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On the characterization of quantum jumps in a coherently-driven Raman system

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Abstract. We examine the characteristics of quantum jumps in a Raman (Λ) system driven by two coherent fields in terms of the joint probability for successive arrivals of photons. In particular, we derive the general expression for $P(1, T_1; 0, T_2; 1, T_3)$, the probability that a photon is counted in the interval $(t, t+T_1)$ and $(t+T_1+T_2, t+T_1+T_2+T_3)$ with no photon in the intermediate interval $(t+T_1, t+T_1+T_2)$, and discuss its behaviour.

1. Introduction

In the recent past a considerable amount of work has been reported on the observation of quantum jumps in a single ion using transitions either in Λ or V configurations of atomic systems [1–4]. The fluctuations in the photon emission in quantum jumps have been theoretically analysed in terms of the intensity–intensity correlations or in terms of the statistics of dark and bright intervals [5–20]. The statistics constitute the calculation of the photon counting distribution $P(n, T)$ which describes the probability of emission of 'n' photons in time interval T. The calculation of $P(n, T)$ requires knowledge of intensity correlations of all orders. However, in order to keep track of the photons emitted in given time intervals the joint probability $P(n_1, T_1; n_2, T_2; n_3, T_3)$, describing the probability of counting n_1 photons in the interval $(t, t+T_1)$, n_2 photons in the interval $(t+T_1; t+T_1+T_2)$ and n_3 photons in the interval $(t+T_1+T_2; t+T_1+T_2+T_3)$, is useful. In particular for the problem of successive arrival of photons, the probability $P(1, T_1; 0, T_2; 1, T_3)$ is of importance.

In the present work we use the joint probability described above to characterize the photon emission process occurring in a three-level Raman (Λ) system driven coherently by an external field satisfying quantum jump conditions. The joint probability refers to the emission of photons corresponding to the excitation of the strong Rayleigh transition.

The paper is organized as follows. In section 2, we discuss the time interval approach for photon statistics. In particular, we derive a general expression for a threefold generating function from which the required joint probability can be obtained. In section 3, we discuss the mathematical formulation describing various intensity correlations pertaining to a three-level Λ system. We then derive the general expression for the joint probability $P(1, T_1; 0, T_2; 1, T_3)$ for this case. An approximate analytical form of this quantity is also derived which allows us to study some broad features. Finally in section 4, we discuss the results based on exact numerical evaluation of the joint probability.

2. Photon statistics: a time interval approach

Statistical properties of optical fields are usually analysed by photoelectron counting techniques. These constitute the most useful and appropriate methods for analysing the properties of light fields and are closest to the experimental detection techniques.

A way of studying the statistical properties of an optical field is by considering the distribution of separation times between the individual photons. A systematic way for studying such time-interval probabilities is through the generating function approach first introduced by Glauber [21]. A review, which includes a formal definition and the uses of such generating functions, is given by Barakat and Blake [22].

Glauber's first-order generating function is given by

$$\begin{aligned} Q(\lambda, T) &= \sum_{n=0}^{\infty}(1-\lambda)^n P(n, T) \\ &= \langle T_{\mathrm{N}} \exp[-\lambda W]\rangle \end{aligned} \tag{1}$$

where $P(n, T)$ is the probability of registering n photocounts in the time interval $[t, t+T]$, and W is related to the intensity $I(t)$ by

$$W = q \int_t^{t+T} I(t')\,\mathrm{d}t' \tag{2}$$

with q the quantum efficiency for detection of photons. The symbol T_{N} denotes the time-ordering, normal ordering operator. It is clear that the quantity of interest $P(n, T)$ is related to the generating function [22–23]

$$P(n, T) = \frac{(-1)^n}{n!}\frac{\partial^n Q(\lambda)}{\partial\lambda^n}\bigg|_{\lambda=1}. \tag{3}$$

Note that the generating function $Q(\lambda)$ involves intensity correlations of all orders, which may be easily seen by expressing $\exp[-\lambda W]$ as a power series in W. The photon distribution $P(n, T)$ in resonance fluorescence from a single atom has been obtained by Mandel [24]. In this case the higher-order intensity correlations can be expressed as products of intensity correlations of second order. This property also holds for a three-level atom interacting with two coherent fields sharing a common level, as in the case of a Λ or V system. In the case of quantum jumps in three-level systems, $P(n, T)$ yields information about statistics of the dark and bright intervals [10, 19].

The higher-order generating functions [23] may be defined by a straightforward generalization of (1). In particular, in connection with quantum jumps, one may be interested in describing the most general situation, namely, the arrival of a photon followed by a gap and subsequently the arrival of another photon. This quantity requires a third-order generating function which is defined as

$$Q(\lambda_1, \lambda_2, \lambda_3) = \sum_{n_1 n_2 n_3=0}^{\infty} \prod_{i=1}^{3}(1-\lambda_i)^{n_i} P(n_1, T_1; n_2, T_2; n_3, T_3) \tag{4}$$

where $P(n_1, T_1; n_2, T_2; n_3, T_3)$ is the joint probability of registering n_i photocounts in the time interval $[t_i, t_i + T_i]$, $(i = 1, 2, 3)$. The quantum optical definition of this generating function is given by

$$Q(\lambda_1, \lambda_2, \lambda_3) = \left\langle T_{\mathrm{N}} \exp\left[-\sum_{i=1}^{3}\lambda_i W_i\right]\right\rangle \tag{5}$$

where W_i is given by equation (2), but defined over the range $[t_i, t_i + T_i]$.

The joint probability $P(n_1, T_1; n_2, T_2; n_3, T_3)$ is obtained from Q by the following formula

$$P(n_1, T_1; n_2, T_2; n_3, T_3) = \left(\frac{(-1)^{n_1+n_2+n_3}}{n_1!n_2!n_3!}\right) \frac{\partial^{n_1+n_2+n_3}}{\partial\lambda_1^{n_1}\partial\lambda_2^{n_2}\partial\lambda_3^{n_3}} Q(\lambda_1, \lambda_2, \lambda_3)|_{\lambda_1=\lambda_2=\lambda_3=1}. \tag{6}$$

Thus the third-order generating function Q is first obtained from equation (5), and subsequently use of equation (6) yields the joint probability distribution. The evaluation of $Q(\lambda_1, \lambda_2, \lambda_3)$ is lengthy and is discussed in the appendix.

It is shown in the appendix that the factorization property allows us to express the result in terms of the normalized intensity correlation of second order only. It is more convenient to introduce the following triple Laplace transform of $Q(\lambda_1, \lambda_2, \lambda_3)$ denoted by $\tilde{Q}(\lambda_1, \lambda_2, \lambda_3)$:

$$\tilde{Q}(\lambda_1, \lambda_2, \lambda_3) = \int_0^\infty \mathrm{d}T_3\, \mathrm{e}^{-z_3 T_3} \int_0^\infty \mathrm{d}T_2\, \mathrm{e}^{-z_2 T_2} \int_0^\infty \mathrm{d}T_1\, Q(\lambda_1, \lambda_2, \lambda_3) \mathrm{e}^{-z_1 T_1}. \tag{7}$$

The expression of $\tilde{Q}(\lambda_1, \lambda_2, \lambda_3)$ is obtained in the appendix.

Next we derive the expression for $P(1, T_1; 0, T_2; 1, T_3)$, which is used to characterize the quantum jump behaviour in atomic systems. This is the joint probability of detecting one photon in the interval $[t_1, t_1 + T_1]$, no photon in the interval $[t_2, t_2 + T_2]$ and one photon in the interval $[t_3, t_3 + T_3]$.

