Optimum field squeezing from atomic sources: Three-level atoms

B.J. Dalton,[1,2] Z. Ficek,[1] and P.L. Knight[2]
[1] *Department of Physics, The University of Queensland, Queensland 4072, Australia*
[2] *Optics Section, Blackett Laboratory, Imperial College of Science, Technology and Medicine, London SW7 2BZ, United Kingdom*
(Received 23 November 1993)

Light emitted from laser-driven atoms often has squeezed quadrature fluctuations reflecting the phase dependence of the atomic source excited into a coherent superposition by the driving field. In this paper, we study the maximum squeezing in the emitted light fields from three-level atoms, paying particular attention to the role of atomic coherences in governing the optimum squeezing which is possible. Some phase-dependent nonlinear optical processes have the potential for substantial quantum noise reduction at useful intensities: for example, four-wave mixing involving atomic systems has already been used to generate quadrature-squeezed light. The theoretical description of such a source is generally complicated with damping processes, coherent and incoherent pumping, and so on, all needing to be taken into account. Fully quantum treatments are available, but the basic physics of the process is often difficult to grasp in the necessarily complicated analysis. Although less important from the point of view of efficient practical sources of squeezed light, studying the generation of squeezed light in simpler systems such as two- or three-level atoms interacting with one or two modes (as in Jaynes-Cummings models) or in spontaneous emission or resonance fluorescence gives greater insight into the fundamentals of the squeezing process. In particular, the relationship between the atomic coherences and the degree of squeezing in the generated fields can be explored. We study the maximum (optimized) squeezing that can be obtained in the emitted fields from the irreversible decay from three-level atoms for any choice of initial conditions, excitations, and decay process, for all quadrature components, and for all possible configurations (V, Λ, and "ladder" systems). We show that for V and Λ systems, optimum squeezing is actually associated with a two-level state (in a suitable basis) involving a single one-photon coherence, but for ladder systems the state for maximum noise reduction is not equivalent to a two-level system and involves a single two-photon coherence and no intermediate state population. We pay particular attention to whether the total field is in a minimum uncertainty state and the nature of the atomic state associated with maximum squeezing, especially whether it is a mixed or pure state. In the case where the initial free field has a zero amplitude at the detector and the source atoms are confined to a region small compared to the wavelength (Dicke source) we show that the squeezing in the total field is given in terms of the squeezing in the source field and hence related to atomic populations and atomic one-photon and two-photon coherences for the case of three-level atom. Previously unconsidered source-free field interferences terms are shown to be zero. The choice of quadrature phase and frequency is optimized to minimize the source field normally ordered variance. We then minimize further with respect to the choice of atomic density matrix elements subject to the constraints that the density matrix is Hermitean, positive, and has a trace equal to unity. In all cases we find that the optimum squeezing is produced when the source field is in a minimum uncertainty state.

PACS number(s): 42.50.Dv

I. INTRODUCTION

The squeezing properties of the electromagnetic (em) field are defined in terms of its quadrature components measured at a particular frequency and phase [1]. Squeezing occurs when the variance of a quadrature component of the electric field becomes less than that for the vacuum state, and this situation signifies a nonclassical state of the field. In general this variance will be time dependent, though with a suitable choice of the quadrature frequency a stationary contribution to the variance can occur and any high frequency oscillating terms could be averaged away. Apart from its significance as a nonclassical realization of the em field, squeezed light fields exemplify situations of phase-dependent quantum fluctuations or noise and have potential applications in high precision interferometry, optical communications, and atomic spectroscopy [1].

The experimental realization of squeezed states of light may be accomplished using a variety of phase-dependent nonlinear optical processes [1]. Some (including parametric amplifiers [1,2], second harmonic generation [1,3], and four-wave mixing [1,4]) can involve nonresonant interactions between quantized light fields and atomic systems. Others (including two-photon correlated emission lasers [1(c),5] and Rydberg atom lasers driven by coherent fields [1(c),6]) can involve interactions closer to resonance. All involve the atomic systems being subjected to phase-dependent excitation mechanisms resulting in the generation of squeezing in the light fields to which

the atoms are coupled. Nonzero atomic coherences will signify the phase dependences generated in the atomic system along with the phase-dependent features of the field that characterize squeezing.

The theoretical description of the type of system described above is generally complicated with damping processes, coherent and incoherent pumping process, and so on having to be taken into account. In all of the above cases fully quantum mechanical treatments are of course available, as, for example, in four-wave mixing when the fields are close to resonance and spontaneous emission effects need to be included [7]. However, the physics of the atomic systems involved is often lost in the details; indeed for the nonresonant processes the atoms are effectively described in terms of nonlinear optical susceptibilities. Although less important from the point of view of experimentally generating squeezed light, studying the generation of squeezed light in simpler systems such as a two-level atom or a three-level atom interacting with single-mode cavity fields, two-mode cavity fields (as in Jaynes-Cummings models), or coupled to empty multimode fields (as in spontaneous emission and resonance fluorescence) is significant in terms of giving greater insight into the fundamentals of the squeezing generation process. In particular the relation between the atomic coherence properties and the amount of squeezing can be studied.

A large number of studies of such simple systems have been carried out [1] for the *reversible* single- or two-mode field cases (Jaynes-Cummings models) where the squeezing can be related to the phase-dependent initial state of the atom(s) and the field. In these models coupled Heisenberg equations of motion for the cavity electric field operators and the atomic transition operators can be derived. There is no simple relationship between the electric field and atomic operators at the same time and so the squeezing cannot be related in a simple way to the atomic coherences.

Squeezing can, however, be obtained in a variety of situations including (i) an excited two-level atom coupled in a single-mode cavity in a coherent state [8(a)], (ii) a two-level atom excited coherently and coupled to a single-mode cavity in a vacuum state [8(b)], (iii) a three-level cascade atom with the outer states excited coherently coupled to a two-mode cavity in a vacuum state [8(b)], (iv) a pair of two-level atoms in a multiatom (atomic) squeezed state coupled to a single-mode cavity in a vacuum state [8(c)], (v) many two-level atoms coupled to a single mode cavity with a variety of initial conditions (atoms in a coherent state with the cavity in the vacuum state, atoms all in the excited state with the cavity in the coherent state, and atoms all in the ground state with the cavity in a coherent state) [8(d)–8(f)], and so on.

Other studies of simple systems have been carried out [1(c)] for the *irreversible* multimode field cases. Here the total electric field at large distances can be expressed as the sum of a free field and a source-field [9]. The source-field can be related to the atomic transition operators at suitable retarded times. On the basis that the source-field effects can be separated from the free-field effects and that the latter field does not contribute to squeezing, properties of the total field relate to the atomic coherences and populations of the source atoms at the retarded time. The atomic coherences and populations could be in a steady state as in resonance scattering experiments with a constant exciting field or they could be changing as in experiments where coherences are initially produced by a pulsed light field and allowed to spontaneously decay.

Squeezing in the resonance scattering case for a two-level system was predicted by Walls and Zoller [10]. Resonance scattering squeezing for the cases of a pair of two-level atoms also occurs [11(a),11(b)] as it does for the case of many two-level atoms [12]. Three-level Λ systems in a somewhat unsymmetrical configuration produce steady state squeezing [13(a)] in resonance fluorescence, though only transient squeezing in a symmetrical configuration with a nonoptimized choice of quadrature components [13(b)]. With recent developments in the study of resonance fluorescence from single laser-cooled trapped ions, it may be possible to study the squeezing from such simple sources using heterodyne detection of the fluorescence [14(a),14(b)].

Pertinent to applications of squeezed light is the amount of squeezing that can be achieved. For perfect squeezing in a quadrature component the Heisenberg uncertainty principle demands that the variance in the other quadrature component becomes infinite. Collett and Walls [15] show that for critical point conditions in several nonlinear optical processes (optical bistability, parametric oscillation, and second harmonic generation) perfect squeezing is theoretically possible. Quantum electrodynamic bounds on squeezing have also been determined [16]. Squeezing at the 70% level (-5.2 dB) has been achieved [17] experimentally in purely optical systems.

