}$$

$$G_{12}(t)=Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}$$

$$\langle(0)|\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\,(1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a}\,|(0)\rangle\,|(0)\rangle \tag{39}$$

$$+\,\langle(0)|\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\,\langle n\rangle^{\frac{1}{2}}\,\mathbf{b}^{\dagger}\,|(0)\rangle\,|(0)\rangle \tag{40}$$

$$G_{12}(t)=Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}\,\langle n\rangle^{\frac{1}{2}}\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle\,\langle(0)|\,e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\mathbf{b}^{\dagger}\,|(0)\rangle \tag{41}$$

$$G_{12}(t)=Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}\,\langle n\rangle^{\frac{1}{2}}\,\langle(0)|\,e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle\,\langle(0)|\,e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\,|(1)\rangle \tag{42}$$

Now let us calculate $G_{12}(t)$ in terms of Franck-Condon Factors. Since

$$\langle(0)|\, e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\,|(0)\rangle=\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} \tag{43}$$

and

$$\langle(0)|\, e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\,|(1)\rangle=\left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{44}$$

we get

$$G_{12}(t)=Kf(t)q^{\circ\circ 2}e^{i\omega^{\circ\circ}t}\,\langle n\rangle^{\frac{1}{2}}\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\}\left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{45}$$

3 Appendix 3: Calculation of $G_{21}(t)$ and $G_{22}(t)$

3.1 Calculation of $G_{21}(t)$

$$G_{21}(t) = Kf(t)q^{\circ\circ 2} tr\left\{e^{-\lambda \mathbf{a}^\dagger \mathbf{a}}\left(\mathbf{a}^\dagger\right)e^{\Phi(t)\mathbf{a}^\dagger - \Phi(t)^*\mathbf{a}}\right\} \tag{46}$$

Theorem:

$$\varepsilon tr\left\{\exp\left\{-\lambda \mathbf{a}^\dagger \mathbf{a}\right\} f\left(\left(\mathbf{a}, \mathbf{a}^\dagger\right)\right)\right\} = \langle(0)|\langle(0)|\ f\left[(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} + \langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger, (1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \langle n\rangle^{\frac{1}{2}}\mathbf{b}\right]|(0)\rangle|(0)\rangle \tag{47}$$

$$G_{21}(t) = Kf(t)q^{\circ\circ 2}$$

$$\langle(0)|\langle(0)|\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \langle n\rangle^{\frac{1}{2}}\mathbf{b}\right)e^{\Phi(t)\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}|(0)\rangle|(0)\rangle \tag{48}$$

$$G_{21}(t) = Kf(t)q^{\circ\circ 2}\langle(0)|\langle(0)|(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger e^{\Phi(t)\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}|(0)\rangle|(0)\rangle \tag{49}$$

$$+\langle(0)|\langle(0)|\langle n\rangle^{\frac{1}{2}}\mathbf{b}e^{\Phi(t)\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}|(0)\rangle|(0)\rangle \tag{50}$$

$$G_{21}(t) = Kf(t)q^{\circ\circ 2}\langle n\rangle^{\frac{1}{2}}\langle(0)|e^{\Phi(t)\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}|(0)\rangle\langle(0)|\mathbf{b}e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}|(0)\rangle \tag{51}$$

$$G_{21}(t) = Kf(t)q^{\circ\circ 2}\langle n\rangle^{\frac{1}{2}}\langle(0)|e^{\Phi(t)\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}|(0)\rangle\langle(1)|e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}|(0)\rangle \tag{52}$$

since

$$\langle(0)|e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}|(0)\rangle = \left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} \tag{53}$$

and

$$\langle(0)|e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}|(1)\rangle = \left\{\Gamma_{1,0}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{54}$$

$$G_{21}(t) = Kf(t)q^{\circ\circ 2}\langle n\rangle^{\frac{1}{2}}\left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\}\left\{\Gamma_{1,0}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{55}$$

3.2 Calculation of $G_{22}(t)$

$$G_{22}(t) = Kf(t)q^{\circ\circ 2} tr\left\{e^{-\lambda \mathbf{a}^\dagger \mathbf{a}}\left(\mathbf{a}\right)e^{\Phi(t)\mathbf{a}^\dagger - \Phi(t)^*\mathbf{a}}\right\} \tag{56}$$