Taking the Laplace transform of this quantity we have

$$\begin{aligned}\tilde{P} &\equiv \int_0^\infty \mathrm{d}T_1 \int_0^\infty \mathrm{d}T_2 \int_0^\infty \mathrm{d}T_3 \exp\left(-\sum_i Z_i T_i\right) P(1, T_1; 0, T_2; 1, T_3) \\ &= \left.\frac{\partial^3 \tilde{Q}}{\partial\lambda_1\partial\lambda_2\partial\lambda_3}\right|_{\lambda_1=\lambda_2=\lambda_3=1}.\end{aligned} \tag{8}$$

Now each term in the expression for $\tilde{Q}$ (equation (A26)) is such so as to render an easy expression for $\tilde{P}$ which can be written as

$$\begin{aligned}\tilde{P} = {}& \frac{(qI_\infty)^2}{(1 + qI_\infty F_1)^2(1 + qI_\infty F_3)^2}\kappa_3 - \frac{(qI_\infty)^3}{(1 + qI_\infty F_1)^2(1 + qI_\infty F_2)(1 + qI_\infty F_3)^2}\kappa_4 \\ & - \frac{(qI_\infty)^3}{(1 + qI_\infty F_2)(1 + qI_\infty F_3)^2} \sum_p \frac{\kappa_p}{[1 + qI_\infty F(z_1 + \nu_p)]^2}\end{aligned} \tag{9}$$

where F_i, χ_i and ν_p are defined in the appendix.

This is as far as we can go formally. In the next section we use the specific form of $f(t)$ for a problem related to quantum jumps in a three-level system.

3. Intensity correlations and evaluation of $P(1, T_1; 0, T_2; 1, T_3)$

We consider a three-level Λ system shown in figure 1. Here we assume that the Stokes transition is very weak and the system is driven by two coherent fields E_1 and E_2 at the respective transition frequencies. The transition between the levels $|2\rangle$ and $|3\rangle$ is also very weak since it is a dipole forbidden transition. In the present model population trapping does not take place since we assume $\nu_1 \neq 0$, $\nu_2 \neq 0$. The Rabi frequencies associated with the fields are $G_1 = -d_{13}E_1/\hbar$, and $G_2 = -d_{12}E_2/\hbar$ respectively. The intensity–intensity correlation function of the field can be expressed in terms of the correlation function of the atomic operators. For instance, the intensity–intensity correlation

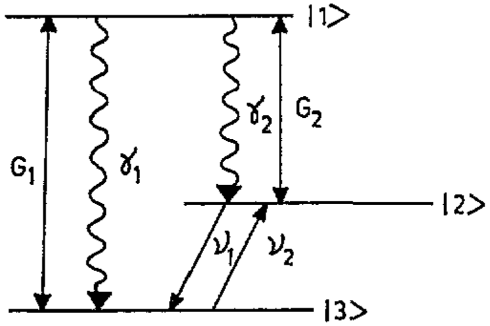

Figure 1. The three-level system in Raman (Λ) configuration.

function for the field emitted on the strong transition $|1\rangle \to |3\rangle$ can be obtained from $\gamma_1^2\langle A_{13}(t)A_{13}(t+\tau)A_{31}(t+\tau)A_{31}(t)\rangle$ which can be denoted by $\gamma_1^2\langle T_N[I_R(t)I_R(t+\tau)]\rangle$. Using the complete set of density matrix equations it has been shown [19] that in the limit of strong driving fields such as $(|G_1|^2+|G_2|^2)^{1/2} \gg \gamma_1, \gamma_2, \nu_1$ and ν_2, the normalized intensity correlation on the strong transition is given by

$$\frac{\langle T_N[I_R(t)I_R(t+\tau)]\rangle}{\langle I_R(t)\rangle\langle I_R(t+\tau)\rangle} = f(\tau) = [1-\exp(-\beta_4\tau)] + \frac{|G_1|^2\beta_4}{G_0^2\beta_5}[\exp(-\beta_4\tau) - \cos(2G_0\tau)\exp(-\beta_2\tau)]. \tag{10}$$

Various decay constants in equation (10) are given by the following expressions:

$$\beta_2 = (\gamma_1+\gamma_2) + \left[\left\{\frac{\gamma_1|G_2|^2}{2G_0^2} + \frac{\nu_1}{4G_0^4}(2|G_2|^4 + |G_1|^2|G_2|^2)\right\} + (1 \rightleftarrows 2)\right] \tag{11}$$

$$\beta_4 = \left[\left\{\frac{\gamma_1|G_2|^2}{G_0^2} + \frac{\nu_1}{2G_0^4}(|G_2|^4 + 2|G_1|^4)\right\} + (1 \rightleftarrows 2)\right] \tag{12}$$

$$\beta_5 = (\nu_1|G_1|^4 + \nu_2|G_2|^4)/G_0^4. \tag{13}$$

The effective Rabi frequency G_0 is defined as

$$G_0 = (|G_1|^2 + |G_2|^2)^{1/2}. \tag{14}$$

The excited-state population in the steady state is

$$I_R(\infty) = \beta_5/2\beta_4. \tag{15}$$

From the preceding equations it is clear that there exists three time scales β_2^{-1}, β_4^{-1} and G_0^{-1} which determine the behaviour of the intensity correlation. The magnitude of these time scales depends in turn on the relative magnitudes of γ_1, γ_2 and ν_1, etc. Furthermore, we assume that $\gamma_1 \gg \gamma_2, \nu_1, \nu_2$ and $G_1 \gg G_2$. Under such conditions one can approximate the β_i's by

$$\beta_2 \approx \frac{3}{2}\gamma_1 + \gamma_2 + \frac{\nu_1|G_2|^2}{4|G_1|^2} \approx \frac{3}{2}\gamma_1 \tag{16a}$$

$$\beta_4 \approx \frac{\gamma_1|G_2|^2}{|G_1|^2} + \gamma_2 + \nu_1 + \frac{\nu_2}{2} \tag{16b}$$

$$\beta_5 \approx \nu_1 + \nu_2\frac{|G_2|^4}{|G_1|^4} - \frac{2\nu_1|G_2|^2}{|G_1|^2} \approx \nu_1. \tag{16c}$$

3.1. Evaluation of $P(1, T_1; 0, T_2; 1, T_3)$

We cast the above expression (10) for the intensity–intensity correlation in the following form for the ease of calculation:

$$f(t) = 1 + a\mathrm{e}^{-\beta_4 t} - \frac{b}{2}[\mathrm{e}^{-\xi} + \mathrm{e}^{\xi^*}] \tag{17}$$

where

$$b = \frac{|G_1|^2\beta_4}{G_0^2\beta_5} = \frac{|G_1|^2}{2G_0^2 I_\infty} \qquad a = b - 1 \qquad \xi = (\beta_2 + 2\mathrm{i}G_0). \tag{18}$$

Next we take the Laplace transform of $f(t)$ with respect to t and obtain

$$F(z) = \frac{1}{z} + \frac{a}{z+\beta_4} - \frac{b(z+\beta_2)}{(z+\xi)(z+\xi^*)}. \tag{19}$$

We define

$$\frac{1}{1 + qI_\infty F(z)} = \frac{N(z)}{D(z)} \tag{20}$$

where

$$N(z) = z(z+\beta_4)(z+\xi)(z+\xi^*) \tag{21}$$

$$D(z) = z^4 + d_3 z^3 + d_2 z^2 + d_1 z + d_0 \equiv 0 \tag{22}$$

with

$$d_0 = qI_\infty[\beta_2^2 + 4G_0^2]\beta_4 \tag{23}$$

$$d_1 = \beta_4[\beta_2^2 + 4G_0^2] + qI_\infty[(\beta_2^2 + 4G_0^2)b + \beta_2\beta_4(1-a)] \tag{24}$$

$$d_2 = (\beta_2^2 + 4G_0^2) + 2\beta_2\beta_4 + qI_\infty(\beta_2 b - \beta_4 a) \tag{25}$$

$$d_3 = 2\beta_2 + \beta_4. \tag{26}$$

The roots of $D(z) = 0$ are denoted by ω_i ($i = 1, 2, 3$ and 4). For further analysis we might need the approximate values of these roots given by

$$\omega_1 = -2\beta_4 I_\infty(|G_0/G_1|)^2 \qquad \omega_2 = -(q/2)(|G_0/G_1|)^2$$
$$\omega_3 = \omega_4^* = -[\xi - (q/4)(|G_0/G_1|)^2]. \tag{27}$$