For the simpler systems of interest here involving the irreversible behavior of two-level and three-level atoms our aim will be to find out the maximum squeezing that could be achieved in the total electric field for *all* times and for all choices of the quadrature frequency and phase and to determine the nature of the atomic state associated with maximum squeezing. In particular, is the atomic state *pure* or *mixed*? Also does the source field correspond to a minimum uncertainty state? Naturally for any *specific* process the amount of squeezing and its time dependence will depend on the detailed dynamics involved in the process and an optical Bloch equation treatment would be needed to find the best squeezing in a specific process. For example, light emitted in resonance fluorescence from a two-level atom [10] can produce steady state squeezing which is largest for exact resonance when the Rabi frequency is equal to the upper level spontaneous decay rate divided by $\sqrt{6}$, and in this situation the atom is *not* in a pure state. However, the squeezing so obtained is not the largest it can be for light emitted from a two-level atom [11(b)]. Barnett and Knight [18] show that the maximum squeezing actually occurs for a pure atomic state whose Bloch vector is at 120° from the orientation of the pure excited state. No such analysis has yet been carried out for three-level atoms and this paper therefore deals with such three-level

cases.

As will be emphasized in this paper the determination of the optimum squeezing does *not* involve a consideration of detailed dynamics, but can be obtained from a consideration of the atomic density matrix elements that determine the variance of the quadrature components at the retarded time for this irreversible case. As we will see there are constraints on the matrix elements of the atomic density operator $\hat{\rho}$ that follow from consideration of the Hermiticity, unit trace, and positiveness of $\hat{\rho}$ (details are given in Appendix B). Again we emphasize that the optimum squeezing found is the best that could ever occur in *any* choice of process, quadrature frequency and phase, and indeed observation time. Such an amount of squeezing may not be achieved in any *particular* process and if it did it could correspond to a negative oscillation of a time-dependent variance. Having obtained the optimum squeezing it would then be of interest to examine *specific* processes (such as resonance fluorescence from driven three-level systems) to see if this optimum can be reached, especially at steady state and/or with time averaged observation conditions.

In this paper we study the optimum squeezing obtained for three-level systems in the Λ, V, and ladder configurations. We will relate the squeezing in the total electric field as specified in terms of the normally ordered variance of the quadrature components to the equivalent variance for the source field and hence to atomic populations, atomic one-photon coherences, and atomic two-photon coherences for the three-level atom. The choice of quadrature phase, quadrature frequency, and observation time here will be optimized to minimize the source field normally ordered variance. We then minimize further with respect to the choice of atomic density matrix elements subject to the aforementioned constraints. For the Λ and V configurations we first show that a change of atomic basis establishes the equivalence of the three-level system to a two-level system plus an uninvolved third level. The optimum squeezing can then be obtained analytically using the method of Lagrange undetermined multipliers. The ladder configuration cannot be reduced to a two-level case (or indeed a pair of two-level cases) and the optimum squeezing is obtained via numerical methods. For all three systems we find that the source field is in a minimum uncertainty state.

The concept of squeezing in the total electric field and its criterion in terms of the normally ordered variance of the quadrature component is briefly reviewed in Sec. II. The relationship of squeezing in the total field to squeezing in the source field does not seem to have been examined in detail elsewhere, unlike for the case of the spectrum where it has been shown [19,20] that the two-time correlation function for the total electric field at an asymptotically placed detector is given by the two-time correlation function of the source field, provided the initial (driving) field involves classical field amplitudes that are zero at the detector. Here the normalized variance of the total electric field quadrature component is first shown to be the sum of equivalent variances for the free field and for the source field together with interference terms between the source field and the free field, which depend on the quadrature phase. For the case where the total field involves classical field amplitudes that are zero at an asymptotically placed detector, we show that the normally ordered variance of the free-field quadrature component together with most of the interference terms can be set to zero. The elimination of the final two (phase-dependent) interference terms requires a consideration of certain free-field–source-field commutators (details are in Appendix A) similar to those considered in other contexts [20–22].

In general the commutators are nonzero, but for the equal time situation of interest they can be shown to be zero for the case where the source atoms are located in a region whose dimension is small compared to the resonant wavelength (Dicke atom source). The single-atom system under study here is such a source. For such Dicke atom sources the final interference terms are then zero and the major result that the normally ordered variance for the total field equals the equivalent variance for the source field is demonstrated. Thus optimizing the source field squeezing also optimizes the total field squeezing. Expressions used for the study of the source field for the three-level atom case are obtained. A short review of source-field squeezing and criteria for a minimum uncertainty state in the source field is also given in Sec. II, along with the expressions for the optimum value of the normally ordered variance of the source-field quadrature component for all choices of quadrature phase, frequency, and wave vector.

Section III deals with the specific three-level atom source in Λ, V, and ladder configurations. Following the description of one-photon and two-photon coherences in terms of the original and modified atomic basis states, five distinct types of squeezing for three-level sources are distinguished, including the important cases of two-level and three-level squeezing. Specific expressions in the normally ordered and nonordered variances of the source-field quadrature components are given and the results of optimizing the choice of atomic state for all three configurations are presented. The details are given in Appendix C. Results are summarized in Sec. IV.

II. THEORY OF SQUEEZING FOR MULTIATOM SOURCES

A. General squeezing expressions

The electric field operator $\hat{E}$ for the quantum em field can be written in terms of quadrature components $\hat{E}_\phi$ and $\hat{E}_{\phi-\frac{\pi}{2}}$ as [1]

$$\hat{E}(\mathbf{R},t) = \hat{E}_\phi \cos(\omega t - \mathbf{k}\cdot\mathbf{R} + \phi) + \hat{E}_{\phi-\frac{\pi}{2}} \sin(\omega t - \mathbf{k}\cdot\mathbf{R} + \phi). \quad (2.1)$$

The quadrature component of the electric field $\hat{\mathrm{E}}_\phi$ at frequency ω, wave vector $\mathbf{k}$, and phase ϕ is given in terms of the positive, negative components $\hat{\mathrm{E}}^{\pm}$ as [1]

$$\hat{E}_\phi = \hat{E}^+ e^{i\xi} + \hat{E}^- e^{-i\xi}, \quad (2.2)$$

where

$$\xi = \omega t - \mathbf{k}\cdot\mathbf{R} + \phi. \tag{2.3}$$

For convenience we will generally interpret $\hat{E}^{\pm}$, $\hat{E}_{\phi}$, etc. as specific vectorial components along unit vector $\mathbf{e}$, unless the vectorial nature of the quantity is specifically indicated. The operators $\hat{E}^{\pm}$, etc. are Heisenberg field operators at position $\mathbf{R}$ at time t.

The positive and negative frequency components can be written in terms of the modes λ for the quantum em field as

$$\hat{\mathbf{E}}^{+} = \sum_{\lambda} \mathbf{f}_{\lambda}(\mathbf{R})\,\hat{a}_{\lambda} \tag{2.4}$$

$$= \left(\hat{\mathbf{E}}^{-}\right)^{\dagger}, \tag{2.5}$$

where

$$\mathbf{f}_{\lambda}(\mathbf{R}) = i\left(\frac{\hbar\omega_{\lambda}}{2\epsilon_{o}V}\right)^{\frac{1}{2}} e^{i\mathbf{k}_{\lambda}\cdot\mathbf{R}}\,\mathbf{e}_{\lambda}, \tag{2.6}$$

and where mode λ has wave vector $\mathbf{k}_{\lambda}$, frequency ω_{λ}, and polarization vector $\mathbf{e}_{\lambda}$. The annihilation and creation operators $\hat{a}_{\lambda}$ and $\hat{a}^{\dagger}_{\lambda}$ obey the usual Bose commutation rules

$$\left[\hat{a}_{\lambda}, \hat{a}^{\dagger}_{\mu}\right] = \delta_{\lambda\mu}. \tag{2.7}$$

It is well known that simultaneous precise measurements of the two quadrature components cannot occur as they satisfy the commutation rule

$$\left[\hat{E}_{\phi}, \hat{E}_{\phi-\frac{\pi}{2}}\right] = 2i\left[\hat{E}^{+}, \hat{E}^{-}\right] \tag{2.8}$$

and a Heisenberg uncertainty principle result for the variances $\left\langle \Delta\hat{E}^{2}_{\phi}\right\rangle, \left\langle \Delta\hat{E}^{2}_{\phi-\frac{\pi}{2}}\right\rangle$ is satisfied

$$\left\langle \Delta\hat{E}^{2}_{\phi}\right\rangle \left\langle \Delta\hat{E}^{2}_{\phi-\frac{\pi}{2}}\right\rangle \geq \left|\left\langle\left[\hat{E}^{+}, \hat{E}^{-}\right]\right\rangle\right|^{2}. \tag{2.9}$$

The case where equality occurs in (2.9) is that of a so-called *minimum uncertainty state* (MUS) for the total electric field $\hat{E}$. Here the variances $\left\langle \Delta\hat{\Omega}^{2}\right\rangle$ and the mean $\left\langle\hat{\Omega}\right\rangle$ are defined as

$$\left\langle \Delta\hat{\Omega}^{2}\right\rangle = \mathrm{Tr}\left[\left(\hat{\Omega} - \left\langle\hat{\Omega}\right\rangle\right)^{2}\hat{W}(0)\right], \tag{2.10}$$

$$\left\langle\hat{\Omega}\right\rangle = \mathrm{Tr}\left[\hat{\Omega}\hat{W}(0)\right], \tag{2.11}$$

where $\hat{W}(0)$ is the initial density operator for the quantum em field and its sources and the trace is taken over their quantum states.

