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LETTER TO THE EDITOR

Correlation functions and the AC Stark effect†

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Abstract. We present a Heisenberg picture QED treatment of the resonance fluorescence of a two-level atom. It is shown that the equations of motion for first-order correlation functions have an asymmetry with respect to the two time arguments. This asymmetry arises in a correct and careful evaluation of the unequal-time commutator between free-field and atomic operators. General solutions of these equations are found without going to the stationary limit.

A deceptively simple-sounding problem has recently received much attention. That problem is the behaviour of a two-level atom interacting with a constant amplitude monochromatic field tuned near resonance. The calculation of the spectral density of the fluorescent light has received particular attention (see Kimble and Mandel 1975a, b and references therein), especially since the report of recent experiments observing the characteristic three-peaked spectrum of the AC Stark effect by Schuda *et al* (1974). This spectrum is of some fundamental interest as it demonstrates the existence of quantum fluctuations in the atomic radiation. This interest makes it desirable to examine carefully the assumptions and approximations made in the theoretical derivations.

Mollow (1969) has carried out a semiclassical treatment in which he assumed that the interaction is a stationary Markovian process. Oliver *et al* (1971), Agarwal (1974), Milonni (1974), Smithers and Freedhoff (1974), Carmichael and Walls (1975), Hassan and Bullough (1975) and Mollow (1975a) have approached the problem using fully quantized formalisms and end up with equivalent results.

If one carries out such a calculation in the Heisenberg picture, one inevitably encounters atomic correlation functions which must be evaluated to determine the spectrum of the scattered light. Some previous attempts (Oliver 1971, Agarwal 1974) to solve this problem have avoided the difficult task of explicitly calculating these correlation functions and the associated two-time operator products by appealing to the fluctuation-regression theorem (Lax 1963, Haake 1973). Another approach sometimes used (Milonni 1974, Hassan and Bullough 1975) is to assume that the free-field operators commute with the atomic variables at all times, which is similar to assuming a c-number external field (Mollow 1969, Herrmann *et al* 1973). Other techniques are used, but none of these approaches has been justified from first principles, so that if one wishes to be sure of the exact predictions to be compared with experiment, one must determine the correlation functions directly. (This point has

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been mentioned by Hassan and Bullough (1975), who were unable to carry out the calculation. Mollow (1975b) also seems to be aware of this problem.)

The purpose of this letter is to carry out a careful and direct evaluation of these correlation functions by finding the equations of motion for the two-time operators themselves. It is shown that the assumptions in most, but not all, of the previous treatments are justified, when one actually determines the behaviour of the particular unequal-time commutators between the free-field and atomic operators.

Consider a two-level atom interacting with a field which initially has only one mode populated. The atomic lowering and raising operators σ_- and σ_+, and the projection operators σ_{22} and σ_{11}, for the upper and lower atomic states respectively, are defined in the usual way (Ackerhalt and Eberly 1974). Each field mode (wavevector $\boldsymbol{k}$, polarization $\lambda = 1,2$) is characterized by a frequency ω_k and annihilation and creation operators $a_{k\lambda}$ and $a_{k\lambda}^\dagger$. The total Hamiltonian in the rotating-wave approximation is

$$\mathscr{H} = \tfrac{1}{2}\hbar\Omega(\sigma_{22} - \sigma_{11}) + \sum_\lambda \int \mathrm{d}\boldsymbol{k}\hbar\omega_k(a_{k\lambda}^\dagger a_{k\lambda} + \tfrac{1}{2}) - \mathrm{i}\hbar \sum_\lambda \int \mathrm{d}\boldsymbol{k}(g_{k\lambda}\sigma_+ a_{k\lambda} - g_{k\lambda}^* a_{k\lambda}^\dagger \sigma_-) \quad (1)$$

where Ω is the atomic energy separation and $g_{k\lambda}$ is the usual coupling constant. The interaction Hamiltonian is written in a kind of normal order in which photon annihilation operators are placed to the extreme right and creation operators to the extreme left of atomic operators. This ordering leads to simplifications when expectation values are taken, as is discussed by Ackerhalt and Eberly (1974).