Theorem:

$$\varepsilon tr\left\{\exp\left\{-\lambda \mathbf{a}^\dagger \mathbf{a}\right\} f\left(\left(\mathbf{a}, \mathbf{a}^\dagger\right)\right)\right\} = \langle(0)|\langle(0)|\ f\left[(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} + \langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger, (1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \langle n\rangle^{\frac{1}{2}}\mathbf{b}\right]|(0)\rangle|(0)\rangle \tag{57}$$

$$G_{21}(t) = Kf(t)q^{\circ\circ 2}$$

$$\langle(0)|\langle(0)|\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} + \langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger\right)e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}|(0)\rangle|(0)\rangle \tag{58}$$

$$G_{21}(t) =$$

$$Kf(t)q^{\circ\circ 2}(1+\langle n\rangle)^{\frac{1}{2}}\langle(0)|\langle(0)|\mathbf{a}e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger}|(0)\rangle|(0)\rangle \tag{59}$$

$$+Kf(t)q^{\circ\circ 2}\langle n\rangle^{\frac{1}{2}}\langle(0)|\langle(0)|\mathbf{b}^{\dagger}e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}|(0)\rangle|(0)\rangle \quad (60)$$

$$G_{21}(t)=Kf(t)q^{\circ\circ 2}(1+\langle n\rangle)^{\frac{1}{2}}\langle(0)|\langle(0)|\mathbf{a}e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}|(0)\rangle|(0)\rangle \quad (61)$$

$$G_{21}(t)=Kf(t)q^{\circ\circ 2}(1+\langle n\rangle)^{\frac{1}{2}}\langle(1)|e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}|(0)\rangle\langle(0)|e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}|(0)\rangle \quad (62)$$

since

$$\langle(1)|e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}|(0)\rangle=\left\{\Gamma_{1,0}(\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}})\right\} \quad (63)$$

and

$$\langle(0)|e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}|(0)\rangle=\left\{\Gamma_{0,0}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} \quad (64)$$

$$G_{21}(t)=Kf(t)q^{\circ\circ 2}(1+\langle n\rangle)^{\frac{1}{2}}\left\{\Gamma_{1,0}(\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}})\right\}\left\{\Gamma_{0,0}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} \quad (65)$$

3.3 Calculation of $G_3(t)$

3.3.1 Calculation of $G_{31}(t)$

$$\begin{aligned}G_{31}(t) &= K^2q^{\circ\circ 2}f(t)e^{-i\omega^{\circ\circ}t}\langle(0)|\langle(0)| \\ &\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\langle n\rangle^{\frac{1}{2}}\mathbf{b}\right)e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}} \\ &\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\langle n\rangle^{\frac{1}{2}}\mathbf{b}\right) \quad (66)\end{aligned}$$

$$\begin{aligned}&G_{31}(t)= \\ &K^2q^{\circ\circ 2}f(t)e^{-i\omega^{\circ\circ}t}\langle(0)|\langle(0)| \\ &\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}\right)e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}\right)|(0)\rangle|(0)\rangle \\ &+K^2q^{\circ\circ 2}f(t)e^{-i\omega^{\circ\circ}t}\langle(0)|\langle(0)| \\ &\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}\right)e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\left(\langle n\rangle^{\frac{1}{2}}\mathbf{b}\right)|(0)\rangle|(0)\rangle \\ &+K^2q^{\circ\circ 2}f(t)e^{-i\omega^{\circ\circ}t}\langle(0)|\langle(0)| \\ &\left(\langle n\rangle^{\frac{1}{2}}\mathbf{b}\right)e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\left((1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}\right)|(0)\rangle|(0)\rangle \\ &+K^2q^{\circ\circ 2}f(t)e^{-i\omega^{\circ\circ}t}\langle(0)|\langle(0)| \\ &\left(\langle n\rangle^{\frac{1}{2}}\mathbf{b}\right)e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}\left(\langle n\rangle^{\frac{1}{2}}\mathbf{b}\right)|(0)\rangle|(0)\rangle\end{aligned}$$

Simplifying

$$\begin{aligned}&G_{31}(t)= \\ &K^2q^{\circ\circ 2}f(t)e^{-i\omega^{\circ\circ}t}(1+\langle n\rangle)^{\frac{1}{2}}\langle n\rangle^{\frac{1}{2}}\langle(0)|e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}\mathbf{a}^{\dagger}|(0)\rangle\langle(0)|\ \mathbf{b}e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}|(0)\rangle\end{aligned}$$

$$G_{31}(t)=K^2q^{\circ\circ 2}f(t)e^{-i\omega^{\circ\circ}t}(1+\langle n\rangle)^{\frac{1}{2}}\langle n\rangle^{\frac{1}{2}}\langle(0)|e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}|(1)\rangle\langle(1)|\ e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}|(0)\rangle \quad (67)$$

since

$$\langle(0)|e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^{\dagger}-\Phi(t)^{*}(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}}|(1)\rangle=\left\{\Gamma_{0,1}(\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}})\right\} \quad (68)$$

and

$$\langle(1)|e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b}-\Phi(t)^{*}\langle n\rangle^{\frac{1}{2}}\mathbf{b}^{\dagger}}|(0)\rangle=\left\{\Gamma_{1,0}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} \quad (69)$$

$$G_{31}(t)=K^2q^{\circ\circ 2}f(t)e^{-i\omega^{\circ\circ}t}(1+\langle n\rangle)^{\frac{1}{2}}\langle n\rangle^{\frac{1}{2}}\left\{\Gamma_{0,1}(\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}})\right\}\left\{\Gamma_{1,0}(\Phi(t)\langle n\rangle^{\frac{1}{2}})\right\} \quad (70)$$