The quadrature component $\hat{E}_{\phi}$ of the total em field is said to be *squeezed* relative to the other quadrature component $\hat{E}_{\phi-\frac{\pi}{2}}$ if the variance $\left\langle \Delta\hat{E}^{2}_{\phi}\right\rangle$ is smaller than that for the MUS, that is,

$$\left\langle \Delta\hat{E}^{2}_{\phi}\right\rangle < \left|\left\langle\left[\hat{E}^{+},\ \hat{E}^{-}\right]\right\rangle\right|. \tag{2.12}$$

The normally ordered variance of the quadrature component $\hat{E}_{\phi}$ is defined by

$$\begin{aligned}\left\langle :\Delta\hat{E}^{2}_{\phi}:\right\rangle = &\left(\left\langle\hat{E}^{+2}\right\rangle - \left\langle\hat{E}^{+}\right\rangle^{2}\right)e^{2i\xi}\\ &+\left(\left\langle\hat{E}^{-2}\right\rangle - \left\langle\hat{E}^{-}\right\rangle^{2}\right)e^{-2i\xi}\\ &+2\left(\left\langle\hat{E}^{-}\hat{E}^{+}\right\rangle - \left\langle\hat{E}^{-}\right\rangle\left\langle\hat{E}^{+}\right\rangle\right)\end{aligned} \tag{2.13}$$

and is related to the variance $\left\langle \Delta\hat{E}^{2}_{\phi}\right\rangle$ via

$$\left\langle \Delta\hat{E}^{2}_{\phi}\right\rangle = \left\langle :\Delta\hat{E}^{2}_{\phi}:\right\rangle + \left\langle\left[\hat{E}^{+}, \hat{E}^{-}\right]\right\rangle, \tag{2.14}$$

since it is constructed from the variance by putting all creation operator terms such as $\hat{E}^{-}$ to the left of all annihilation operator terms such as $\hat{E}^{+}$.

Since the commutator $\left[\hat{E}^{+}, \hat{E}^{-}\right]$ for the total field operators $\hat{E}^{+}$,$\hat{E}^{-}$ is a positive c number

$$\left[\hat{E}^{+}, \hat{E}^{-}\right] = \sum_{\lambda} f^{\mathbf{e}}_{\lambda}\, f^{\mathbf{e}*}_{\lambda} \geq 0, \tag{2.15}$$

with $f^{\mathbf{e}}_{\lambda} = \mathbf{e}\cdot\mathbf{f}_{\lambda}(\mathbf{R})$, it follows that

$$\left\langle\left[\hat{E}^{+}, \hat{E}^{-}\right]\right\rangle = \left|\left\langle\left[\hat{E}^{+}, \hat{E}^{-}\right]\right\rangle\right| = \sum_{\lambda} f^{\mathbf{e}}_{\lambda}\, f^{\mathbf{e}*}_{\lambda}. \tag{2.16}$$

Hence the *criterion for squeezing* is that the *normally ordered variance* must be *negative*

$$\left\langle :\Delta\hat{E}^{2}_{\phi}:\right\rangle < 0. \tag{2.17}$$

The multimode coherent state $|\alpha\rangle = |\alpha_1, \alpha_2, \ldots, \alpha_{\lambda}, \ldots\rangle$ is an eigenstate for $\hat{\mathbf{E}}^{+}$ with associated classical field $\mathcal{E}(\mathbf{R})$

$$\hat{\mathbf{E}}^{+}|\alpha\rangle = \mathcal{E}(\mathbf{R})\,|\alpha\rangle, \tag{2.18}$$

with

$$\mathcal{E}(\mathbf{R}) = \sum_{\lambda} \alpha_{\lambda}\,\mathbf{f}_{\lambda}(\mathbf{R}). \tag{2.19}$$

For such a multimode coherent state with $\hat{W}(0) = |\alpha\rangle\langle\alpha|$ it is easy to show that the normally ordered variance in any quadrature component $\left\langle \Delta\hat{E}^{2}_{\phi}\right\rangle, \left\langle :\Delta\hat{E}^{2}_{\phi}:\right\rangle$ is zero, so that

$$\begin{aligned}\left\langle \Delta\hat{E}^{2}_{\phi}\right\rangle_{\text{coherent}} &= \left\langle\left[\hat{E}^{+}, \hat{E}^{-}\right]\right\rangle\\ &= \sum_{\lambda} f^{\mathbf{e}}_{\lambda}\, f^{\mathbf{e}*}_{\lambda} \geq 0.\end{aligned} \tag{2.20}$$

Thus a squeezed state has a variance less than that for any coherent state, including the vacuum state

$$\left\langle \Delta E^{2}_{\phi}\right\rangle < \left\langle \Delta\hat{E}^{2}_{\phi}\right\rangle_{\text{coherent}}, \tag{2.21}$$

which is another *criterion for a squeezed state.*

At this point we introduce the concept of *optimum squeezing*. This means that the variance $\left\langle \Delta E_\phi^2 \right\rangle$ of *one* quadrature component $\hat{E}_\phi$ is to be as small as it can be taking into account *all* possible choices of the quadrature components $(\phi, \omega, \mathbf{k})$ and *all* possible choices of the state $\hat{W}(0)$ for the field and its sources. Since the variance $\left\langle \Delta \hat{E}_\phi^2 \right\rangle$ satisfies (2.14), with $\left\langle \left[\hat{E}^+, \hat{E}^-\right] \right\rangle$ given by the positive result (2.16), it follows that the optimum squeezing requires us to find the situation where $\left\langle : \Delta \hat{E}_\phi^2 : \right\rangle$ is as *negative as possible*. In this case the variance $\left\langle \Delta \hat{E}_\phi^2 \right\rangle$ will be smaller than that for coherent state by the largest possible amount. This situation is referred to as that of *maximum* squeezing also.

The previous discussion applies to the general case of the quantum em field irrespective of any particular state the field and its sources might be in and for all types of sources of the field. Next we consider the specific case of interest, where the field sources are atomic systems.

B. Squeezing for the field with multiatom sources

The combined system of the quantum em field and the atomic systems may be described via the multipolar Hamiltonian [23] and the interaction between field and atoms given to a good approximation by the electric dipole term. The *total electric field* operator $\hat{\mathbf{E}}$ can then be expressed without further approximation as the sum of a *free-field* term $\hat{\mathbf{E}}_F$ and a *source-field* term $\hat{\mathbf{E}}_S$ [9,20,22]

$$\hat{\mathbf{E}} = \hat{\mathbf{E}}_F + \hat{\mathbf{E}}_S, \tag{2.22}$$

where

$$\begin{aligned}\hat{\mathbf{E}}_F &= \sum_\lambda \mathbf{f}_\lambda(\mathbf{R})\,\hat{a}_\lambda(0)\,e^{-i\omega_\lambda t} + \text{H.c.} \\ &= \sum_\lambda i\left(\frac{\hbar\omega_\lambda}{2\epsilon_0 V}\right)^{\frac{1}{2}} \mathbf{e}_\lambda e^{i(\mathbf{k}_\lambda\cdot\mathbf{R}-\omega_\lambda t)}\hat{a}_\lambda(0) + \text{H.c.}\end{aligned} \tag{2.23}$$

and

$$\begin{aligned}\hat{\mathbf{E}}_S = \boldsymbol{\nabla}\times\Bigg[\boldsymbol{\nabla}\times\frac{1}{4\pi\epsilon_0}\sum_A \frac{\hat{\mathbf{d}}_A\left(t-\frac{|\mathbf{R}-\mathbf{R}_A|}{c}\right)}{|\mathbf{R}-\mathbf{R}_A|} \\ \times\theta\left(t-\frac{|\mathbf{R}-\mathbf{R}_A|}{c}\right)\Bigg].\end{aligned} \tag{2.24}$$

For the source field the sum in (2.24) is over different atoms A whose position is at $\mathbf{R}_A$ and whose electric dipole operator $\hat{\mathbf{d}}_A \cdot \theta$ is the usual Heaviside function, zero for negative argument and unity for positive argument. No arbitrary assumptions on boundary conditions are actually required to produce the above *retarded* form for the source field, where the field at $\mathbf{R},t$ depends on the dipole operator $\hat{\mathbf{d}}_A$ at the retarded time $t-|\mathbf{R}-\mathbf{R}_A|/c$. The total electric field $\hat{\mathbf{E}}$ is the new electric field produced via the Power-Zienau transformation [23] (equal to the old electric displacement) and, as it is the operator involved in any subsequent theory of photodetection of the field, it is this new electric field and its squeezing properties that we will continue to focus on. The free field has the same time dependence that would apply if there were no atomic systems present.

