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Time factorization of the higher-order intensity correlation functions in the theory of resonance fluorescence

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The intensity correlation functions $\mathrm{Tr}\{\rho(0)s^+(t)s^+(t+\tau_1)\cdots s^+(t+\Sigma_{i=1}^n\tau_i)s^-(t+\Sigma_{i=1}^n\tau_i)\cdots s^-(t+\tau_1)s^-(t)\}$ associated with a two-level atom undergoing Markovian dynamics [$s^\pm(t)$ being the spin-1/2 operators for the atom] are shown to factorize in the form $f(t)\Pi_{i=1}^n g(\tau_i)$ with $f(t)$ [$g(t)$] giving the probability of finding the atom in the excited state when initially it is in the state $\rho(0)$ [ground state].

The second-order intensity correlation function of the radiation emitted by a two-level atom which is also driven by a strong coherent field has been found to have the time-factorization property[1-3]

$$\langle E_i^{(-)}(t)E_j^{(-)}(t+\tau)E_k^{(+)}(t+\tau)E_l^{(+)}(t)\rangle=\alpha_{ijkl}f(t)g(\tau)\,,\qquad(1)$$

where $\vec{E}^{(+)}$ and $\vec{E}^{(-)}$ are the positive- and negative-frequency parts of the field operator $\vec{E}$ and α_{ijkl} is a geometrical factor. The correlation function (1) is proportional to the probability of simultaneously detecting two photons, one at time t and the other at time $t+\tau$. The function $g(\tau)$ is found to be identical to the probability of finding the atom in the excited state if initially it was in the ground state. The intensity correlation function appearing in (1) has been used to discuss the antibunching effects[1-3] in the theory of resonance fluorescence.[4]

In this note we prove the following theorem: Atomic correlation functions of the type

$$\Gamma_{n+1}\equiv\left\langle s^+(t)s^+(t+\tau_1)s^+(t+\tau_1+\tau_2)\cdots s^+\left(t+\sum_{i=1}^n\tau_i\right)s^-\left(t+\sum_{i=1}^n\tau_i\right)\cdots s^-(t+\tau_1)s^-(t)\right\rangle\quad(\tau_i>0)\,,$$

where the $s^\pm(t)$ are the spin-$\frac{1}{2}$ operators associated with the two-level atom, factorize as follows:

$$\left\langle s^+(t)s^+(t+\tau_1)s^+(t+\tau_1+\tau_2)\cdots s^+\left(t+\sum_{i=1}^n\tau_i\right)s^-\left(t+\sum_{i=1}^n\tau_i\right)\cdots s^-(t)\right\rangle=f(t)\prod_{i=1}^n g(\tau_i)\,,\qquad(2)$$

in which $\langle\ \rangle$ denotes the ensemble average over the initial density matrix $\rho(0)$ of the atomic system. In Eq. (2) $g(\tau)$ gives the probability that the atom is found in the excited state if at time $t=0$ it was in the ground state, whereas $f(t)$ gives the probability of finding the atom in the excited state if initially it was the state $\rho(0)$. The above has been derived under the assumption that the dynamics of the two-level system is Markovian and by using the properties of spin-$\frac{1}{2}$ algebra:

$$\begin{gathered}s^+s^-=\tfrac{1}{2}+s^z,\quad s^{+2}=s^{-2}=0\,,\\ -s^zs^+=s^+s^z=-\tfrac{1}{2}s^+,\quad s^zs^-=-s^-s^z=-\tfrac{1}{2}s^-\,.\end{gathered}\qquad(3)$$

The above theorem is immediately applicable to the case of the resonance fluorescence as the dynamics of the two-level atom is taken to be Markovian. The result for the intensity correlations follows from (2) and from the following result[10-12] between the positive frequency part of the electric field operator in the far zone and the atomic operators:

$$E_i^{(+)}(\vec{r},t)\sim E_{0i}^{(+)}(\vec{r},t)+\beta_i s^{(-)}(t)\,,\qquad(4)$$

where β_i is a geometrical factor given by

$$\beta_i=\sum_j d_j\frac{\omega^2}{c^2}(\delta_{ij}-\hat{r}_i\hat{r}_j)\frac{e^{i(\omega/c)r}}{r}\quad(\hbar=1)\,,\qquad(5)$$

where $\vec{d}$ is the dipole-moment matrix element connecting the two states of the two-level atom having energy separation ω.