The operator equations of motion follow directly:

$$\dot{\sigma}_{22}(t) = -\sum_\lambda \int \mathrm{d}\boldsymbol{k}(g_{k\lambda}\sigma_+ a_{k\lambda} + g_{k\lambda}^* a_{k\lambda}^\dagger \sigma_-) \quad (2a)$$

$$\dot{\sigma}_-(t) = -\mathrm{i}\Omega\sigma_- + \sum_\lambda \int \mathrm{d}\boldsymbol{k}\, g_{k\lambda}(\sigma_{22} - \sigma_{11})a_{k\lambda} \quad (2b)$$

$$\dot{a}_{k\lambda}(t) = -\mathrm{i}\omega_k a_{k\lambda} + g_{k\lambda}^* \sigma_- . \quad (2c)$$

The field equation (2*c*) can be formally integrated from the initial time $t = 0$ of the interaction:

$$a_{k\lambda}(t) = a_{k\lambda}(0)\exp(-\mathrm{i}\omega_k t) + g_{k\lambda}^* \int_0^t \sigma_-(t')\exp[-\mathrm{i}\omega_k(t - t')]\,\mathrm{d}t'. \quad (3)$$

The interpretation of these two terms as free-field and source-field is discussed by Ackerhalt and Eberly (1974). When the expression (3) is substituted for the field operators in equations (2*a*, *b*), the atomic operator equations are found to contain only initial-time field operators.

It is customary at this point to make the *harmonic approximation* in which the lowering operator evolves as it would in the absence of any atom–field coupling: $\sigma_-(t_1) = \sigma_-(t_2)\exp[-\mathrm{i}\Omega(t_1 - t_2)]$. The validity of this approximation is restricted to times much shorter than a Rabi period or a natural lifetime. It will only be employed in terms such as those in equation (4), where the σ_- appears under a double integral over time and frequency,

$$\int_0^\infty \mathrm{d}\omega f(\omega) \int_0^t \sigma_-(t_1)\exp[-\mathrm{i}\omega(t_2 - t_1)]\,\mathrm{d}t_1$$
$$\simeq \sigma_-(t_2) \int_0^\infty \mathrm{d}\omega f(\omega) \int_0^t \exp[-\mathrm{i}(\omega - \Omega)(t_2 - t_1)]\,\mathrm{d}t_1 \quad (4)$$

and where $f(\omega)$ is assumed to be a well behaved function which takes care of all possible divergences. The harmonic approximation is justified here, because the frequency integral behaves like a delta function in time $\delta(t_2 - t_1)$ with an effective width of the order of a few optical periods. When $t_2 = t$, the upper limit in equation (4), the usual treatment in the dipole approximation leads to a decay constant and a frequency shift (Ackerhalt and Eberly 1974).

With these substitutions, the atomic equations of motion become:

$$\dot{\sigma}_{22}(t) = -2\beta\sigma_{22} - \sum_{\lambda} \int \mathrm{d}\boldsymbol{k}\,[g_{\boldsymbol{k}\lambda}\sigma_+ a_{\boldsymbol{k}\lambda}(0)\exp(-\mathrm{i}\omega_k t) + g^*_{\boldsymbol{k}\lambda} a^\dagger_{\boldsymbol{k}\lambda}(0)\sigma_- \exp(\mathrm{i}\omega_k t)] \tag{5a}$$

$$\dot{\sigma}_-(t) = -(\beta + \mathrm{i}\bar{\Omega})\sigma_- + \sum_{\lambda} \int \mathrm{d}\boldsymbol{k} g_{\boldsymbol{k}\lambda}(\sigma_{22} - \sigma_{11}) a_{\boldsymbol{k}\lambda}(0)\exp(-\mathrm{i}\omega_k t). \tag{5b}$$

The atomic frequency including the Lamb shift is denoted by $\bar{\Omega}$, and β is half the Einstein A coefficient.

We find it convenient to remove the oscillations at optical frequency by transforming to slowly varying operators:

$$\chi_{11}(t) = \sigma_{11}(t), \qquad \chi_{22}(t) = \sigma_{22}(t) \qquad \text{and} \qquad \chi_\pm(t) = \sigma_\pm(t)\exp(\mp\mathrm{i}\omega_0 t).$$