The above expressions are valid for all $\mathbf{R},t$. In the usual case where we detect the field at a large distance from the source atoms, the asymptotic form of $\hat{\mathbf{E}}_S$ is important and for $t > |\mathbf{R}-\mathbf{R}_A|/c$ with R large, we have

$$\begin{aligned}\hat{\mathbf{E}}_S = \frac{1}{4\pi\epsilon_0 c^2}\sum_A\left(\hat{\mathbf{R}}-\hat{\mathbf{R}}_A\right) \\ \times\left(\frac{\left(\hat{\mathbf{R}}-\hat{\mathbf{R}}_A\right)\times\ddot{\hat{\mathbf{d}}}_A\left(t-\frac{|\mathbf{R}-\mathbf{R}_A|}{c}\right)}{|\mathbf{R}-\mathbf{R}_A|}\right),\end{aligned} \tag{2.25}$$

showing the inverse distance dependence at large $R=|\mathbf{R}|$ and the dependence on the dipole acceleration.

The electric dipole operator for the A atom can be written as the sum of an upward component $\hat{\mathbf{d}}_A^+$ and a downward component $\hat{\mathbf{d}}_A^-$ as

$$\hat{\mathbf{d}}_A = \hat{\mathbf{d}}_A^+ + \hat{\mathbf{d}}_A^-, \tag{2.26}$$

where with atomic states $|i\rangle,|j\rangle$ whose energies are $\hbar\omega_i$, $\hbar\omega_j$, etc. and i >j signifies $\omega_i > \omega_j$

$$\hat{\mathbf{d}}_A^+ = \sum_{i>j}\hat{\Lambda}_{ij}^A\mathbf{d}_{ij} = \left(\hat{\mathbf{d}}_A^-\right)^\dagger. \tag{2.27}$$

The $\hat{\Lambda}_{ij}^A = |i\rangle\langle j|$ are atomic transition operators and $\mathbf{d}_{ij} = \langle i|\hat{\mathbf{d}}_A|j\rangle$ are dipole elements. Approximating $\hat{\mathbf{d}}_A$ by its free evolution expression gives the familiar form for $\hat{\mathbf{E}}_S$ at large distances

$$\begin{aligned}\hat{\mathbf{E}}_S = \frac{-1}{4\pi\epsilon_0 c^2}\sum_{ijA}\left(\hat{\mathbf{R}}-\hat{\mathbf{R}}_A\right) \\ \times\left(\frac{\left(\hat{\mathbf{R}}-\hat{\mathbf{R}}_A\right)\times\hat{\Lambda}_{ij}^A\left(t-\frac{|\mathbf{R}-\mathbf{R}_A|}{c}\right)\omega_{ij}^2\,\mathbf{d}_{ij}}{|\mathbf{R}-\mathbf{R}_A|}\right).\end{aligned} \tag{2.28}$$

The positive and negative frequency components of $\hat{\mathbf{E}}$ are defined by (2.4) and (2.5). Solving the Heisenberg equations of motion for $\hat{a}_\lambda,\hat{a}_\lambda^\dagger$ along with the use of Laplace transform methods and standard mode summation methods that enabled the result (2.22) to be obtained also enables expressions for $\hat{\mathbf{E}}_F^\pm,\hat{\mathbf{E}}_S^\pm$ to be derived.

For the free-field terms

$$\hat{\mathbf{E}}_F^+ = \sum_\lambda \mathbf{f}_\lambda(\mathbf{R})\hat{a}_\lambda(0)\,e^{-i\omega_\lambda t} \tag{2.29}$$

$$= \left(\hat{\mathbf{E}}_F^-\right)^\dagger. \tag{2.30}$$

For the source-field terms we find that, provided the typical atomic transition frequencies $\omega_{ij} \sim \omega_0$ are large com-

pared to the decay rates, to a good approximation

$$\hat{\mathbf{E}}_S^+ = \nabla \times \left[\nabla \times \frac{1}{4\pi\epsilon_0} \sum_A \frac{\hat{\mathbf{d}}_A^- \left(t - \frac{|\mathbf{R}-\mathbf{R}_A|}{c} \right)}{|\mathbf{R}-\mathbf{R}_A|} \times \theta \left(t - \frac{|\mathbf{R}-\mathbf{R}_A|}{c} \right) \right], \tag{2.31}$$

$$\hat{\mathbf{E}}_S^- = \nabla \times \left[\nabla \times \frac{1}{4\pi\epsilon_0} \sum_A \frac{\hat{\mathbf{d}}_A^+ \left(t - \frac{|\mathbf{R}-\mathbf{R}_A|}{c} \right)}{|\mathbf{R}-\mathbf{R}_A|} \times \theta \left(t - \frac{|\mathbf{R}-\mathbf{R}_A|}{c} \right) \right]. \tag{2.32}$$

Similar asymptotic expressions for $\hat{\mathbf{E}}_S^+, \hat{\mathbf{E}}_S^-$ can be obtained

$$\hat{\mathbf{E}}_S^+ = -\frac{1}{4\pi\epsilon_0 c^2} \sum_{i>jA} \left(\hat{\mathbf{R}} - \hat{\mathbf{R}}_A \right) \times \left(\frac{\left(\hat{\mathbf{R}} - \hat{\mathbf{R}}_A \right) \times \hat{\Lambda}_{ji}^A \left(t - \frac{|\mathbf{R}-\mathbf{R}_A|}{c} \right) \mathbf{d}_{ji} \omega_{ji}^2}{|\mathbf{R}-\mathbf{R}_A|} \right), \tag{2.33}$$

$$\hat{\mathbf{E}}_S^- = -\frac{1}{4\pi\epsilon_0 c^2} \sum_{i>j} \left(\hat{\mathbf{R}} - \hat{\mathbf{R}}_A \right) \times \left(\frac{\left(\hat{\mathbf{R}} - \hat{\mathbf{R}}_A \right) \times \hat{\Lambda}_{ij}^A \left(t - \frac{|\mathbf{R}-\mathbf{R}_A|}{c} \right) \mathbf{d}_{ij} \omega_{ij}^2}{|\mathbf{R}-\mathbf{R}_A|} \right). \tag{2.34}$$

The component along a unit vector $\mathbf{e}$ which is perpendicular to $\mathbf{R} - \mathbf{R}_A$ is easily obtained as [9]

$$\hat{E}_S^+ = \mathbf{e} \cdot \hat{\mathbf{E}}_S^+ = \frac{1}{4\pi\epsilon_0 c^2} \sum_{i>jA} \frac{\omega_{ji}^2 \mathbf{e} \cdot \mathbf{d}_{ji} \hat{\Lambda}_{ji}^A \left(t - \frac{|\mathbf{R}-\mathbf{R}_A|}{c} \right)}{|\mathbf{R}-\mathbf{R}_A|}, \tag{2.35}$$

$$\hat{E}_S^- = \mathbf{e} \cdot \hat{\mathbf{E}}_S^- = \frac{1}{4\pi\epsilon_0 c^2} \sum_{i>jA} \frac{\omega_{ij}^2 \mathbf{e} \cdot \mathbf{d}_{ij} \hat{\Lambda}_{ij}^A \left(t - \frac{|\mathbf{R}-\mathbf{R}_A|}{c} \right)}{|\mathbf{R}-\mathbf{R}_A|}. \tag{2.36}$$

These expressions are to be used in the treatment which follows of the fields produced by three-level atoms.