3.3.2 Calculation of $\mathbf{G_{32}(t)}$

$$G_{32}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ} t} \langle(0)| \langle(0)| \left((1+\langle n\rangle)^{\frac{1}{2}} \mathbf{a}^\dagger + \langle n\rangle^{\frac{1}{2}} \mathbf{b} \right) e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \left((1+\langle n\rangle)^{\frac{1}{2}} \mathbf{a} + \langle n\rangle^{\frac{1}{2}} \mathbf{b}^\dagger \right) |(0)\rangle |(0)\rangle \tag{71}$$

$$G_{32}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ} t} (1+\langle n\rangle) \langle(0)| \langle(0)| \ \mathbf{a}^\dagger e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} \mathbf{a} |(0)\rangle\, e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} |(0)\rangle$$
$$+K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ} t} (1+\langle n\rangle)^{\frac{1}{2}} \langle n\rangle^{\frac{1}{2}} \langle(0)| \ \mathbf{a}^\dagger e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} |(0)\rangle \langle(0)| e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \mathbf{b}^\dagger |(0)\rangle$$
$$+K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ} t} \langle n\rangle^{\frac{1}{2}} (1+\langle n\rangle)^{\frac{1}{2}} \langle(0)| e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} \mathbf{a} |(0)\rangle \langle(0)| \ \mathbf{b} e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} |(0)\rangle$$
$$+K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ} t} \langle n\rangle \langle(0)| \ e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} |(0)\rangle \langle(0)| \mathbf{b} e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \mathbf{b}^\dagger |(0)\rangle$$
$$G_{32}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ} t} \langle n\rangle \langle(0)| \ e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} |(0)\rangle \langle(0)| \mathbf{b} e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \mathbf{b}^\dagger |(0)\rangle$$
$$G_{32}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ} t} \langle n\rangle \langle(0)| \ e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} |(0)\rangle \langle(1)| e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} |(1)\rangle$$

since

$$\langle(0)| e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} |(0)\rangle = \left\{ \Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,) \right\} \tag{72}$$

and

$$\langle(1)| e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} |(1)\rangle = \left\{ \Gamma_{1,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}}) \right\} \tag{73}$$

$$G_{32}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ} t} \langle n\rangle \left\{ \Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,) \right\} \left\{ \Gamma_{1,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}}) \right\} \tag{74}$$

3.3.3 Calculation of $\mathbf{G_{33}(t)}$

$$G_{33}(t) = K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ} t} \langle(0)| \langle(0)| \left((1+\langle n\rangle)^{\frac{1}{2}} \mathbf{a} + \langle n\rangle^{\frac{1}{2}} \mathbf{b}^\dagger \right) e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \left((1+\langle n\rangle)^{\frac{1}{2}} \mathbf{a}^\dagger + \langle n\rangle^{\frac{1}{2}} \mathbf{b} \right) |(0)\rangle |(0)\rangle \tag{75}$$

$$G_{33}(t) = K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ} t} (1+\langle n\rangle) \langle(0)| \langle(0)| \ (\mathbf{a})\, e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \left(\mathbf{a}^\dagger\right) |(0)\rangle |(0)\rangle \tag{76}$$
$$+K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ} t} (1+\langle n\rangle)^{\frac{1}{2}} \langle n\rangle^{\frac{1}{2}} \langle(0)| \langle(0)| \ (\mathbf{a})\, e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} (\mathbf{b}) |(0)\rangle |(0)\rangle$$
$$+K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ} t} \langle n\rangle^{\frac{1}{2}} (1+\langle n\rangle)^{\frac{1}{2}} \langle(0)| \langle(0)| \ \left(\mathbf{b}^\dagger\right) e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \left(\mathbf{a}^\dagger\right) |(0)\rangle |(0)\rangle$$
$$+K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ} t} \langle n\rangle \langle(0)| \langle(0)| \ \left(\mathbf{b}^\dagger\right) e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} (\mathbf{b}) |(0)\rangle |(0)\rangle$$

$$G_{33}(t) = K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ} t} (1+\langle n\rangle) \langle(0)| \ (\mathbf{a})\, e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} \left(\mathbf{a}^\dagger\right) |(0)\rangle \langle(0)| e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} |(0)\rangle \tag{77}$$

$$G_{33}(t) = K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ} t} (1+\langle n\rangle) \langle(1)| \ e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} \left(\mathbf{a}^\dagger\right) |(1)\rangle \langle(0)| e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} |(1)\rangle \tag{78}$$

since

$$\langle(1)| e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} |(1)\rangle = \left\{ \Gamma_{1,1}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,) \right\} \tag{79}$$

and

$$\langle(0)| e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} |(1)\rangle = \left\{ \Gamma_{1,0}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}}) \right\}$$

$$G_{33}(t) = K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ} t} (1+\langle n\rangle) \left\{ \Gamma_{1,1}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,) \right\} \left\{ \Gamma_{1,0}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}}) \right\}$$