After taking expectation values in a product state of a monochromatic coherent state of frequency ω_0 for the field and an arbitrary state for the two-level system, we find a set of linear equations. In matrix notation, they take the form:

$$\dot{\boldsymbol{V}}(t) = \frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix}\langle\chi_{22}(t)\rangle\\ \langle\chi_-(t)\rangle\\ \langle\chi_+(t)\rangle\\ \langle\chi_{11}(t)\rangle\end{bmatrix} = \begin{bmatrix}-2\beta & -\tfrac{1}{2}\epsilon^* & -\tfrac{1}{2}\epsilon & 0\\ \tfrac{1}{2}\epsilon & -\beta + \mathrm{i}\alpha & 0 & -\tfrac{1}{2}\epsilon\\ \tfrac{1}{2}\epsilon^* & 0 & -\beta - \mathrm{i}\alpha & -\tfrac{1}{2}\epsilon^*\\ 2\beta & \tfrac{1}{2}\epsilon^* & \tfrac{1}{2}\epsilon & 0\end{bmatrix} \cdot \begin{bmatrix}\langle\chi_{22}(t)\rangle\\ \langle\chi_-(t)\rangle\\ \langle\chi_+(t)\rangle\\ \langle\chi_{11}(t)\rangle\end{bmatrix} = \boldsymbol{A}\cdot\boldsymbol{V}(t) \tag{6}$$

where $\boldsymbol{V}(t)$ and $\boldsymbol{A}$ have the obvious definitions, $\alpha = \omega_0 - \bar{\Omega}$ is the detuning and ϵ is the Rabi frequency. The solution of equation (6) is straightforward,

$$\boldsymbol{V}(t) = \sum_{m=1}^{4} \boldsymbol{Q}(s_m)\cdot\boldsymbol{V}(0)\exp(s_m t) \tag{7}$$

where it can be shown that the 4×4 matrix $\boldsymbol{Q}(s_m)$ is given by

$$\boldsymbol{Q}(s_m) = -\lim_{s\to s_m}[(s - s_m)(\boldsymbol{A} - s\boldsymbol{I})^{-1}] = \boldsymbol{v}_m\cdot\boldsymbol{w}^\dagger_m \tag{8}$$

where s_m, $\boldsymbol{v}_m$ and $\boldsymbol{w}_m$ $(m = 1, \ldots, 4)$ are the eigenvalues, eigenvectors and reciprocal eigenvectors respectively of $\boldsymbol{A}$, and $\boldsymbol{I}$ is the 4×4 identity matrix. (Note that the product $\boldsymbol{v}_m\cdot\boldsymbol{w}^\dagger_m$ is a 4×4 matrix, since both $\boldsymbol{v}_m$ and $\boldsymbol{w}_m$ are column vectors.) Similar treatments have been given by Kimble and Mandel (1975) and Hassan and Bullough (1975).

The spectral density of the fluorescent light is related to the mean photon number in a given field mode $\boldsymbol{k},\lambda$ (excluding the incident field mode). Since the $\boldsymbol{k},\lambda$ mode is initially unoccupied, one finds directly from equation (3) that

$$\langle n_{\boldsymbol{k}\lambda}(t)\rangle = |g_{\boldsymbol{k}\lambda}|^2 \int_0^t \int_0^t \langle\sigma_+(t'')\sigma_-(t')\rangle \exp[-\mathrm{i}\omega_k(t'' - t')]\,\mathrm{d}t'\,\mathrm{d}t''. \tag{9}$$

We shall derive equations of motion for general two-time correlation functions $\langle\sigma(t)\sigma_-(t')\rangle$. A particular example appears in equation (9), where $\sigma = \sigma_+$. The time derivatives will be taken with respect to t', the time argument of the operator on the

right. This is easily done by considering all time arguments in equations (5*a*,*b*) to be t' and then multiplying from the left by $\sigma(t)$. In order to put these equations in the normal order explained previously (with the field creation operators to the extreme left), we must evaluate the commutation expression:

$$\kappa(t,t') = \sum_{\lambda} \int \mathrm{d}\boldsymbol{k}\, g^{*}_{\boldsymbol{k}\lambda}\,[\sigma(t), a^{\dagger}_{\boldsymbol{k}\lambda}(0)]\exp(\mathrm{i}\omega_k t'). \tag{10}$$

The creation operator can be expressed according to equation (3). Since the equal-time atom–field commutator is zero, it follows that

$$\kappa(t,t') = -\sum_{\lambda} \int \mathrm{d}\boldsymbol{k}|g_{\boldsymbol{k}\lambda}|^2\left[\sigma(t), \int_0^t \sigma_+(t_1)\exp\{\mathrm{i}\omega_k(t'-t_1)\}\,\mathrm{d}t_1\right]. \tag{11}$$

Now we can use the harmonic approximation as in equation (4), noting that the singularity of the frequency integral occurs at $t_1 = t'$. Since t_1 can be within or outside the interval of integration $[0, t]$, $\kappa(t,t')$ is proportional to a step function in time. Assuming that t' as well as $t - t'$ are much larger than an optical period, equation (11) becomes:

$$\kappa(t,t') = -2\beta[\sigma(t), \sigma_+(t')]\,U(t-t'). \tag{12}$$

Here $U(t-t')$ is one for $(t-t') > 0$ and zero for $(t-t') < 0$. Equation (12) is not valid for $t = t'$.