We now refer to our expression for $\tilde{P}$ derived in section 2 (see equation (9)). Here we have

$$\{\nu_p\} \equiv \beta_4, \xi, \xi^* \qquad \{a_p\} = \left\{a, -\frac{b}{2}, -\frac{b}{2}\right\}. \tag{28}$$

The explicit expression for $\tilde{P}$ can now be written in the following form:

$$\tilde{P} = (qI_\infty)^2\left\{\frac{\Phi_1(z_1)\Phi_1(z_3)}{z_2} + \frac{a\Phi_2(z_1)\Phi_2(z_3)}{z_2+\beta_4} - \frac{b\Phi_3(z_1)\Phi_3(z_3)}{2(z_2+\xi)} - \frac{b\Phi_4(z_1)\Phi_4(z_3)}{2(z_2+\xi^*)}\right\}$$
$$-(qI_\infty)^3\left\{\Phi_1(z_1)\Psi_1(z_2) + a\Phi_2(z_1)\Psi_2(z_2)\right.$$
$$\left.-\frac{b}{2}\Phi_3(z_1)\Psi_3(z_2) - \frac{b}{2}\Phi_4(z_1)\Psi_4(z_2)\right\}\Phi_1(z_3)$$
$$-a(qI_\infty)^3\left\{\Phi_1(z_1+\beta_4)\Psi_2(z_2) + a\Phi_2(z_1+\beta_4)\Psi_5(z_2)\right.$$

$$-\frac{b}{2}\Phi_3(z_1+\beta_4)\Psi_6(z_2)-\frac{b}{2}\Phi_4(z_1+\beta_4)\Psi_7(z_2)\Big\}\Phi_2(z_3)$$
$$+\frac{b}{2}(qI_\infty)^3\Big\{\Phi_1(z_1+\xi)\Psi_3(z_2)+a\Phi_2(z_1+\xi)\Psi_6(z_2)$$
$$-\frac{b}{2}\Phi_3(z_1+\xi)\Psi_8(z_2)-\frac{b}{2}\Phi_4(z_1+\xi)\Psi_9(z_2)\Big\}\Phi_3(z_3)$$
$$+\frac{b}{2}(qI_\infty)^3\Big\{\Phi_1(z_1+\xi^*)\Psi_4(z_2)+a\Phi_2(z_1+\xi^*)\Psi_7(z_2)$$
$$-\frac{b}{2}\Phi_3(z_1+\xi^*)\Psi_9(z_2)-\frac{b}{2}\Phi_4(z_1+\xi^*)\Psi_{10}(z_2)\Big\}\Phi_4(z_3) \tag{29}$$

in which the quantities Φ_i and Ψ_i are given by

$$\Phi_i(z)=\frac{1}{z(z+x_i)(1+qI_\infty F)^2} \tag{30a}$$

where $x_i = 0$, β_4, ξ, ξ^*, (for i = 1–4 respectively);

$$\Psi_i(z)=\frac{1}{(z+x_i)(z+y_i)(1+qI_\infty F)} \tag{30b}$$

here $x_i = 0$ and $y_i = 0, \beta_4, \xi, \xi^*$ (for i = 1–4 respectively); $x_i = \beta_4$ and $y_i = \beta_4, \xi, \xi^*$, (for i = 5–7 respectively); $x_i = \xi$ and $y_i = \xi, \xi^*$ (for i = 8, 9 respectively); $x_{10} = y_{10} = \xi^*$.

The complete expression for the joint probability $P(1, T_1; 0, T_2; 1, T_3)$ may be obtained from equation (29) by taking the Laplace inverse of the various functions $\phi_i(z)$ and $\psi_i(z)$.

However, for further discussion, we need to consider only an approximate expression for P, where only $\phi_{1,2}(T)$ and $\psi_{1,2,5}(T)$ are involved. Explicit expressions for these functions are as follows:

$$\phi_1(t)=\sum_{j=1}^{4}\frac{[(\omega_j+\beta_4)(\omega_j+\xi)(\omega_j+\xi^*)]^2}{\prod_{j=1}^{4}(\omega_j-\omega_i)^2}$$
$$\times\exp(\omega_j t)\left\{t+2\left(\frac{1}{\omega_j+\beta_4}+\frac{1}{\omega_j+\xi}+\frac{1}{\omega_j+\xi^*}\right)-2\sum_{i\neq j}\frac{1}{\omega_j-\omega_i}\right\} \tag{31a}$$

$$\phi_2(t)=\sum_{j=1}^{4}\frac{\omega_j(\omega_j+\beta_4)(\omega_j+\xi)^2(\omega_j+\xi^*)^2}{\prod_{j=1}^{4}(\omega_j-\omega_i)^2}$$
$$\times\exp(\omega_j t)\left\{t+\left(\frac{1}{\omega_j+\beta_4}+\frac{1}{\omega_j}+\frac{2}{\omega_j+\xi}+\frac{2}{\omega_j+\xi^*}\right)-2\sum_{i\neq j}\frac{1}{\omega_j-\omega_i}\right\} \tag{31b}$$

$$\Psi_1(t)=L^{-1}[\Psi_1(z)]=\frac{1}{qI_\infty}+\sum_{i=1}^{4}\frac{(\omega_i+\beta_4)(\omega_i+\xi)(\omega_i+\xi^*)\eta_i(t)}{\omega_i} \tag{32a}$$

$$\Psi_2(t)=L^{-1}[\Psi_2(z)]=\sum_{i=1}^{4}(\omega_1+\xi)(\omega_i+\xi^*)\eta_i(t) \tag{32b}$$

$$\Psi_5(t)=L^{-1}[\Psi_5(z)]=\sum_{i=1}^{4}\frac{\omega_i(\omega_i+\xi)(\omega_i+\xi^*)\eta_i(t)}{\omega_i+\beta_4}-\frac{\beta_4(\beta_4-\xi)(\beta_4-\xi^*)}{\prod_{j=1}^{4}(\omega_j+\beta_4)}\exp(-\beta_4 t) \tag{32c}$$

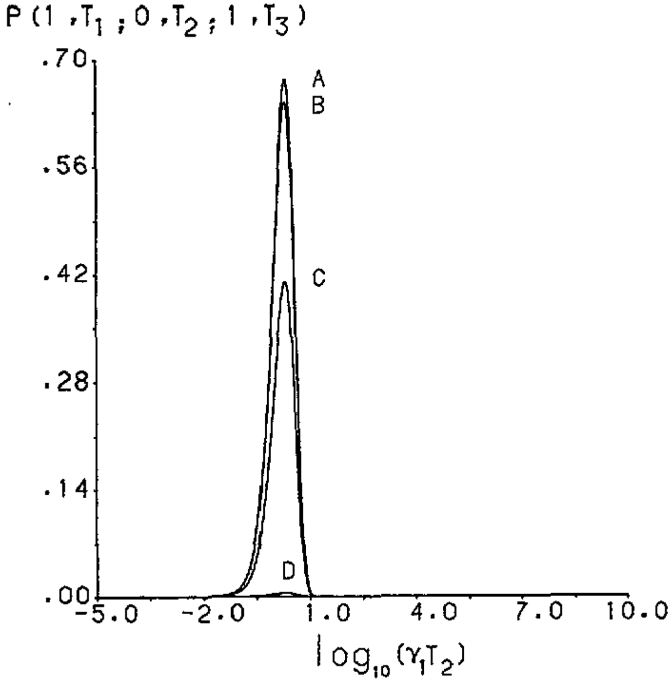

Figure 2. The joint probability $P(1, T_1; 0, T_2; 1, T_3) \times 10^{-1}$ as a function of interval $T_1 = T_3 = T$ (in units of γ^{-1}) with $G_1 = 10$, $G_2 = 0.0$, $\gamma_1 = 1.0$, $\gamma_2 = 10^{-6}\gamma_1$, $\nu_1 = 10^{-6}\gamma_1$, $\nu_2 = 0.0$ and the quantum efficiency parameter $q = 1$. Curves A, B, C, D correspond to the values of $\gamma_1 T_2 = 0.01, 0.1, 1.0, 10.0$, respectively. The oscillatory character of P for very short times is not shown here.

