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Density operator with coherent, number, and squeezed states

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A new approach to developing the series expansion of the density operator of quantum optics is given, and the result is applied to finding the second-order polarization vector. Coherent, number, and squeezed states are each investigated and the results compared.

The cornerstone of nonlinear optics is the nonlinear susceptibility, and, since Bloembergen's pioneering work,[1] much effort has gone into the theoretical (as well as experimental) evaluation of the nonlinear susceptibilities. Generally, in these calculations, the incident radiation field is assumed to take on its classical form, the justification owing itself to the relatively large magnitude of the incident field. Recently, however, the second-order susceptibility was calculated for a fully quantized system using coherent states, and it was shown, for example, that the second-order polarization vector carries information about the phase of the coherent beam.[2,3] Thus quantum-mechanical treatments do yield information not carried by the classical approximation. Moreover, there are some situations, such as squeezed states, that cannot be described classically. For these reasons, a quantum-optical calculation of the polarization vector to second order will be carried out, and in doing so, various states of the radiation field will be considered. Also, a new way to obtain the density operator will be given which significantly reduces the computations involved in finding higher-order terms. Depending on the circumstances, one may wish to use number states, coherent states, or squeezed states to describe the radiation field, and, using the developments described above, results for these various cases will be compared.

The use of coherent states to describe nonlinear-optical processes is a natural choice since these states well describe laser radiation. Moreover, coherent states are often more realistic descriptions than number states. For example, if one computes the expectation value of a free electric field, a coherent-state representation yields a result resembling a classical field, while a null result is obtained using the number states. This simple example gives a case where the result of a number-state calculation does not go over to the classical result in the limit of large n (number of photons) whereas the coherent-state calculation does. The question presents itself: Are there other situations where number states are inadequate to describe a physical process, but where coherent states are suitable? It will be shown that the answer is yes, and in particular, a situation is examined whereby coherent states give frequency doubling while number states do not.

In addition to coherent states, squeezed states are becoming increasingly important. Ever since their discovery by Yuen,[4] squeezed states of the radiation field have received considerable attention, and afford a precision in measurements that transcends the limit that was previously held to exist,[5] and recently, there has been interest and theoretical work done in high-intensity squeezed states.[6] Since squeezing is accomplished in a nonlinear medium, one would expect nonlinear effects *from* squeezed states, if they are intense. Since squeezed states have no classical analog, they may react in a nonlinear way which cannot be accounted for with the classical field approximation.

Coherent states may be defined as eigenstates of the annihilation operator $a_\sigma|\alpha_\sigma\rangle=\alpha_\sigma|\alpha_\sigma\rangle$, where $|\alpha_\sigma|^2$ is the mean number of photons of the coherent state $|\alpha_\sigma\rangle$ of frequency ω_σ. To simplify notation, the above equation, and those to follow, will be written as $a|\alpha\rangle=\alpha|\alpha\rangle$. Writing $\alpha=|\alpha|e^{i\xi}$, ξ is called the phase of the coherent state. Squeezed states may be defined by the equation $b|\beta\rangle=\beta|\beta\rangle$ where $b=\mu a+\nu a^\dagger$. It is also assumed that $|\mu|^2-|\nu|^2=1$. Throughout, naturalized ($\hbar=c=1$) Gaussian units are used.

In view of the foregoing remarks, the first step here is obtain the polarization vector in second order. In order to do this, the density-matrix techniques will be used. With this, one has[1]

$$\langle \mathbf{P}\rangle=\mathrm{Tr}(\rho\mathbf{P}) , \tag{1}$$

where $\mathbf{P}$ is the polarization and ρ is the density matrix, and the trace is over the radiation and atomic fields. One may then assume that $\mathbf{P}=-N\boldsymbol{\mu}$ were N is the number density of elementary dipoles $\boldsymbol{\mu}$, and local field corrections may be made.[7]

From its definition, it follows that the density operator obeys the equations

$$\frac{i\partial\rho}{\partial t}=[H,\rho] , \tag{2}$$

which is in the interaction picture, and where H is the interaction Hamiltonian, which is given, in the Schrödinger picture, by $H^{\mathrm{SP}}=-\boldsymbol{\mu}\cdot\mathbf{E}$. In the standard approach, this equation may be integrated iteratively to obtain ρ to any order.[3]