Choosing the same initial state as before, we find that the correlation function equations of motion for the slowly varying operators read

$$\frac{\mathrm{d}}{\mathrm{d}t'}\langle\chi(t)\chi_{22}(t')\rangle = -\frac{\epsilon}{2}\langle\chi(t)\chi_+(t')\rangle - \frac{\epsilon^*}{2}\langle\chi(t)\chi_-(t')\rangle - 2\beta\langle\chi(t)\chi_{22}(t')\rangle$$
$$+ \langle[\chi(t), \chi_+(t')]\chi_-(t')\rangle\, 2\beta\, U(t-t') \tag{13a}$$

$$\frac{\mathrm{d}}{\mathrm{d}t'}\langle\chi(t)\chi_-(t')\rangle = \frac{\epsilon}{2}(\langle\chi(t)\chi_{22}(t')\rangle - \langle\chi(t)\chi_{11}(t')\rangle) - (\beta - \mathrm{i}\alpha)\langle\chi(t)\chi_-(t')\rangle \tag{13b}$$

$$\frac{\mathrm{d}}{\mathrm{d}t'}\langle\chi(t)\chi_+(t')\rangle = \frac{\epsilon^*}{2}(\langle\chi(t)\chi_{22}(t')\rangle - \langle\chi(t)\chi_{11}(t')\rangle) - (\beta + \mathrm{i}\alpha)\langle\chi(t)\chi_+(t')\rangle$$
$$- (\langle[\chi(t), \chi_+(t')]\chi_{22}(t')\rangle - \langle[\chi(t), \chi_+(t')]\chi_{11}(t')\rangle)2\beta U(t-t') \tag{13c}$$

$$\frac{\mathrm{d}}{\mathrm{d}t'}\langle\chi(t)\chi_{11}(t')\rangle = -\frac{\mathrm{d}}{\mathrm{d}t'}\langle\chi(t)\chi_{22}(t')\rangle. \tag{13d}$$

This system of equations exhibits a very important asymmetry in $(t-t')$, due to the presence of terms containing the step function. For $t' > t$ the set is closed as written. For $t' < t$ additional terms appear which have been omitted in previous treatments. We will investigate the solution in both regimes.

When $t' > t$ the equations of motion can be written (cf equation (6))

$$\frac{\mathrm{d}}{\mathrm{d}t'}\boldsymbol{W}(t,t') = \frac{\mathrm{d}}{\mathrm{d}t'}\begin{bmatrix}\langle\chi(t)\chi_{22}(t')\rangle\\ \langle\chi(t)\chi_-(t')\rangle\\ \langle\chi(t)\chi_+(t')\rangle\\ \langle\chi(t)\chi_{11}(t')\rangle\end{bmatrix} = \boldsymbol{A}\,.\,\boldsymbol{W}(t,t') \tag{14}$$

where the definition of $W(t,t')$ is obvious and A is the same matrix that appears in equation (6). The solution is

$$W(t,t') = \sum_{l=1}^{4} Q(s_l).W(t,t)\exp[s_l(t'-t)] \tag{15}$$

with s_l and Q the same quantities appearing in equation (7). The same-time vector $W(t,t)$ is trivially found to be $T.V(t)$. Here T is a simple 4×4 matrix which depends upon the choice for $\chi(t)$. For example, if $\chi(t)$ is $\chi_+(t)$, then the (2,1) and (4,3) elements of T are one and all other elements are zero. By substituting the above expression for $W(t,t)$ into equation (15), we can write the correlation functions in terms of single-time expectation values, where the time is the earlier of the two times in the correlation functions. This is just the form prescribed by the fluctuation-regression theorem, although we did not make use of this theorem.