where

$$\eta_i(t) = \frac{e^{i\omega_i t}}{\prod_{i\# j}(\omega_i - \omega_j)} \tag{33}$$

3.2. Approximate expression for $P(1, T_1; 0, T_2; 1, T_3)$

Basically, in quantum jump problems we are interested in slow time scales. On this scale the rapidly oscillating part involving $\exp(-\xi T)$ and $\exp(-\xi^* T)$ are neglected. The expression for $P(1, T_1; 0, T_2; 1, T_3)$ now takes the simplified form

$$\begin{aligned} P(1, T_1; 0, T_2; 1, T_3) &= (qI_\infty)^2\{\Phi_1(T_1)\Phi_1(T_3) + a\Phi_2(T_1)\Phi_2(T_3)e^{-\beta_4 T_2}\} \\ &\quad -(qI_\infty)^3\{\Phi_1(T_1)\Psi_1(T_2) + a\Phi_2(T_1)\Psi_2(T_2)\}\Phi_1(T_3) \\ &\quad -a(qI_\infty)^3\{\Phi_1(T_1)\Psi_2(T_2) + a\Phi_2(T_1)\Psi_5(T_2)\}\Phi_2(T_3)e^{-\beta_4 T_1} \\ &= (qI_\infty)^2[1 - qI_\infty\Psi_1(T_2)]\Phi_1(T_1)\Phi_1(T_3) \\ &\quad +a(qI_\infty)^2\{e^{-\beta_4 T_2} - aqI_\infty e^{-\beta_4 T_1}\Psi_5(T_2)\}\Phi_2(T_1)\Phi_2(T_3) \\ &\quad -a(qI_\infty)^3\{\Phi_2(T_1)\Phi_1(T_3) + \Phi_2(T_3)\Phi_1(T_1)e^{-\beta_4 T_1}\}\Psi_2(T_2). \end{aligned} \tag{34}$$

The approximate roots ω_1 and ω_3 (cf equation (27)) need to be considered now. For the special case when $G_2 = 0$, these roots are

$$\omega_1 \simeq -2\beta_4 I_\infty \qquad \omega_3 \simeq -q/2. \tag{35}$$

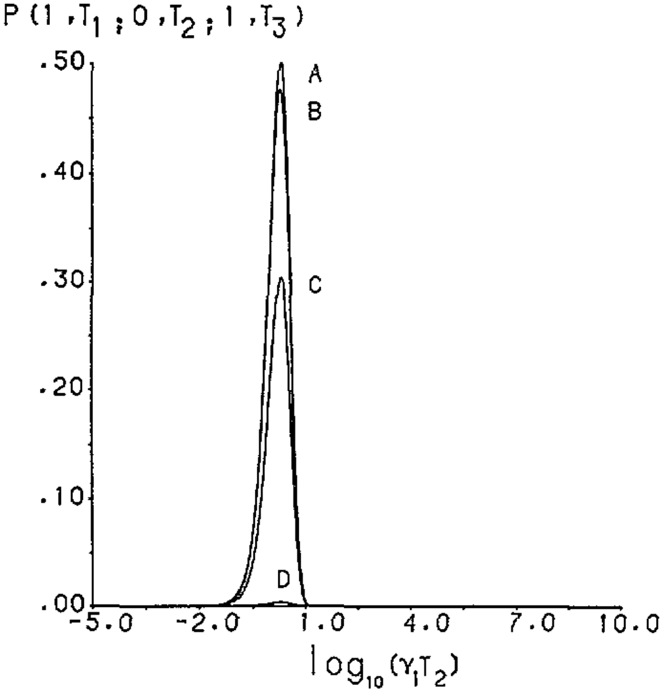

Figure 3. The joint probability $P(1, T_1; 0, T_2; 1, T_3) \times 10^{-2}$ as a function of interval $T_1 = T_3 = T$ (in units of γ^{-1}) with $G_2 = 0.05$. The other parameters are the same as in figure 2.

In this case, we have the following expressions for the Φ's and Ψ's:

$$\phi_1(t) = \frac{4\beta_4}{q^2}(1-2I_\infty)[\beta_4(1-2I_\infty)t - 2]\mathrm{e}^{-2\beta_4 I_\infty t} + \left[t + \frac{8\beta_4}{q^2}(1-2I_\infty)\right]\mathrm{e}^{-qt/2} \qquad (36a)$$

$$\phi_2(t) = \left(-\frac{4\beta_4}{q^2}\right)[\beta_4 I_\infty(1-2I_\infty)t - (1-4I_\infty)]\mathrm{e}^{-2\beta_4 I_\infty t} + \left[t + \frac{4}{q^2}(1-4I_\infty)\beta_4\right]\mathrm{e}^{-qt/2} \qquad (36b)$$

$$\Psi_1(t) = \frac{1}{qI_\infty}[1 - \mathrm{e}^{-2\beta_4 I_\infty t}] + \frac{2}{q}[\mathrm{e}^{-2\beta_4 I_\infty t} - \mathrm{e}^{-qt/2}] \qquad (36c)$$

$$\Psi_2(t) = \frac{2}{q}[\mathrm{e}^{-2\beta_4 I_\infty t} - \mathrm{e}^{-qt/2}] \qquad (36d)$$

$$\Psi_5(t) = \frac{2}{q(1-2I_\infty)}[\mathrm{e}^{-\beta_4 t} - 2I_\infty \mathrm{e}^{-2\beta_4 I_\infty t}] - \frac{2}{q}\mathrm{e}^{-qt/2}. \qquad (36e)$$

Note that as $t \to 0$, $\phi_{1,2}(t)$ and $\Psi_{1,2,5}(t) \to 0$. We next examine the behaviour of $P(1, T_1; 0, T_2; 1, T_3)$ for various situations.

(i) First, we keep T_1 and T_3 fixed and examine P as T_2 is varied. For $T_2 \to 0$, $\Psi_1(T_2)$, $\Psi_2(T_2)$ and $\Psi_5(T_2) \to 0$, and we have

$$P(1, T_1; 0, T_2; 1, T_3) \to (qI_\infty)^2\{\Phi_1(T_1)\Phi_1(T_3) + a\Phi_2(T_1)\Phi_2(T_3)\}. \qquad (37)$$

Hence the initial value of P as a function of T_2 depends on T_1 and T_3 as expected. For small, as well as very large values of T_1 and T_3, P is small. However, P attains a maximum for some intermediate values of T_1 and T_3.

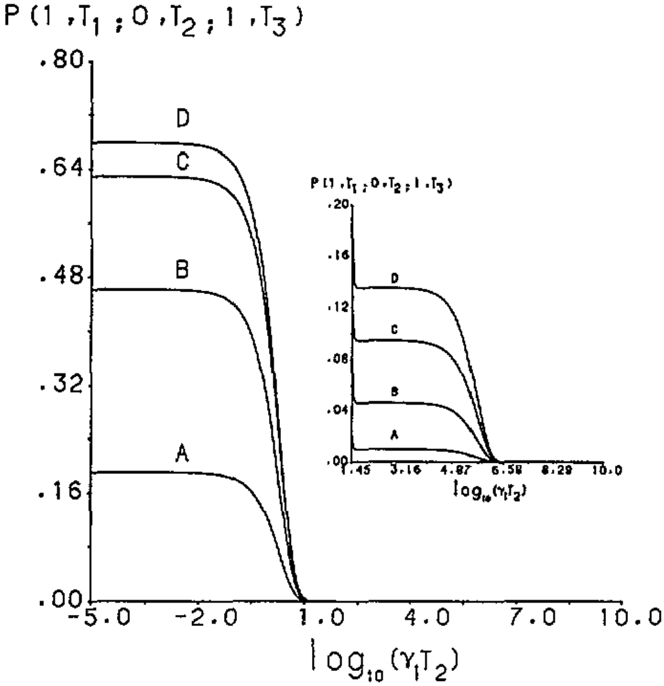

Figure 4. The joint probability $P(1, T_1; 0, T_2; 1, T_3) \times 10^{-1}$ as a function of interval T_2 (in units of γ^{-1}) with $G_1 = 10$, $G_2 = 0.0$, $\gamma_1 = 1.0$, $\gamma_2 = 10^{-6}\gamma_1$, $\nu_1 = 10^{-6}\gamma_1$ and $\nu_2 = 0.0$. Here we have taken $T_1 = T_3 = T$ and the curves A, B, C, D correspond to the values of $\gamma_1 T = 0.5$, 1.0, 1.5, 2.0, respectively. The inset shows $P(1, T_1; 0, T_2; 1, T_3) \times 10^{-6}$ as a function of T_2 from $\log_{10}(\gamma_1 T_2) = 1.45$ onwards.