The reduced density operator of the two-level atom is assumed to satisfy the following Markovian master equation:

$$\frac{\partial\rho}{\partial t}=+\mathcal{L}\rho(t)\,,\qquad(6)$$

where $\mathcal{L}$ is the appropriate time-independent Liouville operator. For example, in the theory of resonance fluorescence $\mathcal{L}$ has the form[6]

$$\begin{aligned}\mathcal{L}G=&-i[(\omega-\omega_0)s^z-\tfrac{1}{2}|\vec{d}|E_0(s^++s^-),G]\\&-\gamma(s^+s^-G-2s^-Gs^++Gs^+s^-)\,,\end{aligned}\qquad(7)$$

where the driving field has been taken to be $E_0 e^{-i\omega_0 t}$. The dipole moment $\langle s^{\pm}(t)\rangle$ and the energy $\langle s^z(t)\rangle$ obey a set of linear equations, and it is clear that the solution for $\langle s^z(t)\rangle$ can be expressed in the form

$$\begin{aligned}\langle s^z(t+\tau)\rangle &= A(\tau)\langle s^+(t)\rangle + A^*(\tau)\langle s^-(t)\rangle \\ &\quad + B(\tau)\langle s^z(t)\rangle + C(\tau)\,. \qquad (8)\end{aligned}$$

We now make use of the quantum regression theorem[13] to obtain for the two-time correlation functions

$$\begin{aligned}\langle s^+(t)s^z(t+\tau)s^-(t)\rangle &= A(\tau)\langle s^+(t)s^+(t)s^-(t)\rangle \\ &\quad + A^*(\tau)\langle s^+(t)s^-(t)s^-(t)\rangle \\ &\quad + B(\tau)\langle s^+(t)s^z(t)s^-(t)\rangle \\ &\quad + C(\tau)\langle s^+(t)s^-(t)\rangle\,,\end{aligned}$$

which on using the operator algebra (3) simplifies to

$$\langle s^+(t)s^z(t+\tau)s^-(t)\rangle = [C(\tau) - \tfrac{1}{2}B(\tau)]\langle s^+(t)s^-(t)\rangle\,, \qquad (9)$$

and hence

$$\begin{aligned}&\langle s^+(t)s^+(t+\tau)s^-(t+\tau)s^-(t)\rangle \\ &\qquad = \tfrac{1}{2}\langle s^+(t)s^-(t)\rangle + \langle s^+(t)s^z(t+\tau)s^-(t)\rangle \\ &\qquad = \langle s^+(t)s^-(t)\rangle[\tfrac{1}{2} + C(\tau) - \tfrac{1}{2}B(\tau)] \\ &\qquad = f(t)g(\tau) \equiv \Gamma_2(t, t+\tau)\,, \qquad (10)\end{aligned}$$

with

$$f(t) = \langle s^+(t)s^-(t)\rangle\,, \qquad (11)$$

$$g(\tau) = \tfrac{1}{2} + C(\tau) - \tfrac{1}{2}B(\tau)\,. \qquad (12)$$

We have thus shown the factorization property of the second-order intensity correlation using the operator algebra (3) and the Markovian dynamics. It is clear that the function $g(\tau)$ does not depend on the initial state of the atomic system (i.e., it depends only on dynamics), whereas $f(t)$ does. $f(t)$ obviously gives the probability of finding the atom in the excited state if at time $t=0$ it was in the state $\rho(0)$. Let us now examine the function $g(\tau)$.

From (8) one finds that

$$\begin{aligned}f(t) &\equiv \langle s^+(t)s^-(t)\rangle = \tfrac{1}{2} + \langle s^z(t)\rangle \\ &= A(t)\langle s^+(0)\rangle + A^*(t)\langle s^-(0)\rangle + B(t)[\langle s^z(0)\rangle + \tfrac{1}{2}] + g(t)\,,\end{aligned} \qquad (13)$$

and thus if at time $t=0$ the atom is in the ground state $\langle s^+(0)\rangle = \langle s^-(0)\rangle = 0$, $\langle s^z(0)\rangle = -\frac{1}{2}$, then

$$\langle s^+(t)s^-(t)\rangle = g(t)\,. \qquad (14)$$

Hence $g(t)$ gives the probability of finding the atom in the excited state given that it was in the ground state at time $t=0$. It is perhaps also interesting to note that

$$\lim_{t\to\infty} f(t) = \lim_{t\to\infty} g(t)\,, \quad g(0) = 0\,. \qquad (15)$$