Finally, using equation (7) to bring in the initial conditions explicitly, one finds

$$W(t,t') = \sum_{l=1}^{4}\sum_{m=1}^{4} [Q(s_l).T.Q(s_m).V(0)]\exp[s_l(t'-t)]\exp(s_m t) \qquad t'>t. \tag{16}$$

Because the real parts of the eigenvalues s_l are non-positive, it can be seen from equation (16) that in the limit of large time, for $t-t'$ kept constant, $W(t,t')$ becomes a function only of the time difference. This shows that the interaction process becomes stationary in the wide sense. It is then simple to calculate the time derivative of $\langle n_{k\lambda}(t)\rangle$ in equation (9) in the limit of t going to infinity and recover Mollow's stationary emission spectrum for a two-level atom.

When $t'<t$ the step function in equation (13) is equal to one and new terms appear. Some can be reduced to two-operator product expectation values, but others remain in a sandwich form where the operator at time t is between operators at time t'. They are $\langle\chi_+(t')\chi(t)\chi_{22}(t')\rangle$, $\langle\chi_+(t')\chi(t)\chi_-(t')\rangle$ and $\langle\chi_+(t')\chi(t)\chi_{11}(t')\rangle$. These equations are solved by taking derivatives with respect to t' of all 16 different possible sandwich terms (for a given $\chi(t)$). This set of equations is closed and linear. Defining a 16×1 vector $Y(t,t')$ in analogy with $W(t,t')$, but with each element now a sandwich correlation function, we can write the equations of motion as follows:

$$\frac{\mathrm{d}}{\mathrm{d}t'}Y(t,t') = B.Y(t,t') \qquad t'<t. \tag{17}$$

With a suitable choice of basis in our 16-dimensional space, B assumes the form $B=(I\otimes A - A^{\mathrm{T}}\otimes I)$. I is the 4×4 identity matrix, A is as defined in equation (6), $\otimes$ denotes a direct product, and A^{T} is the transpose of A. The particular form of B allows us to write the solution in terms of s_m and $Q(s_m)$ of A, which we have previously defined. After some manipulation, we find the solution

$$Y(t,t') = \sum_{l=1}^{4}\sum_{m=1}^{4} [(Q(s_l).\hat{e}_j)\otimes(Q(s_m).V(0))]\exp(s_l t)\exp[(s_m-s_l)t'] \qquad t'<t \tag{18}$$

where $\hat{e}_j$ is a 4×1 unit vector and the j specifies the particular $\chi(t)$ which appears in $Y(t,t')$ ($\langle\chi\rangle$ is the jth component of $V(t)$ in equation (7)). This relation is quite general for $t'<t$ and gives the behaviour of all 64 possible sandwich correlation functions, whatever the initial atomic conditions are.

That solutions (16) and (18) are consistent can be seen by a direct comparison. For example, consider $\langle\chi_+(t_1)\chi_-(t_2)\rangle$, $t_1<t_2$. This expression as given by equation (16)

is seen to be identical to $\langle\chi_+(t_1)\chi_-(t_2)\chi_{11}(t_1)\rangle + \langle\chi_+(t_1)\chi_-(t_2)\chi_{22}(t_1)\rangle$, $t_1 < t_2$, as given by equation (18).

Of course, the usefulness of the 16-dimensional space of sandwich correlations lies in the insight it provides into the question of time ordering in the evaluation of correlation functions. Furthermore, it shows the consistency of equations (13) in the two time regimes $t' \gtrless t$. This involved formalism need not be used in any particular first-order correlation function calculation. As long as one takes derivatives with respect to the larger time first, one is working in the domain where the step function $U(t, t')$ is zero and therefore one remains in the 4-dimensional space of the single-time operator expectation values. This explains why some previous treatments have yielded the same results, even when incorrect or unjustified assumptions were made about the behaviour of the unequal-time commutators.

But one must always be aware of the time ordering. For example, suppose that equations (13) with $U(t, t') = 0$ properly describe the evolution of $\langle\chi_+(t)\chi_-(t')\rangle$ when $t' < t$. This would lead to the prediction that the number of photons scattered by the atom in any mode diverges exponentially in time. This cannot be. This illustrates one possible consequence of assuming the commutator of equation (10) to be negligible or of higher order.

The results presented here will be used in a future paper to evaluate higher-order correlation functions. There we will also discuss non-stationary effects including the proper emission spectrum calculation for an experimental situation in which transients are important.

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