(ii) For small values of T_2, $e^{-\beta_4 T_2}$ and $e^{-2\beta_4 I_\infty T_2} \cong 1$ and hence,

$$\Psi_{1,2,5}(t) \rightarrow \frac{2}{q}[1 - e^{-qt/2}]. \tag{38}$$

Therefore

$$\begin{aligned} P(1, T_1; 0, T_2; 1, T_3) \cong (qI_\infty)^2\{&[(1-2I_\infty) + 2I_\infty e^{-qT_2/2}]\Phi_1(T_1)\Phi_1(T_3) \\ &+a[1 - 2aI_\infty e^{-\beta_4 T_1}(1 - e^{-qT_2/2})]\Phi_2(T_1)\Phi_2(T_3) \\ &-2aI_\infty[\Phi_2(T_1)\Phi_1(T_3) + \Phi_1(T_1)\Phi_2(T_3)e^{-\beta_4 T_1}](1 - e^{-qT_2/2})\}. \end{aligned} \tag{39}$$

Now as T_2 is increased, P decreases steadily from its initial value (37), the decreased rate being governed by the decay rate $q/2$. Eventually the factor $e^{-qT_2/2}$ tends to zero and P attains a plateau with the value given by

$$\begin{aligned} P_{\text{at plateau}} = (qI_\infty)^2\{&(1-2I_\infty)\Phi_1(T_1)\Phi_1(T_3) + a(1 - 2aI_\infty e^{-\beta_4 T_1})\Phi_2(T_1)\Phi_2(T_3) \\ &-2aI_\infty[\Phi_2(T_1)\Phi_1(T_3) + \Phi_1(T_1)\Phi_2(T_3)e^{-\beta_4 T_1}]\}. \end{aligned} \tag{40}$$

(iii) For larger values of T_2 where $e^{-qT_2/2} \rightarrow 0$ but $e^{-\beta_4 T_2}$ and $e^{-2\beta_4 I_\infty T_2}$ are significant we have

$$\Psi_1(T_2) = \frac{1}{qI_\infty} - \frac{2a}{q}e^{-2\beta_4 I_\infty T_2} \tag{41}$$

where $a = (1 - 2I_\infty)/2I_\infty$

$$\Psi_2(T_2) = \frac{2}{q}e^{-2\beta_4 I_\infty T_2} \tag{42}$$

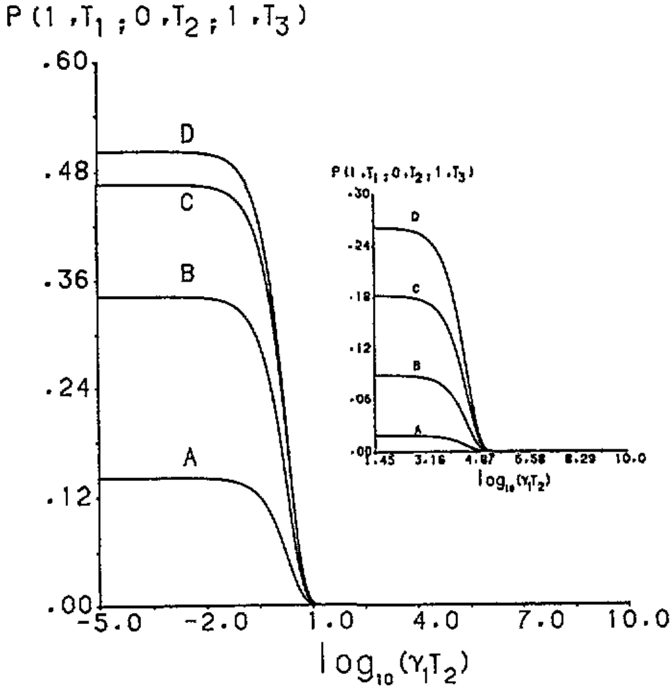

Figure 5. The joint probability $P(1, T_1; 0, T_2; 1, T_3) \times 10^{-2}$ as a function of interval T_2 (in units of γ^{-1}) with $G_2 = 0.05$. The other parameters are the same as in figure 4. The inset shows $P(1, T_1; 0, T_2; 1, T_3) \times 10^{-6}$ as a function of T_2 from $\log_{10}(\gamma_1 T_2) = 1.45$ onwards.

$$\Psi_5(T_2) = \frac{1}{qaI_\infty}[\mathrm{e}^{-\beta_4 T_2} - 2I_\infty \mathrm{e}^{-2\beta_4 I_\infty T_2}]. \tag{43}$$

This leads to

$$\begin{aligned} P(1, T_1; 0, T_2; 1, T_3) = (qI_\infty)^2((1 - 2I_\infty)\mathrm{e}^{-2\beta_4 I_\infty T_2}\Phi_1(T_1)\Phi_1(T_3) \\ +a[\mathrm{e}^{-\beta_4 T_2} - (\mathrm{e}^{-\beta_4 T_2} - 2I_\infty \mathrm{e}^{-2\beta_4 I_\infty T_2})\mathrm{e}^{-\beta_4 T_1}]\Phi_2(T_1)\Phi_2(T_3) \\ -2aI_\infty[\Phi_2(T_1)\Phi_1(T_3) + \Phi_1(T_1)\Phi_2(T_3)\mathrm{e}^{-\beta_4 T_1}]\mathrm{e}^{-2\beta_4 I_\infty T_2}). \end{aligned} \tag{44}$$

It is clear from equation (44) that the probability P eventually decays to zero for very long times, characterized by $2I_\infty \beta_4 T_2 \gg 1$.

Note, however, that in the above analysis, we have set $G_2 = 0$. The formula will still hold if we let

$$I_\infty \to I'_\infty = I_\infty \frac{|G_0|^2}{|G_1|^2} \qquad \text{and} \qquad q \to q' = q\frac{|G_1|^2}{|G_0|^2} \tag{45}$$

in the above approximate expressions. These changes will give the behaviour of P for different values of G_2.