A different derivation of ρ will be presented here which gives a form that entails less computation, and is in a form usuable for quantum-field-theoretic techniques. The basic idea is to assume that the state vector has a time evolution that is given by

$$|\psi(t)\rangle = U|\psi(t_0)\rangle \ , \tag{3}$$

where $|\psi\rangle$ is in the interaction picture and U is a unitary operator with $U(t_0)=1$. Then, since

$$\rho = \sum_{\psi} c_{\psi}|\psi\rangle\langle\psi| \ , \tag{4}$$

it follows that

$$\rho(t) = U\rho(t_0)U^{\dagger} \ . \tag{5}$$

From (3) however, one has

$$U = T\exp\left[-i\int_{t_0}^{t} H(t')dt'\right] \ , \tag{6}$$

where T stands for chronological product. Thus, using (6) with (5), the density operator may be computated to any order. Although this, ultimately, is equivalent to the conventional formalism, there are two advantages in using (6). In the case of quantum-field theory, assuming that the interaction Hamiltonian is normally ordered, then U is given in terms of the chronological product of normally ordered pairs, which may be reduced to the usual Feynmann terms describing all virtual processes. The other advantage of (6) is that, under certain conditions to be discussed, it immediately yields a form whereby all of the upper limits on the integrals become t, which significantly reduces the amount of work needed to compute the integrals.

Thus, to find the density operator to second order, writing $U = U_0 + U_1 + \cdots$, one has from (5), and the unitarity of U at all orders

$$\rho(t) = \rho_0 + [U_1,\rho_0] + [U_2,\rho_0] + [\rho_0, U_1]U_1 + \cdots \ , \tag{7}$$

where the dots denote third-order terms and higher.

Denoting the second-order part by $\rho^{(2)}$, one obtains from (7)

$$\begin{aligned} i^2\rho^{(2)} = {} & (T/2)\int_{t_0}^{t} dt_1 \int_{t_0}^{t} dt_2 [H(t_1)H(t_2)\rho(t_0) \\ & \qquad - \rho(t_0)H(t_1)H(t_2)] \\ & + \int_{t_0}^{t} dt_1 [\rho(t_0)H(t_1) - H(t_1)\rho(t_0)] \\ & \qquad \times \int_{t_0}^{t} dt_1 H(t_1) \ . \end{aligned} \tag{8}$$

Here, as previously discussed, all upper limits are t, but for actual computation, this expression can be put in the form

$$\begin{aligned} i^2\rho^{(2)} = {} & \int_{t_0}^{t} dt_1 \int_{t_0}^{t} dt_2 \{[\rho(t_0), H(t_1)H(t_2)]/2 \\ & \qquad - H(t_1)\rho(t_0)H(t_2) \\ & \qquad + H(t_1)H(t_2)\rho(t_0)\} \\ & - \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 [H(t_1), H(t_2)]\rho(t_0) \ . \end{aligned} \tag{9}$$

In this expression time ordering has been used, and the only term involving different upper limits contains the commutation of the interaction Hamiltonian at different times. This is a useful form of the density operator because, far from resonance such that the incident frequency ω is large compared to the transition frequencies ω_{if}, this last term in (9) becomes negligible. This can be shown explicitly, but the calculation is not given here. In what follows, this far-from-resonance condition will be assumed to hold so that the last term in (9) will be dropped.

Another standard way to calculate the density operator is to derive the master equation for the reduced density operator. There are generally two approximations that are made in that approach that are not needed in the present course. One is that the Markoff approximation is usually made, and the other is that the rotating-wave approximation is also usually adopted. The new approach presented here obviates the need for these assumptions yet still provides a simple form of the density operator.

Now, (9) can be used, with (1), to find the expectation value of any operator. Consider now $\mathbf{P}$, the polarization vector. To proceed, assume that initially the system is in some definite state, so that $\rho_0 = |I\rangle\langle I|$, where capitals will always represent product states of the radiation and atomic field. For example, $|A\rangle = |a\rangle|\alpha\rangle$ where $|a\rangle$ represents the atomic state and $|\alpha\rangle$ the coherent state. Also, since the atomic and radiation fields are independent, the density operator at $t = t_0 \equiv 0$ may be written in product form, i.e., $\rho_0 = \rho_a(0)\rho_r(0)$ where a and r stand for atomic and radiation part.

In what follows, although it is not necessary, the definite initial-state assumption is used since it avoids unnecessary terms. The effect of the more general ρ_0 has been discussed elsewhere.[3] Finally, it will be assumed that a two-level system is under consideration. For the results given below, the basic conclusion will be the same for a multilevel system, and no real advantage is gained by considering the more complex situation for the purposes here. Thus, using (9) in (1), one obtains

$$i^2\langle\mathbf{P}\rangle = \sum_{A,B} \mathbf{P}_{IA}S_{AB}S_{BI} - \mathbf{P}_{BA}S_{AI}S_{IB} \ , \tag{10}$$

where $S_{AB} = \int_0^t dt' H_{AB}(t')$, and $H_{AB} = \langle A|H|B\rangle$.