The squeezing in the total field can be considered in terms of substituting $E_F^+ + E_S^+$ for $E^\pm$ in the expression for the quadrature components, variances, etc., with $E_{\phi S}, E_{\phi F}$ given in (2.2) but with $E^\pm$ replaced by $E_S^\pm$ or $E_F^\pm$. The normally ordered variance of the total field is given as

$$\begin{aligned} \left\langle : \Delta \hat{E}_\phi^2 : \right\rangle = & \left\langle : \Delta \hat{E}_{\phi F}^2 : \right\rangle + \left\langle : \Delta \hat{E}_{\phi S}^2 : \right\rangle + \left(\left\langle \hat{E}_F^+ \hat{E}_S^+ \right\rangle + \left\langle \hat{E}_S^+ \hat{E}_F^+ \right\rangle - 2 \left\langle \hat{E}_F^+ \right\rangle \left\langle \hat{E}_S^+ \right\rangle \right) e^{2i\xi} \\ & + \left(\left\langle \hat{E}_F^- \hat{E}_S^- \right\rangle + \left\langle \hat{E}_S^- \hat{E}_F^- \right\rangle - 2 \left\langle \hat{E}_F^- \right\rangle \left\langle \hat{E}_S^- \right\rangle \right) e^{-2i\xi} \\ & + 2 \left(\left\langle \hat{E}_F^- \hat{E}_S^+ \right\rangle + \left\langle \hat{E}_S^- \hat{E}_F^+ \right\rangle - \left\langle \hat{E}_F^- \right\rangle \left\langle \hat{E}_S^+ \right\rangle - \left\langle \hat{E}_F^+ \right\rangle \left\langle \hat{E}_S^- \right\rangle \right). \end{aligned} \tag{2.37}$$

Thus the normally ordered variance equals the sum of the normally ordered variances for the free field and for the source field together with possible interference terms involving both the free field and the source field. These interference terms are phase dependent. The following question then arises: under what conditions can the normally ordered variance for the total field be equated to the normally ordered variance for the source-field atoms? A similar situation occurs for the spectrum of the em field, where the usual practice is to replace the total electric field by the source field in the two-time correlation functions that determine the spectrum [19,20]. In practice the atomic systems are driven by fields which would have classical electric fields that are zero at the position $\mathbf{R}$ of any photodetectors and ultimately this enables us to eliminate most of the interference and the free field terms here, as it does for the spectrum. A further condition such as Dicke atom sources is required, however, to eliminate the remainder of the interference terms.

The density operator $\hat{W}(0)$ can be written as

$$\hat{W}(0) = \hat{\rho}_A(0) \; \hat{\rho}_F(0), \tag{2.38}$$

where $\hat{\rho}_A(0)$ is the initial density operator for the atoms and $\hat{\rho}_F(0)$ for the field. The initial field density operator can be expressed in terms of the Glauber-Sudarshan P representation as

$$\hat{\rho}_F(0) = \int d^2\alpha \; P(\alpha, \alpha^*) |\alpha\rangle \langle\alpha|. \tag{2.39}$$

Then

$$\hat{E}_F^+ \hat{\rho}_F(0) = \int d^2\alpha \, P(\alpha, \alpha^*) \, \mathcal{E}_F(\mathbf{R},t) |\alpha\rangle \langle\alpha|,$$

$$\hat{\rho}_F(0) \hat{E}_F^- = \int d^2\alpha \; P(\alpha, \alpha^*) |\alpha\rangle \langle\alpha| \, \mathcal{E}_F^*(\mathbf{R},t),$$

where

$$\mathcal{E}_F(\mathbf{R},t) = \sum_\lambda \alpha_\lambda e^{-i\omega_\lambda t} \mathbf{e} \cdot \mathbf{f}_\lambda(\mathbf{R}). \tag{2.40}$$

The situation of the driving fields being zero at the detector is expressed as follows: for important α in phase space [where $P(\alpha, \alpha^*)$ is significant] the corresponding classical fields $\mathcal{E}_F(\mathbf{R},t)$ are zero at the detector.

This condition enables *most* of the terms in (2.37) to be set to zero. First, the normally ordered variance for the free field is zero. For some typical terms

$$\left\langle \hat{E}_F^+ \right\rangle = \mathrm{Tr}\ \hat{E}_F^+\ \hat{\rho}_A(0)\ \hat{\rho}_F(0)$$
$$= \int d^2\alpha\ \ P(\alpha,\alpha^*)\,\mathrm{Tr}\left[\hat{E}_F^+ |\alpha\rangle\langle\alpha|\ \hat{\rho}_A(0)\right] = \int d^2\alpha\ \ P(\alpha,\alpha^*)\,\mathcal{E}_F(\mathbf{R},t)\,\mathrm{Tr}\left[|\alpha\rangle\langle\alpha|\ \hat{\rho}_A(0)\right] = 0, \tag{2.41}$$

$$\left\langle \hat{E}_F^{+2} \right\rangle = \int d^2\alpha\ \ P(\alpha,\alpha^*)\,\mathcal{E}_F^2(\mathbf{R},t)\,\mathrm{Tr}\left[|\alpha\rangle\langle\alpha|\ \hat{\rho}_A(0)\right] = 0, \tag{2.42}$$

$$\left\langle \hat{E}_F^- \hat{E}_F^+ \right\rangle = \mathrm{Tr}\left[\hat{E}_F^-\ \hat{E}_F^+\ \hat{\rho}_F(0)\ \hat{\rho}_A(0)\right]$$
$$= \mathrm{Tr}\left[\hat{E}_F^+\ \hat{\rho}_F(0)\ \hat{E}_F^-\ \hat{\rho}_A(0)\right]$$
$$= \int d^2\alpha\ \ P(\alpha,\alpha^*)\,\mathrm{Tr}\left[\hat{E}_F^+ |\alpha\rangle\langle\alpha|\ \hat{E}_F^-\ \hat{\rho}_A(0)\right]$$
$$= \int d^2\alpha\ \ P(\alpha,\alpha^*)\,\mathcal{E}_F(\mathbf{R},t)\,\mathcal{E}_F^*(\mathbf{R},t)\,\mathrm{Tr}\left[|\alpha\rangle\langle\alpha|\ \hat{\rho}_A(0)\right] = 0. \tag{2.43}$$

Hence

$$\left\langle :\Delta\hat{E}_{\phi F}^2: \right\rangle = 0. \tag{2.44}$$

Second, many of the interference terms are zero. For some typical terms

$$\left\langle \hat{E}_S^+\ \hat{E}_F^+ \right\rangle = \mathrm{Tr}\left[\hat{E}_S^+\ \hat{E}_F^+\ \hat{\rho}_F(0)\ \hat{\rho}_A(0)\right]$$
$$= \int d^2\alpha\ \ P(\alpha,\alpha^*)\,\mathrm{Tr}\left[\hat{E}_S^+\ \hat{E}_F^+ |\alpha\rangle\langle\alpha|\ \hat{\rho}_A(0)\right]$$
$$= \int d^2\alpha\ \ P(\alpha,\alpha^*)\,\mathcal{E}_F(\mathbf{R},t)\,\mathrm{Tr}\left[\hat{E}_S^+\ |\alpha\rangle\langle\alpha|\ \hat{\rho}_A(0)\right] = 0, \tag{2.45}$$

$$\left\langle \hat{E}_F^-\ \hat{E}_S^- \right\rangle = \mathrm{Tr}\left[\hat{E}_F^-\ \hat{E}_S^-\ \hat{\rho}_F(0)\ \hat{\rho}_A(0)\right]$$
$$= \mathrm{Tr}\left[\hat{E}_S^-\ \hat{\rho}_F(0)\ \hat{E}_F^-\ \hat{\rho}_A(0)\right]$$
$$= \int d^2\alpha\ \ P(\alpha,\alpha^*)\,\mathrm{Tr}\left[\hat{E}_S^- |\alpha\rangle\langle\alpha|\ \hat{E}_F^-\ \hat{\rho}_A(0)\right]$$
$$= \int d^2\alpha\ \ P(\alpha,\alpha^*)\,\mathcal{E}_F^*(\mathbf{R},t)\,\mathrm{Tr}\left[\hat{E}_S^- |\alpha\rangle\langle\alpha|\ \hat{\rho}_A(0)\right] = 0. \tag{2.46}$$

Hence

$$\left\langle \hat{E}_S^+\hat{E}_F^+ \right\rangle = \left\langle \hat{E}_F^-\hat{E}_S^- \right\rangle = \left\langle \hat{E}_F^-\hat{E}_S^+ \right\rangle = \left\langle \hat{E}_S^-\hat{E}_F^+ \right\rangle = 0. \tag{2.47}$$