4. Discussion of results

We have evaluated numerically the complete expression for the joint probability $P(1, T_1; 0, T_2; 1, T_3)$ for the parameters of interest. The numerical results are consistent with the qualitative features discussed in section 3 with the help of the approximate analytical expression (34). In figure 2 we show the behaviour of the joint probability with respect to time $T_1 = T_3 = T$ for various values of time T_2 when no field is applied to drive the weak transition $|1\rangle \rightarrow |2\rangle (G_2 = 0)$. As expected on physical grounds, the probability shows a sharp maximum when $\gamma_1 T \simeq 1$. However, as the time interval (T_2) between successive emissions is increased, the height of the maximum decreases. This is because the successive photons arrive within a time interval that is of the order of γ_1^{-1}. Also, as shown in figure 3, when a field is applied to drive the weak transition $|1\rangle \rightarrow |2\rangle$ the heights of the maxima decrease, as compared to the previous case ($G_2 = 0$), due to the increased probability of shelving of the electron in the level $|2\rangle$. Figure 4 displays the behaviour of joint probability P with respect to time T_2 for some fixed values of $T_1 = T_3 = T(\gamma_1 T \geqslant 1)$ with $G_2 = 0$. For small values of T_2, the probability P is nearly constant and thereafter it rapidly decreases. This decrease is essentially governed by the time constant $q/2$. The inset in figure 4, however, shows the existence of a plateau when T_2 approaches the second time scale ($\sim \gamma^{-1}$). During the plateau region the probability that no photon is emitted remains constant due to the shelving effect. Eventually, for very large values of T_2, the quantity P decreases to zero since an interval of that length contains both dark and bright periods of the quantum telegraph. Figure 5 shows the behaviour of the joint probability as a function of time T_2 when a small field is applied to drive the transition $|1\rangle \rightarrow |2\rangle$. The behaviour is similar to that shown in figure 4 except that the values of P are lower because of the enhanced probability for the electron to be shelved in level $|2\rangle$.

Appendix. Evaluation of the third-order generating function $Q(\lambda_1, \lambda_2, \lambda_3)$

In this appendix, we outline the steps leading to the evaluation of $\tilde{Q}(\lambda_1, \lambda_2, \lambda_3)$ as defined in equation (7) which is subsequently used in the main text to evaluate $\tilde{P}$.

We start with the expression

$$\begin{aligned} Q(\lambda_1, \lambda_2, \lambda_3) &= \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \langle T_N(\lambda_1 W_1 + \lambda_2 W_2 + \lambda_3 W_3)^n \rangle \\ &= 1 + \sum_{n=1}^{\infty} \frac{(-1)^n}{n!} \sum_{k=0}^{n} {}^nC_k \lambda_1^k \langle T_N W_1^k (\lambda_2 W_2 + \lambda_3 W_3)^{n-k} \rangle \\ &= 1 + \sum_{n=1}^{\infty} \sum_{k=0}^{n} \sum_{m=0}^{n-k} \frac{(-1)^n}{n!} {}^nC_k \, {}^{n-k}C_m \lambda_1^k \lambda_2^m \lambda_3^{n-k-m} \langle T_N W_1^k W_2^m W_3^{n-k-m} \rangle. \end{aligned} \tag{A1}$$

Typical terms which occur in the above expression are $\langle T_N W_i^k \rangle$, $\langle T_N W_i^k W_j^{n-k} \rangle$ ($i, j = 1$–3, $i < j$) and $\langle T_N W_1^k W_2^m W_3^{n-k-m} \rangle$.

We evaluate the contribution of each of these terms towards $\tilde{Q}$. First consider a typical term

$$\begin{aligned} \langle T_N W_1^n \rangle &= q^n \int_t^{t+T_1} \int_t^{t+T_1} \mathrm{d}t_1 \cdots \mathrm{d}t_n \, \langle T_N I(t_1) I(t_2) \cdots I(t_n) \rangle \\ &= q^n n! \int_0^{T_1} \mathrm{d}t_n \cdots \int_0^{t_2} \mathrm{d}t_1 \, \langle T_N I(t_1) I(t_2) \cdots I(t_n) \rangle \\ &= q^n \langle I \rangle^n \int_0^{T_1} \mathrm{d}t_n \cdots \int_0^{t_k} \mathrm{d}t_1 \prod_{s=2}^{n} f(t_s - t_{s-1}) \end{aligned} \tag{A2}$$

where $f(t)$ is the normalized intensity correlation of second order defined by

$$f(t) = \langle T_N I(t_0) I(t_0 + t) \rangle / [\langle I(t_0) \rangle \langle I(t_0 + t) \rangle]. \tag{A3}$$

In the step (A2) we have used the so-called factorization property valid for a single atom. This property allows us to express higher-order correlations as a product of correlations of second order, e.g.

$$\langle T_{\mathrm{N}} I(t_1) I(t_2)\rangle = \langle I\rangle^2 f(t_2 - t_1). \tag{A4}$$

Therefore, the knowledge of this normalized intensity correlation $f(t)$ allows us to evaluate every other correlation.

We thus have

$$\langle T_{\mathrm{N}} W_1^n\rangle = \langle q I_\infty\rangle^n n! \int_0^{T_1} \mathrm{d}t_n \cdots \int_0^{t_k} \mathrm{d}t_1 \prod_{s=2}^{n} f(t_s - t_{s-1}) \tag{A5}$$

where $\langle I\rangle = I_\infty$.

We define a triple Laplace transform

$$\widetilde{\langle T_{\mathrm{N}} W_1^n\rangle} = \int_0^\infty \mathrm{d}T_3\, \mathrm{e}^{-z_3 T_3} \int_0^\infty \mathrm{d}T_2\, \mathrm{e}^{-z^2 T_2} \int_0^\infty \mathrm{d}T_1 \langle T_{\mathrm{N}} W_1^n\rangle \mathrm{e}^{-z_1 T_1} \tag{A6}$$

and using the convolution theorem we get

$$\widetilde{\langle T_{\mathrm{N}} W_i^n\rangle} = (q I_\infty)^n n! \frac{[F(Z_i)]^{n-1}}{Z_1 Z_2 Z_3} \qquad i = 1\text{–}3 \tag{A7}$$

$$\langle \tilde{1}\rangle = \int_0^\infty \mathrm{d}T_3\, \mathrm{e}^{-z_3 T_3} \int_0^\infty \mathrm{d}T_2\, \mathrm{e}^{-z_2 T_2} \int_0^\infty \mathrm{d}T_1\, \mathrm{e}^{-z_1 T_1} = \frac{1}{Z_1 Z_2 Z_3}. \tag{A8}$$

Here $F(z)$ is the Laplace transform of the normalized intensity correlation function $f(t)$ defined earlier.

The contribution of these terms to $\tilde{Q}$ is

$$A = \frac{1}{Z_1 Z_2 Z_3} - \frac{q I_\infty}{Z_1 Z_2 Z_3} \sum_{i=1}^{3} \frac{\lambda_i}{Z_i (1 + \lambda_i q I_\infty F_i)} \tag{A9}$$

where $F_i = F(Z_i)$.

Next we consider terms of the type

$$\begin{aligned}\langle T_{\mathrm{N}} : W_1^k W_2^{n-k}\rangle &= k!(n-k)! \langle q I_\infty\rangle^n \int_0^{T_2} \mathrm{d}t_n \cdots \int_0^{t_{k+2}} \mathrm{d}t_{k+1} \int_0^{T_1} \mathrm{d}t_k \\ &\cdots \int_0^{t_2} \mathrm{d}t_1 \left(\prod_{s=k+2}^{n} f(t_s - t_{s-1})\right) f(t_{k+1} + T_1 - t_k) \left(\prod_{r=1}^{k} f(t_r - t_{r-1})\right)\end{aligned} \tag{A10}$$

and evaluate the triple Laplace transform

$$\begin{aligned}&\int_0^\infty \mathrm{d}T_3\, \mathrm{e}^{-z_3 T_3} \int_0^\infty \mathrm{d}T_2\, \mathrm{e}^{-z_2 T_2} \int_0^\infty \mathrm{d}T_1\, \mathrm{e}^{-z_1 T_1} \int_0^{T_2} \mathrm{d}t_n \cdots \int_0^{t_{k+2}} \mathrm{d}t_{k+1} \int_0^{T_1} \mathrm{d}t_k \\ &\quad \cdots \int_0^{t_2} \mathrm{d}t_1 \left(\prod_{s=k+2}^{n} f(t_s - t_{s-1})\right) f(t_{k+1} + T_1 - t_k) \left(\prod_{r=1}^{k} f(t_r - t_{r-1})\right) \\ &= \frac{1}{Z_3} \int_0^\infty \mathrm{d}T_2\, \mathrm{e}^{-z_2 T_2} \int_0^\infty \mathrm{d}T_1\, \mathrm{e}^{-z_1 T_1} \int_0^{T_2} \mathrm{d}t \cdots \int_0^{t_{k+2}} \mathrm{d}t_{k+1} \int_0^{T_1} \mathrm{d}t_k \\ &\quad \cdots \int_0^{t_2} \mathrm{d}t_1 \left(\prod_{s=k+2}^{n} f(t_s - t_{s-1})\right) f(t_{k+1} + T_1 - t_k) \left(\prod_{r=2}^{k} f(t_r - t_{r-1})\right) \\ &= \frac{F^{n-k-1}(Z_2)}{Z_2 Z_3} \int_0^\infty \mathrm{d}t_{k+1}\, \mathrm{e}^{-z_2 t_{k+1}} \int_0^\infty \mathrm{d}T_1\, \mathrm{e}^{-z_1 T_1} \int_0^{T_1} \mathrm{d}t_k\, f(t_{k+1} + T_1 - t_k) \\ &\quad \times \int_0^{t_k} \mathrm{d}t_{k-1} \int_0^{t_2} \mathrm{d}t_1 \left(\prod_{r=2}^{k} f(t_r - t_{r-1})\right).\end{aligned} \tag{A11}$$