The above result will be used to express $\langle\mathbf{P}\rangle$ for three situations. First it will be assumed that the incident radiation is described by a coherent state, which is implemented by assuming that $\rho_r(0) = |\sigma\rangle\langle\sigma|$. Using this, (10) yields

$$\begin{aligned} \langle\mathbf{P}^{(2)}\rangle_{\mathrm{coh}} = {} & (eN/L^3)(\boldsymbol{\Delta}/\omega)|\boldsymbol{\mu}_{\mathrm{if}}\cdot\hat{\mathbf{e}}|^2|\alpha|^2 \\ & \times[2 - 2\cos(\omega t) + \cos 2\xi \\ & \qquad + \cos(2\xi - 2\omega t) - 2\cos(2\xi - \omega t)] \ , \end{aligned} \tag{11}$$

where $\boldsymbol{\Delta} = \mathbf{r}_{ii} - \mathbf{r}_{ff}$, $\mathbf{r}_{ab} = \langle a|\mathbf{r}|b\rangle$, and L^3 is the quantization volume. It is assumed that only two-photon processes occur, for which H_{ab} is different from zero only if $a \neq b$. Details concerning the derivation of (4) may be found elsewhere.[3]

Now consider that the incident radiation is described by a number state, so that $\rho_r(0) = |n\rangle\langle n|$, where $|n\rangle$ represents the number state. One obtains, with (1),

$$\begin{aligned} \langle\mathbf{P}^{(2)}\rangle_{\mathrm{num}} = {} & (eN/L^3)(\boldsymbol{\Delta}/\omega)|\boldsymbol{\mu}_{\mathrm{if}}\cdot\hat{\mathbf{e}}|^2 n \\ & \times[2 - 2\cos(\omega t)] \ , \end{aligned} \tag{12}$$

where n is the number of photons, the approximation $\sqrt{n(n+1)} \cong n$ is used in the eigenvalue, and a spontaneous emission term has been neglected.

It is evident that n replaces $|\alpha|^2$ in the number space result, which is to be expected. The more interesting result is that the number state result (12) contains no frequency-doubled component, while the coherent-state result does.

The reason for this result is not hard to see. When using the coherent-state description, the eigenstates $|\alpha\rangle$, being eigenvalues of the annihilation operator, are indefinite number states. Upon taking the trace, this gives nonzero contributions for the operators a^2 and $(a^\dagger)^2$, while in the number state representation, these contribute nothing. As can be seen from (11), it is precisely these terms that give the frequency-doubled contribution. It is also interesting to note that these are the terms that carry the phase information of the coherent state.

Finally, (10) may be used to find the second-order polarization for squeezed states. The result is given by, using similar manipulations as before and retaining only the frequency-doubled part,

$$\begin{aligned}\langle \mathbf{P}^{(2)} \rangle_{\mathrm{sq}} = {} & (eN/L^3)(\Delta/2\omega)|\boldsymbol{\mu}_{\mathrm{if}} \cdot \hat{\mathbf{e}}|^2 \\ & \times \{ e^{-2i\omega t}[\mu^{*2}\beta^2 + \nu^2\beta^{*2} \\ & - |\beta|^2(\nu\mu^* + \mu^*\nu) - \mu^*\nu] + \text{c.c.} \} \ . \end{aligned} \tag{13}$$

The above result also predicts a dc rectification term that depends on the phase of the squeezed state, as well as the amount of squeezing. This result has been discussed elsewhere for the coherent state and will not be considered further here.[8] Sometimes the quantity $|\beta|^2$ is referred to as the number of generalized quanta. It is seen from that, to within a phase factor, the second harmonic would be proportional to the number of generalized quanta were it not for the presence of the term $\mu^*\nu$ in (13). This would correspond to a kind of spontaneous emission, with regard to the generalized quanta (a true spontaneous emission term for the coherent state was neglected since $|\alpha|^2$ is taken to be large).

As a final comment, it is recognized that the two formulations of the density operator must be equivalent, and the advantage of the new formulation, as explained, lies in its form. Nevertheless, it is worthwhile to explicitly demonstrate the equivalence in second order, and to show how the conventional formulation is more cumbersome.

Successive iteration of (2) yields

$$\begin{aligned} i^2\rho^{(2)} = \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 [& H(t_1)H(t_2)\rho(t_0) \\ & - H(t_1)\rho(t_0)H(t_2) \\ & + \rho(t_0)H(t_2)H(t_1) \\ & - H(t_2)\rho(t_0)H(t_1)] \ , \end{aligned} \tag{14}$$

which represents the conventional approach. The difficulty with (14) is that it is not a time-ordered product and the upper limits on the integrals are different. Direct integration of (14), even after it is used under the trace operation, requires more work than (8) does, and also there are more surviving terms. This problem becomes more severe in higher orders. To show (8) and (13) are equivalent, however, one proceeds as follows. In (8) use Dyson's trick to break up the time-ordered product into two integrals, one from t_1 to t_2 to t and the other from t_2 to t_1 to t. Also in (8), relabel t_1 by t_2 in the second time integral in the second term on the right-hand side. With cancellation and relabeling of time integrals this becomes the form given by (9). This demonstrates their equivalence to second order.

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