For the normally ordered variance we now have

$$\left\langle :\Delta\hat{E}_\phi^2: \right\rangle = \left\langle :\Delta\hat{E}_{\phi S}^2: \right\rangle + \left\langle \hat{E}_F^+\hat{E}_S^+ \right\rangle e^{2i\xi} + \left\langle \hat{E}_S^-\hat{E}_F^- \right\rangle e^{-2i\xi}. \tag{2.48}$$

To proceed further we need to consider the commutator

$$\hat{C} = \left[\hat{\mathbf{E}}_F^+(\mathbf{R}_1,t_1), \hat{\mathbf{E}}_S^+(\mathbf{R}_2,t_2)\right] \tag{2.49}$$

$$= \left[\hat{\mathbf{E}}_S^-(\mathbf{R}_2,t_2), \hat{\mathbf{E}}_F^-(\mathbf{R}_2,t_2)\right]^\dagger, \tag{2.50}$$

where we will take $\mathbf{R}_1 = \mathbf{R}_2 = \mathbf{R}$ (asymptotic) and $t_1 = t_2 = t$, where $t > |\mathbf{R} - \mathbf{R}_A|/c$ for all atoms present. Under the same approximation as before that the atomic transition frequencies are large compared to decay rates we find (see Appendix A)

$$\hat{C} = -\sum_{AB} \nabla_2 \times \left\{ \nabla_2 \times \frac{1}{4\pi\epsilon_0 R_{A2}} \times \left[\nabla_1 \times \left(\nabla_1 \times \frac{1}{4\pi\epsilon_0 R_{B1}} \hat{K} \right) \right] \right\}, \tag{2.51}$$

where

$$\hat{K} = \left[\hat{\mathbf{d}}_B^-(t_1-\tau_{B1}), \hat{\mathbf{d}}_A^-(t_2-\tau_{A2})\right] \theta(t_1-\tau_{B1}) \times\theta(t_2-\tau_{A2})\theta((t_2-\tau_{A2})-(t_1-\tau_{B1})) - \left[\hat{\mathbf{d}}_B^-(t_1+\tau_{B1}), \hat{\mathbf{d}}_A^-(t_2-\tau_{A2})\right] \theta(t_1+\tau_{B1}) \times\theta(t_2-\tau_{A2})\theta((t_2-\tau_{A2})-(t_1+\tau_{B1})) \tag{2.52}$$

and the time delays and distances are defined by

$$R_{A2} = |\mathbf{R}_2 - \mathbf{R}_A| = c\tau_{A2}, \tag{2.53}$$

$$R_{B1} = |\mathbf{R}_1 - \mathbf{R}_B| = c\tau_{B1}. \tag{2.54}$$

A similar result has been obtained by Cresser [20] for the case of a single atom situated at the origin.

For our purposes the case of interest is where $t_1 = t_2 = t$ and where $t_1 > \tau_{B1}, t_2 > \tau_{A2}$ for all atoms and where we will take $\mathbf{R}_1 = \mathbf{R}_2 = \mathbf{R}$. For this case $\hat{K}$ is given by

$$\hat{K} = \left[\hat{\mathbf{d}}_B^-(t-\tau_{B1}), \hat{\mathbf{d}}_A^-(t-\tau_{A2})\right]\theta(\tau_{B1}-\tau_{A2}), \tag{2.55}$$

the second terms being zero due to the Heaviside functions $\theta(-\tau_{A2}-\tau_{B1})$ and two of the Heaviside functions in the first term being equal to unity.

For many of the situations of interest all the atoms will be confined in a region close to the origin and small compared to the transition wavelength $\lambda_0 = 2\pi c/\omega_0$. Thus the conditions of the Dicke model will apply and we restrict ourselves to the case

$$|\mathbf{R}_{A2} - \mathbf{R}_{B1}| \ll \lambda_0. \tag{2.56}$$

As $\hat{\mathbf{d}}_B^-(t), \hat{\mathbf{d}}_A^-(t)$ have typical oscillation frequencies ω_0, the difference in phase $\omega_0(\tau_{A2}-\tau_{B1})$ between the two factors in $\hat{K}$ is small compared to 2π. Hence with a common delay $\tau = \tau_{A2} = \tau_{B1}$

$$\hat{K} = \left[\hat{\mathbf{d}}_B^-(t-\tau), \hat{\mathbf{d}}_A^-(t-\tau)\right]\theta(\tau_{B1}-\tau_{A2}). \tag{2.57}$$

For the terms where the atoms are different ($A \neq B$) the commutator is zero, since $\hat{\mathbf{d}}_B^-(0)$ commutes with $\hat{\mathbf{d}}_A^-(0)$. For the terms where the atoms are the same ($A = B$), the dipole operators are the same and hence will commute. For a Dicke atom source we therefore find

$$\left[\hat{\mathbf{E}}_F^+(\mathbf{R},t), \hat{\mathbf{E}}_S^+(\mathbf{R},t)\right] = \left[\hat{\mathbf{E}}_S^-(\mathbf{R},t), \hat{\mathbf{E}}_F^-(\mathbf{R},t)\right] = 0, \tag{2.58}$$

so that

$$\left\langle \hat{E}_F^+ \hat{E}_S^+ \right\rangle = \left\langle \hat{E}_S^+ \hat{E}_F^+ \right\rangle = 0 \tag{2.59}$$

and

$$\left\langle \hat{E}_S^- \hat{E}_F^- \right\rangle = \left\langle \hat{E}_F^- \hat{E}_S^- \right\rangle = 0. \tag{2.60}$$

Thus the normally ordered variance in the total field is given by the normally ordered variance for the source field

$$\left\langle : \Delta\hat{E}_\phi^2 : \right\rangle = \left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle \tag{2.61}$$

and the variance for the total field is given by

$$\left\langle \Delta\hat{E}_\phi^2 \right\rangle = \left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle + \left\langle \Delta\hat{E}_\phi^2 \right\rangle_{\text{coherent state}}. \tag{2.62}$$

It is seen that squeezing in the total field requires $\left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle < 0$. Furthermore optimized squeezing for the total field occurs when the normally ordered variance for the source field is as negative as possible, and this will be the basis of our optimization approach. As it is the total field that is detected it is important to have established that the minimization of the normally ordered variance $\left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle$ for the *source* field will in fact optimize the squeezing in the *total* field.

With regard to a MUS in the total field we require

$$\left\langle \Delta\hat{E}_\phi^2 \right\rangle \left\langle \Delta\hat{E}_{\phi-\frac{\pi}{2}}^2 \right\rangle = \left|\langle [E^+, E^-] \rangle\right|^2 = \left(\left\langle \Delta\hat{E}_\phi^2 \right\rangle_{\text{coherent state}}\right)^2. \tag{2.63}$$

Substituting for $\left\langle \Delta\hat{E}_\phi^2 \right\rangle$ from (2.62) we see that for a MUS we require

$$\left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle \left\langle : \Delta\hat{E}_{\phi-\frac{\pi}{2}S}^2 : \right\rangle + \left(\left\langle \Delta\hat{E}_\phi^2 \right\rangle_{\text{coherent state}}\right) \times \left(\left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle + \left\langle : \Delta\hat{E}_{\phi-\frac{\pi}{2}S}^2 : \right\rangle\right) = 0. \tag{2.64}$$

Since the normally ordered source-field variances are proportional to $\left(\frac{1}{R}\right)^2$ and since $\left\langle \Delta\hat{E}_\phi^2 \right\rangle_{\text{coh}}$ is positive, it does not seem possible that the optimization process could produce normally ordered variances such that (2.63) is satisfied, the first terms being proportional to $\left(\frac{1}{R}\right)^4$ and the second to $\left(\frac{1}{R}\right)^2$. Hence optimization will not produce a MUS for the total field.

C. Squeezing for the source field

It is of some interest to consider the source field $\hat{\mathbf{E}}_S$ as a field in its own right. The commutator for the quadrature components $\hat{E}_{\phi S}, \hat{E}_{\phi-\frac{\pi}{2}S}$ satisfies

$$\left[\hat{E}_{\phi S}, \hat{E}_{\phi-\frac{\pi}{2}S}\right] = 2i\left[\hat{E}_S^+, \hat{E}_S^-\right], \tag{2.65}$$

so that the variances satisfy the Heisenberg uncertainty principle result

$$\left\langle \Delta\hat{E}_{\phi S}^2 \right\rangle \left\langle \Delta\hat{E}_{\phi-\frac{\pi}{2}S}^2 \right\rangle \geq \left|\left\langle \left[\hat{E}_S^+, \hat{E}_S^-\right] \right\rangle\right|^2, \tag{2.66}$$

with the equal sign applying for a MUS for the source field.