In order to evaluate this we write

$$\int_0^\infty dT_1\, e^{-z_1T_1}\int_0^{T_1} dt_k\, f(t_{k+1}+T_1-t_k) = \int_0^\infty dt_k \int_{t_k}^\infty dT_1\, e^{-z_1T_1} f(t_{k+1}+T_1-t_k)$$
$$= \int_0^\infty dt_k\, e^{-z_1t_k}\int_0^\infty dT_1\, e^{-z_1T_1} f(t_{k+1}+T_1)$$
$$= \int_0^\infty dT_1\, e^{-z_1T_1} f(t_{k+1}+T_1)\int_0^\infty dt_k\, e^{-z_1t_k}. \tag{A12}$$

From here on, the convolution theorem is applicable and we may write

$$\langle T_N : \widetilde{W_1^k} W_2^{n-k}\rangle = k!(n-k)!(qI_\infty)^n \frac{[F(Z_2)]^{n-k-1}[F(Z_1)]^{k-1}}{Z_1Z_2Z_3}$$
$$\times \int_0^\infty dt_{k+1}\, e^{-z_2t_{k+1}}\int_0^\infty dT_1\, e^{-z_1T_1} f(t_{k+1}+T_1). \tag{A13}$$

Consider now the term

$$\langle T_N : W_2^k W_3^{n-k}\rangle = k!(n-k)!(qI_\infty)^n \int_0^{T_3} dt_n \cdots \int_0^{t_{k+2}} dt_{k+1}\int_0^{T_2} dt_k$$
$$\cdots \int_0^{t_2} dt_1 \left(\prod_{s=k+2}^n f(t_s-t_{s-1})\right) f(t_{k+1}+T_2-t_k)\left(\prod_{r=1}^k f(t_r-t_{r-1})\right). \tag{A14}$$

Finally, we consider

$$\langle T_N : W_1^k W_3^{n-k}\rangle = \int_t^{t+T_1} dt_1 \cdots \int_t^{t+T_1} dt_k \int_{t+T_1+T_2}^{t+T_1+T_2+T_3} dt_{k+1}$$
$$\cdots \int_{t+T_1+T_2}^{t+T_1+T_2+T_3} dt_n \langle T_N I(t_1)\cdots I(t_k) I(t_{k+1})\cdots I(t_n)\rangle. \tag{A15}$$

Using the factorization theorem, we may write this as

$$\langle T_N : W_1^k W_3^{n-k}\rangle = k!(n-k)!(qI_\infty)^n \int_0^{T_3} dt_n \cdots \int_0^{t_{k+2}} dt_{k+1}\int_0^{t_{k+1}} dt_k$$
$$\cdots \int_0^{T_1} dt_1 \left(\prod_{s=k+2}^n f(t_s-t_{s-1})\right) f(t_{k+1}+T_1+T_2-t_k)\left(\prod_{r=1}^k f(t_r-t_{r-1})\right). \tag{A16}$$

If we take the triple Laplace transform and use the convolution theorem as before, we obtain

$$\langle T_N : \widetilde{W_1^k} W_2^{n-k}\rangle = (n-k)!k!(qI_\infty)^n [F(Z_1)]^{k-1}[F(Z_2)]^{n-k-1}\kappa_1(Z_1Z_2Z_3) \tag{A17a}$$
$$\langle T_N : \widetilde{W_2^k} W_3^{n-k}\rangle = (n-k)!k!(qI_\infty)^n [F(Z_2)]^{k-1}[F(Z_3)]^{n-k-1}\kappa_2(Z_1Z_2Z_3) \tag{A17b}$$
$$\langle T_N : \widetilde{W_1^k} W_3^{n-k}\rangle = (n-k)!k!(qI_\infty)^n [F(Z_1)]^{k-1}[F(Z_3)]^{n-k-1}\kappa_3(Z_1Z_2Z_3) \tag{A17c}$$

where

$$\kappa_1(Z_1Z_2Z_3) = \frac{1}{Z_1Z_2Z_3}\int_0^\infty dt_{k+1}\, e^{-z_2t_{k+1}}\int_0^\infty dT_1\, e^{-z_1T_1} f(t_{k+1}+T_1) \tag{A18a}$$
$$\kappa_2(Z_1Z_2Z_3) = \frac{1}{Z_1Z_2Z_3}\int_0^\infty dt_{k+1}\, e^{-z_3t_{k+1}}\int_0^\infty dT_2\, e^{-z_2T_2} f(t_{k+1}+T_2) \tag{A18b}$$
$$\kappa_3(Z_1Z_2Z_3) = \frac{1}{Z_1Z_3}\int_0^\infty dT_2\, e^{-z_2T_2}\int_0^\infty dT_1\, e^{-z_1T_1}\int_0^\infty dt_{k+1}\, e^{-z_3t_{k+1}} f(t_{k+1}+T_1+T_2). \tag{A18c}$$

On using the standard form of $f(t)$ given by

$$f(t) = 1 + \sum_p a_p \mathrm{e}^{-\nu_p t} \tag{A19}$$

where ν_p are complex quantities, we arrive at

$$\kappa_1 = \frac{1}{Z_1^2 Z_2^2 Z_3} + \sum_p \frac{a_p}{Z_1(Z_1+\nu_p)Z_2(Z_2+\nu_p)Z_3} \tag{A20a}$$

$$\kappa_2 = \frac{1}{Z_1 Z_2^2 Z_3^2} + \sum_p \frac{a_p}{Z_1(Z_2+\nu_p)Z_2(Z_3+\nu_p)Z_3} \tag{A20b}$$

$$\kappa_3 = \frac{1}{Z_1^2 Z_2 Z_3^2} + \sum_p \frac{a_p}{Z_1(Z_1+\nu_p)Z_2(Z_3+\nu_p)Z_3}. \tag{A20c}$$

Contribution from these terms to $\tilde{Q}(\lambda_1, \lambda_2, \lambda_3)$ is

$$\frac{\lambda_1\lambda_2(qI_\infty)^2\kappa_1(Z_1, Z_2, Z_3)}{(1+qI_\infty\lambda_1F_1)(1+qI_\infty\lambda_2F_2)} + \frac{\lambda_2\lambda_3(qI_\infty)^2\kappa_2(Z_1, Z_2, Z_3)}{(1+qI_\infty\lambda_2F_2)(1+qI_\infty\lambda_3F_3)} + \frac{\lambda_3\lambda_1(qI_\infty)^2\kappa_3(Z_1, Z_2, Z_3)}{(1+qI_\infty\lambda_1F_1)(1+qI_\infty\lambda_3F_3)}. \tag{A21}$$