3.3.4 Calculation of $\mathbf{G_{34}(t)}$

$$\begin{aligned} G_{34}(t) &= K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} \langle(0)| \langle(0)| \left((1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a} + \langle n\rangle^{\frac{1}{2}}\,\mathbf{b}^\dagger\right) \\ & e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \\ & \left((1+\langle n\rangle)^{\frac{1}{2}}\,\mathbf{a} + \langle n\rangle^{\frac{1}{2}}\,\mathbf{b}^\dagger\right) |(0)\rangle\, |(0)\rangle \end{aligned} \tag{80}$$

$$G_{34}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} (1+\langle n\rangle) \langle(0)| \langle(0)|\ \mathbf{a} e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \mathbf{a}\, |(0)\rangle\, |(0)\rangle$$
$$+K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} (1+\langle n\rangle)^{\frac{1}{2}} \langle n\rangle^{\frac{1}{2}} \langle(0)| \langle(0)|\ \mathbf{a} e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \mathbf{b}^\dagger\, |(0)\rangle\, |(0)\rangle$$
$$+K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} \langle n\rangle^{\frac{1}{2}} (1+\langle n\rangle)^{\frac{1}{2}} \langle(0)| \langle(0)|\ \mathbf{b}^\dagger e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \mathbf{a}\, |(0)\rangle\, |(0)\rangle$$
$$+K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} \langle n\rangle \langle(0)| \langle(0)|\ \mathbf{b}^\dagger e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger + \Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \mathbf{b}^\dagger\, |(0)\rangle\, |(0)\rangle$$

$$G_{34}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} (1+\langle n\rangle)^{\frac{1}{2}} \langle n\rangle^{\frac{1}{2}} \langle(0)|\ \mathbf{a} e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} |(0)\rangle \langle(0)| e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} \mathbf{b}^\dagger\, |(0)\rangle$$
$$G_{34}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} (1+\langle n\rangle)^{\frac{1}{2}} \langle n\rangle^{\frac{1}{2}} \langle(1)|\ e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} |(0)\rangle \langle(0)| e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} |(1)\rangle$$

since

$$\langle(1)|\, e^{\Phi(t)(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}^\dagger - \Phi(t)^*(1+\langle n\rangle)^{\frac{1}{2}}\mathbf{a}} |(0)\rangle = \left\{\Gamma_{1,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} \tag{81}$$

and

$$\langle(1)|\, e^{+\Phi(t)\langle n\rangle^{\frac{1}{2}}\mathbf{b} - \Phi(t)^*\langle n\rangle^{\frac{1}{2}}\mathbf{b}^\dagger} |(0)\rangle = \left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{82}$$

$$G_{34}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} (1+\langle n\rangle)^{\frac{1}{2}} \langle n\rangle^{\frac{1}{2}} \left\{\Gamma_{1,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} \left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{83}$$

Then we have

$$G_3(t) = G_{31}(t) + G_{32}(t) + G_{33}(t) + G_{34}(t) \tag{84}$$

$$G_{31}(t) = K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ}t} (1+\langle n\rangle)^{\frac{1}{2}} \langle n\rangle^{\frac{1}{2}} \left\{\Gamma_{0,1}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} \left\{\Gamma_{1,0}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{85}$$

$$G_{32}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} \langle n\rangle \left\{\Gamma_{0,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} \left\{\Gamma_{1,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{86}$$

$$G_{33}(t) = K^2 q^{\circ\circ 2} f(t) e^{-i\omega^{\circ\circ}t} (1+\langle n\rangle) \left\{\Gamma_{1,1}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} \left\{\Gamma_{1,0}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{87}$$

$$G_{34}(t) = K^2 q^{\circ\circ 2} f(t) e^{i\omega^{\circ\circ}t} (1+\langle n\rangle)^{\frac{1}{2}} \langle n\rangle^{\frac{1}{2}} \left\{\Gamma_{1,0}(\Phi(t)\,(1+\langle n\rangle)^{\frac{1}{2}}\,)\right\} \left\{\Gamma_{0,1}(\Phi(t)\,\langle n\rangle^{\frac{1}{2}})\right\} \tag{88}$$