The quadrature component $\hat{E}_{\phi S}$ for the source field would be said to be *squeezed* in relation to the quadrature component $\hat{E}_{\phi-\frac{\pi}{2}S}$ if

$$\left\langle \Delta\hat{E}_{\phi S}^2 \right\rangle < \left|\left\langle \left[\hat{E}_S^+, \hat{E}_S^-\right] \right\rangle\right|. \tag{2.67}$$

With the normally ordered variance given similarly to (2.13) as

$$\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle = \left(\left\langle \hat{E}^{+2}_S \right\rangle - \left\langle \hat{E}^{+}_S \right\rangle^2 \right) e^{2i\xi} + \left(\left\langle \hat{E}^{-2}_S \right\rangle - \left\langle \hat{E}^{-}_S \right\rangle^2 \right) e^{-2i\xi} + 2\left(\left\langle \hat{E}^{-}_S \hat{E}^{+}_S \right\rangle - \left\langle \hat{E}^{-}_S \right\rangle \left\langle \hat{E}^{+}_S \right\rangle \right), \quad (2.68)$$

we have, similar to before,

$$\left\langle \Delta\hat{E}^2_{\phi S} \right\rangle = \left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle + \left\langle \left[\hat{E}^{+}_S, \hat{E}^{-}_S \right] \right\rangle. \quad (2.69)$$

In the case of the source field, the commutator $\left[\hat{E}^{+}_S, \hat{E}^{-}_S\right]$ is a Hermitian operator and not a positive c number as in the case of the total field. Its expectation value can be positive or negative depending on the quantum state.

For the case of atomic sources all confined to a region small compared to the transition wavelength, we can use results (2.35) and (2.36) together with the common delay time $\tau = |\mathbf{R} - \mathbf{R}_A|/c$ and common distance $|\mathbf{R} - \mathbf{R}_A| = R$ to obtain, for the commutator,

$$\left[\hat{E}^{+}_S, \hat{E}^{-}_S\right] = \left(1/4\pi\epsilon_0 c^2 R\right)^2 \sum_A \sum_{ijk} \omega^2_{ij}\, \mathbf{e}\cdot\mathbf{d}_{ij}\, \omega^2_{jk}\, \mathbf{e}\cdot\mathbf{d}_{jk} \times S(i,j,k)\, \hat{\Lambda}^A_{ik}(t-\tau), \quad (2.70)$$

where

$$S(i,j,k) = \begin{cases} +1, & i<j,\ k<j \\ -1, & i>j,\ k>j. \end{cases} \quad (2.71)$$

Thus the expectation value of the commutator will depend on single-atom populations and coherences. For a single two-level atom with upper and lower state populations ρ_{22} and ρ_{11}, respectively, the expectation value of the commutators is $d(\rho_{11} - \rho_{22})$, where d is positive.

For the case where the expectation value is positive

$$\left| \left\langle \left[\hat{E}^{+}_S, \hat{E}^{-}_S \right] \right\rangle \right| = \left\langle \left[\hat{E}^{+}_S, \hat{E}^{-}_S \right] \right\rangle, \quad (2.72)$$

and from (2.61) we find that squeezing in the total field $\left\langle :\Delta\hat{E}^2_{\phi}: \right\rangle < 0$ requires $\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle < 0$. From (2.69) and (2.72) it follows that

$$\left\langle \Delta\hat{E}^2_{\phi S} \right\rangle < \left| \left\langle \left[\hat{E}^{+}_S, \hat{E}^{-}_S \right] \right\rangle \right|, \quad (2.73)$$

so that the quadrature component $\hat{\mathrm{E}}_{\phi S}$ for the source field is squeezed with respect to the quadrature component $\hat{E}_{\phi - \frac{\pi}{2} S}$.

If the expectation value is negative

$$\left| \left\langle \left[\hat{E}^{+}_S, \hat{E}^{-}_S \right] \right\rangle \right| = -\left\langle \left[\hat{E}^{+}_S, \hat{E}^{-}_S \right] \right\rangle, \quad (2.74)$$

then we cannot have squeezing in the total field, since from (2.69) and (2.74) the inequality $\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle < 0$ implies that $\left\langle \Delta\hat{E}^2_{\phi S} \right\rangle$ is negative, which is impossible. For the case of the two-level atom we thus require that the population is *not* to be inverted if squeezing in the total field is to occur. For the case where the expectation value is negative one can still have the quadrature component $\hat{\mathrm{E}}_{\phi S}$ squeezed with respect to the quadrature component $\hat{E}_{\phi - \frac{\pi}{2} S}$, even though in these circumstances the total field is not squeezed. This distinction between squeezing in the source field and squeezing in the total field is an important one.

The criterion for the source field to be squeezed in the case where the expectation value is negative is for the *antinormally* ordered variance $\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle$ to be negative [8(b),10,24], where the antinormally ordered variance is given by an expression similar to (2.68), but with $\left\langle \hat{E}^{-}_S\ \hat{E}^{+}_S \right\rangle$ replaced by $\left\langle \hat{E}^{+}_S\ \hat{E}^{-}_S \right\rangle$, and in this case

$$\left\langle \Delta\hat{E}^2_{\phi S} \right\rangle = \left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle - \left\langle \left[\hat{E}^{+}_S, \hat{E}^{-}_S \right] \right\rangle. \quad (2.75)$$

Hence in view of (2.73) and (2.74), squeezing in $\hat{E}_{\phi S}$ relative to $\hat{\mathrm{E}}_{\phi - \frac{\pi}{2} S}$ requires $\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle$ to be negative. These results are in accord with those of previous authors [8(b),10,24]. In terms of the notation of Barnett [24] $\hat{A} \to \hat{E}_{\phi S}$, $\hat{B} \to \hat{E}_{\phi - \frac{\pi}{2} S}$, $\hat{D} \to 2\hat{E}^{-}_S e^{-i\xi}$, $\hat{D}^{\dagger} \to 2\hat{E}^{+}_S e^{i\xi}$, and $\langle :\Delta A^2: \rangle \to \left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle$, so that there is an interchange of the convention for normal and antinormal ordering.

We do, however, find that the source field can be a MUS. From (2.66) and (2.69) we require, for a MUS,

$$\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle \left\langle :\Delta\hat{E}_{\phi - \frac{\pi}{2} S}: \right\rangle + \left\langle \left[\hat{E}^{+}_S, \hat{E}^{-}_S \right] \right\rangle \times \left(\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle + \left\langle :\Delta\hat{E}_{\phi - \frac{\pi}{2} S}: \right\rangle \right) = 0. \quad (2.76)$$

As both terms are proportional to $\left(\frac{1}{R}\right)^4$ a MUS is possible.

D. Optimizing the squeezing with respect to quadrature phase

The normally ordered variance of the source field can be written in units of a suitable normalizing constant K (to be chosen later) as

$$\frac{\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle}{K^2} = \alpha e^{2i\xi} + \alpha^* e^{-2i\xi} + \beta, \quad (2.77)$$

where

$$\alpha = \left(\left\langle \hat{E}^{+2}_S \right\rangle - \left\langle \hat{E}^{+}_S \right\rangle^2 \right)/K^2 \quad (2.78)$$

and

$$\beta = 2\left(\left\langle \hat{E}^{-}_S \hat{E}^{+}_S \right\rangle - \left\langle \hat{E}^{-}_S \right\rangle \left\langle \hat{E}^{+}_S \right\rangle \right)/K^2. \quad (2.79)$$

From (2.69) the variance of the source field can be written in units of K^2 as

$$\frac{\left\langle \Delta\hat{E}^2_{\phi S} \right\rangle}{K^2} = \alpha e^{2i\xi} + \alpha^* e^{-2i\xi} + \beta + \gamma, \quad (2.80)$$

where the expectation value of the commutator is parametrized as

$$\gamma = \frac{\left\langle \left[\hat{E}_S^+, \hat{E}_S^-\right]\right\rangle}{K^2}. \tag{2.81}$$

The normally ordered variance for the source field can be optimized with respect to the choice of quadrature component ϕ, quadrature frequency ω, and observation time t. All these quantities appear only in the quantity $\xi = \omega t - \mathbf{k}\cdot\mathbf{R} + \phi$. Minimizing the normally ordered variance $\left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle$ with respect to ξ we obtain

$$\left(\frac{\left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle}{K^2}\right)_{\text{opt}\,\phi} = -2|\alpha| + \beta, \tag{2.82}$$

with the choice

$$e^{2i\xi} = -\sqrt{\frac{\alpha^*}{\alpha}}. \tag{2.83}$$

For the same ξ the normally ordered variances for the other quadrature component is $\left(\xi \to \xi - \frac{\pi}{2}\right)$

$$\left(\frac{\left\langle : \Delta\hat{E}_{\phi-\frac{\pi}{2}S}^2 : \right\rangle}{K^2}\right)_{\text{opt}\phi} = 2|\alpha| + \beta. \tag{2.84}$$

From (2.80) and (2.82) the expression for the variance of the optimum quadrature component of the source field is

$$\left(\frac{\left\langle \Delta\hat{E}_{\phi S}^2 \right\rangle}{K^2}\right)_{\text{opt}\phi} = -2|\alpha| + \beta + \gamma. \tag{2.85}$$

To obtain a MUS for the source field we require for this optimum choice of quadrature phase that α,β,γ satisfy

$$\left(\beta^2 - 4|\alpha|^2\right) + 2\beta\gamma = 0. \tag{2.86}$$

III. SQUEEZING FOR SINGLE THREE-LEVEL ATOMS

We now consider the specific source consisting of a single three-level atom located at $\mathbf{R} = \mathbf{0}$.