Finally, we consider the following term in equation (7), i.e. $\langle T_\mathrm{N} : W_1^k W_2^m W_3^{n-k-m}\rangle$. The procedure for reduction of this term is as before. We take the triple Laplace transform. The time ordering and factorization property yields

$$\begin{aligned}\langle T_\mathrm{N} : W_1^k W_2^m W_3^{n-k-m}\rangle &= k!m!(n-k-m)!(qI_\infty)^n \\ &\times \int_0^{T_3} \mathrm{d}t_n \cdots \int_0^{t_{k+m+2}} \mathrm{d}t_{k+m+1} \int_0^{T_2} \mathrm{d}t_{k+m+1} \cdots \int_0^{t_{k+2}} \mathrm{d}t_{k+1} \int_0^{T_1} \mathrm{d}t_k \\ &\cdots \int_0^{t_2} \mathrm{d}t_1 \left(\prod_{h=k+m+2}^{n} f(t_h - t_{h-1}) \right) f(t_{k+m+1} + T_1 + T_2 - t_{k+m}) \\ &\times \left(\prod_{i=k+2}^{k+m} f(t_i - t_{i-1}) \right) f(t_{k+1} + T_1 + T_2 - t_k) \left(\prod_{j=1}^{k} f(t_j - t_{j-1}) \right)\end{aligned} \tag{A22}$$

$$\begin{aligned}\langle T_\mathrm{N} W_1^k \widetilde{W_2^m} W_3^{n-k-m}\rangle &= \frac{(qI_\infty)^n}{Z_3}[F(Z_3)]^{n-k-m-1}[F(Z_2)]^{m-1}\left(\frac{n!}{k!m!(n-k-m)!}\right) \\ &\times \int_0^\infty \mathrm{d}t_{k+m+1}\, \mathrm{e}^{-z_3 t_{k+m+1}} \int_0^\infty \mathrm{d}T_2\, \mathrm{e}^{-z_2 T_2} \int_0^\infty \mathrm{d}t_{k+1}\, \mathrm{e}^{-z_2 t_{k+1}} \\ &\times \int_0^\infty \mathrm{d}T_1\, \mathrm{e}^{-z_1 T_1} \int_0^\infty \mathrm{d}t_k \cdots \int_0^\infty \mathrm{d}t_1 \exp\left(-\sum_{i=1}^{k} Z_i t_i\right) \\ &\times \left(\prod_{i=1}^{k} f(t_i)\right) f\left(t_{k+m+1} + T_1 + T_2 + \sum_{i=1}^{k} t_i\right) f(t_{k+1} + T_1).\end{aligned} \tag{A23}$$

After simplification the contribution to $\tilde{Q}$ from this term becomes

$$\begin{aligned}&-\frac{\lambda_1\lambda_2\lambda_3(qI_\infty)^3\kappa_4}{(1+qI_\infty\lambda_1F_1)(1+qI_\infty\lambda_2F_2)(1+qI_\infty\lambda_3F_3)} - \frac{\lambda_1\lambda_2\lambda_3(qI_\infty)^3}{(1+qI_\infty\lambda_2F_2)(1+qI_\infty\lambda_3F_3)} \\ &\quad\times\left\{\sum_p \frac{\kappa_p}{[1+qI_\infty\lambda_1F(Z_1+\nu_p)]}\right\}\end{aligned} \tag{A24}$$

where

$$\kappa_4 = \frac{1}{(Z_1Z_2Z_3)^2} + \sum_p \frac{a_p}{Z_1(Z_1+\nu_p)Z_2(Z_2+\nu_p)Z_3^2} \tag{A25a}$$

$$\kappa_p = \frac{a_p}{(Z_1+\nu_p)(Z_2+\nu_p)Z_3(Z_3+\nu_p)}\left\{\frac{1}{(Z_1+\nu_p)Z_2} + \sum_r \frac{a_p}{(Z_1+\nu_p+\nu_r)(Z_2+\nu_r)}\right\}. \tag{A25b}$$

Finally, adding the contributions term by term we obtain

$$\begin{aligned}\tilde{Q}(\lambda_1,\lambda_2,\lambda_3) = {}& \frac{1}{Z_1Z_2Z_3} - \frac{qI_\infty}{Z_1Z_2Z_3}\sum_{i=1}^{3}\frac{\lambda_i}{Z_i(1+\lambda_i qI_\infty F_i)} \\ & + \frac{\lambda_1\lambda_2(qI_\infty)^2\kappa_1(Z_1,Z_2,Z_3)}{(1+qI_\infty\lambda_1F_1)(1+qI_\infty\lambda_2F_2)} + \frac{\lambda_2\lambda_3(qI_\infty)^2\kappa_2(Z_1,Z_2,Z_3)}{(1+qI_\infty\lambda_2F_2)(1+qI_\infty\lambda_3F_3)} \\ & + \frac{\lambda_3\lambda_1(qI_\infty)^2\kappa_3(Z_1,Z_2,Z_3)}{(1+qI_\infty\lambda_1F_1)(1+qI_\infty\lambda_3F_3)} \\ & - \frac{\lambda_1\lambda_2\lambda_3(qI_\infty)^3\kappa_4}{(1+qI_\infty\lambda_1F_1)(1+qI_\infty\lambda_2F_2)(1+qI_\infty\lambda_3F_3)} \\ & - \frac{\lambda_1\lambda_2\lambda_3(qI_\infty)^3}{(1+qI_\infty\lambda_2F_2)(1+qI_\infty\lambda_3F_3)}\left\{\sum_p \frac{\kappa_p}{[1+qI_\infty\lambda_1F(Z_1+\nu_p)]}\right\}.\end{aligned} \tag{A26}$$

References

[1] Nagourney W, Sandberg J and Dehmelt H 1986 *Phys. Rev. Lett.* **56** 2797
[2] Bergquist J C, Hulet R G, Itano W M and Wineland D J 1986 *Phys. Rev. Lett.* **57** 1699
[3] Sauter Th, Neuhauser W, Blatt R and Toschek P E 1986 *Phys. Rev. Lett.* **17** 1696
[4] Sauter Th, Blatt R, Neuhauser W and Toschek P E 1986 *Opt. Commun.* **60** 287
[5] Cook R J and Kimble H J 1985 *Phys. Rev. Lett.* **54** 1023
[6] Javanainen J 1986 *Phys. Rev.* A **33** 2121
[7] Schenzle A, Devoe R G and Brewer R G 1986 *Phys. Rev.* A **33** 2127
[8] Dalibard J and Cohen-Tannoudji C 1986 *Europhys. Lett.* **1** 441
[9] Kimble H J, Cook R J and Wells A L 1986 *Phys. Rev.* A **34** 3190
[10] Schenzle A and Brewer R J 1986 *Phys. Rev.* A **34** 3127
[11] Pegg D T, Loudon R and Knight P L 1986 *Phys. Rev.* A **33** 4085
[12] Erber T and Putterman S 1985 *Nature* **318** 41
[13] Knight P L, Loudon R and Pegg D T 1986 *Nature* **323** 608
[14] Zoller P, Marte M and Walls D F 1987 *Phys. Rev.* A **35** 198
[15] Kim M S, Knight P L and Wodkiewicz K 1987 *Opt. Commun.* **82** 385
[16] Kim M S and Knight P L 1987 *Phys. Rev.* A **36** 5265
[17] Nienhius G 1987 *Phys. Rev.* A **35** 4639
[18] Porrati M and Putterman S P 1987 *Phys. Rev.* A **36** 929; 1989 *Phys. Rev.* A **39** 3010
[19] Agarwal G S, Lawande S V and D'Sousza R 1988 *Phys. Rev.* A **37** 444
[20] Erber T, Hammerling P, Hockney G, Porrati M and Putterman S 1989 *Ann. Phys.* **190** 254
[21] Glauber R J 1965 *Quantum Optics and Electronics* ed C Dewitt, A Blandin and C Cohen-Tannoudji (New York: Gordon and Breach)
[22] Barakat R and Blake J 1980 *Phys. Rep.* **60** 225
[23] Mandel L and Wolf E 1965 *Rev. Mod. Phys.* **37** 231 Sec. 6.2
[24] Mandel L 1979 *Opt. Lett.* **4** 205