The second property follows from the definition of $g(\tau)$ and the first one from the fact that the steady state should be independent of the initial condition. In view of (15) one has the following relation for the variance of the intensity correlation in the stationary state:

$$\frac{\lim_{t\to\infty}\langle \Delta s^+(t)\Delta s^+(t+\tau)\Delta s^-(t+\tau)\Delta s^-(t)\rangle}{[\lim_{t\to\infty}\langle s^+(t)s^-(t)\rangle]^2} = \frac{g(\tau)}{g(\infty)} - 1\,. \qquad (16)$$

Hence the variance of the intensity correlation goes to -1 in the limit $\tau \to 0$. The approach to -1 can also be studied rather easily. The Liouville operator, in general, will have the structure

$$\mathcal{L}G = AG + GA^+ + BGC + C^+GB^+\,, \qquad (17)$$

where A, B, and C are functions of the atomic operators. On using the orthogonality of the states $|+\rangle$ and $|-\rangle$ we obtain the result

$$g'(0) = \langle + | B | - \rangle\langle - | C | + \rangle + \text{c.c.} \qquad (18)$$

In particular, in case of resonance fluorescence [$\mathcal{L}$ given by Eq. (7)] $g'(0) = 0$.

The proof for the higher-order correlation functions is similar to the one given above. We use the operator algebra to write the $(n+1)$th-order correlation as

$$\begin{aligned}\Gamma_{n+1}\Big(t, t+\tau_1, \ldots, t+\sum_{i=1}^{n}\tau_i\Big) &= \frac{1}{2}\Gamma_n\Big(t, t+\tau_1, \ldots, t+\sum_{i=1}^{n-1}\tau_i\Big) \\ &\quad + \Big\langle s^+(t)\cdots s^+\Big(t+\sum_{i=1}^{n-1}\tau_i\Big)s^z\Big(t+\sum_{i=1}^{n}\tau_i\Big)s^-\Big(t+\sum_{i=1}^{n-1}\tau_i\Big)\cdots s^-(t)\Big\rangle\,,\end{aligned}$$

which by using the quantum regression theorem and the operator algebra reduces to

$$\Gamma_{n+1}\Big(t, t+\tau_1, \ldots, t+\sum_{i=1}^{n}\tau_i\Big) = g(\tau_n)\Gamma_n\Big(t+t+\tau_1+\cdots+t+\sum_{i=1}^{n-1}\tau_i\Big)\,. \qquad (19)$$

Eq. (19) relates the $(n+1)$th-order intensity correlation to the nth-order intensity correlation; hence by the repeated use of (19) we arrive at the result

$$\Gamma_{n+1}\left(t, t+\tau_1, \ldots, t+\sum_{i=1}^{n} \tau_i\right) = f(t) \prod_{i=1}^{n} g(\tau_i)\,. \tag{20}$$

On combining (4) and (20) we get the following result for the intensity correlations in the theory of resonance fluorescence:

$$\langle E_{i_1}^{(-)}(t) E_{i_2}^{(-)}(t+\tau_1) \cdots E_{i_{n+1}}^{(-)}\left(t+\sum \tau_i\right) E_{j_{n+1}}^{(+)}\left(t+\sum \tau_i\right) \cdots E_{j_1}^{(+)}(t)\rangle = f(t) \prod_{i=1}^{n} g(\tau_i) \left(\prod_{\alpha=1}^{n+1} \beta_{i_\alpha}^{*} \beta_{j_\alpha}\right). \tag{21}$$

Finally, we would like to add that the arbitrary type of correlations associated with the two-level atom have much more complicated structure; e.g., the two-time correlation function $\langle s^{+}(t) s^{-}(\tau)\rangle$ will, in general, be of the form $\sum_i \alpha_i(t)\beta_i(\tau)$. Moreover, results analogous to (2) do not appear to hold for multilevel atoms, presumably because of the interference effects.

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Note added in proof. In the case when $\mathcal{L}$ is time dependent then the right-hand side in Theorem (2) should be replaced by

$$f(t) \prod_{i=1}^{n} g\left(t+\sum_{j=1}^{i} \tau_j, t+\sum_{j=1}^{i-1} \tau_j\right),$$

where $g(t_1, t_2)$ now gives the probability of finding the atom in the excited state at time t_1, if it was in the ground state at earlier time t_2. Note that $\mathcal{L}$ would be time dependent if, for example, one was studying the dynamics of a two-level atom in a bichromatic field [cf. R. G. Gush and H. P. Gush, Phys. Rev. A 10, 1474 (1974)]. It should also be noted that the theorem remains valid if other relaxation mechanisms, such as collisional relaxation are included in the dynamics [cf. J. L. Carlsten and A. Szöke, Phys. Rev. Lett. 36, 667 (1976); B. R. Mollow, Phys. Rev. A 2, 76 (1970)].

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