A. Description of the three-level atom states

The Λ, V, and ladder configurations, with allowed electric dipole transitions indicated, are illustrated in Fig. 1. States $|1\rangle$ and $|3\rangle$ have opposite parity to state $|2\rangle$.

For the Λ and V configurations, if new orthogonal states $|1'\rangle$,$|2'\rangle$,$|3'\rangle$ are introduced via the transformations

$$|1'\rangle = C_{11}|1\rangle + C_{31}|3\rangle, \tag{3.1}$$

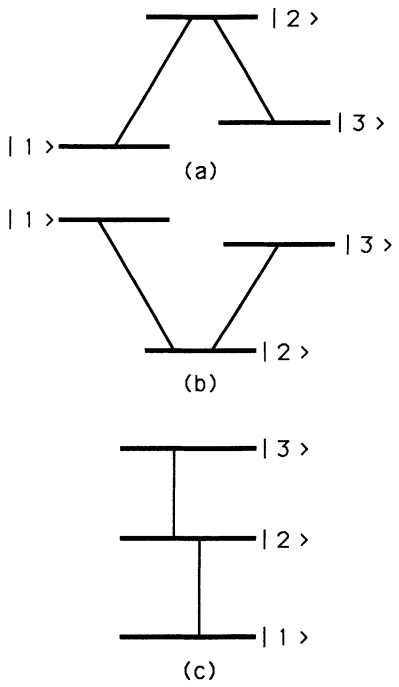

FIG. 1. Three-level systems: (a) Λ, (b) V, and (c) ladder and allowed transitions.

$$|2'\rangle = |2\rangle, \tag{3.2}$$

$$|3'\rangle = C_{13}|1\rangle + C_{33}|3\rangle, \tag{3.3}$$

then the same electric dipole transitions would occur as for the original states and with particular atomic transitions still associated with the same allowed electric dipole processes within the rotating wave approximation. Thus in the Λ case $|1\rangle \to |2\rangle$ and $|1'\rangle \to |2'\rangle$ transitions will involve the *absorption* of a photon for both original and new states whereas in the V case the *emission* of a photon would be involved for these transitions.

For the ladder case no new states introduced as in (3.1)–(3.3) are possible that would have the same allowed dipole and rotating wave approximation processes as for the original. For example, the original $|3\rangle \to |2\rangle$ transition involves the *emission* of a photon. However, for states as in (3.1) – (3.3) $|3'\rangle \to |2'\rangle$ would involve partly the *emission* (the $|3\rangle \to |2\rangle$ component) and partly the *absorption* (the $|1\rangle \to |2\rangle$ component) of a photon. Thus, while we may choose to describe the three-level atom in the Λ and V configuration cases with new orthogonal states in which the original $|1\rangle$ and $|3\rangle$ states are replaced by states $|1'\rangle$ and $|3'\rangle$, this new description is not useful for the ladder configuration.

The transformations (3.1)–(3.3) will be applied below to simplify the discussion of optimum squeezing in Λ and V systems. In this application the coefficients will be chosen in accordance with the relative contributions the $|1\rangle \to |2\rangle$ and $|3\rangle \to |2\rangle$ transitions make to the source field. It is therefore unrelated to any specific process the three-level system may be subjected to, such

as the effect of external laser fields. Transformations of the form (3.1) – (3.3) are also useful in the description of specific processes. For example, in the case of a Λ system driven by two classical laser fields, one of frequency ω_a and phase ϕ_a coupled to the $|1\rangle \rightarrow |2\rangle$ transition and the other of frequency ω_b and phase ϕ_b coupled to the $|3\rangle \rightarrow |2\rangle$ transition, the simple phase rotation given by $c_{11} = \exp[i(\omega_a t + \phi_a)]$, $c_{31} = 0$, and $c_{13} = 0$, $c_{33} = \exp[i(\omega_b t + \phi_b)]$ leads to the new density matrix elements satisfying Bloch equations with time-independent coefficients. Furthermore, when the system is at two-photon resonance ($\omega_1 + \omega_a = \omega_2 + \omega_b$) the overall transformation given by $c_{11} = \xi_a \exp[i(\omega_a t + \phi_a)]/\xi$, $c_{31} = \xi_b \exp[i(\omega_b t + \phi_b)]/\xi$, $c_{31} = -\xi_b \exp[i(\omega_b t + \phi_b)]/\xi$ (where ξ_a,ξ_b, are the two Rabi frequencies and $\xi = [(\xi_a^2 + \xi_b^2)]^{1/2}$) gives new density matrix elements satisfying Bloch equations which show that the state $|3'\rangle$ is decoupled from the other states $|1'\rangle$ and $|2'\rangle$.

This is an example of a dressed atom picture [25] transformation, applied to the case of classical fields. For atoms interacting with quantum fields the situation is more complicated since the "source" now involves these quantum fields and the dressed atom picture will involve combinations of products of atomic states with Fock states of the driving fields. In applications of (3.1) – (3.3) to describing specific processes, the aim is to find a transformation that in effect diagonalizes part of the Hamiltonian.

In all cases we will refer to the atomic density matrix elements as follows. The *one-photon atomic coherences* are ρ_{12},ρ_{21},ρ_{23},ρ_{32}; the *two-photon atomic coherences* are ρ_{13},ρ_{31}, and the *populations* are ρ_{11},ρ_{22},ρ_{33}. For the Λ and V configurations the same terminology is used for the atomic density matrix elements involving the new states $|1'\rangle$,$|2'\rangle$,$|3'\rangle$. The appropriateness of this terminology describing atomic density matrix elements as *n-photon atomic coherences* follows from an examination of the response of two- and three-level atoms prepared in phase-dependent initial states and considered as detectors of the em field. For a two-level atom detector Barnett and Pegg [26] show that the detector response associates the one-photon atomic coherences with the one-photon correlation functions for the field $\left\langle \hat{E}^{\pm}(t) \right\rangle$. For a three-level atom detector Dalton and Knight [27] show that the detector response associates the two-photon atomic coherences with two-photon correlation functions for the field $\left\langle \hat{E}^{s}(t_1)\, \hat{E}^{r}(t_2) \right\rangle$ $(s, r = +, -)$.

The possible states of the atomic system are specified by the atomic density matrix and the five distinct types of state are illustrated in Fig. 2. Note that as the state may change with time, a state with two-photon coherence could become a state with a single one-photon coherence. For the Λ and V configurations the new atomic density matrix elements can be easily related to the old atomic density matrix elements using (3.1) –(3.3).

We find that (a) the *new two-photon coherences* ρ'_{13}, ρ'_{31} are linear combinations of the *old two-photon coherences* ρ_{13}, ρ_{31} and the *old 1,3 populations* ρ_{11}, ρ_{33}; (b) the new *one-photon coherences* $\rho'_{12}, \rho'_{21}, \rho'_{23}, \rho'_{32}$ are linear combinations of the *old one-photon coherences* $\rho_{12}, \rho_{21}, \rho_{23}, \rho_{32}$; (c) the *new 1,3 populations* ρ'_{11}, ρ'_{33} are linear combinations of the *old 1,3 populations* ρ_{11}, ρ_{33} and the old *two-photon coherences* ρ_{13}, ρ_{31}; and (d) the *new 2 population* ρ'_{22} is the same as the *old 2 population* ρ_{22}.

Atomic State	State with:	Atomic Density Matrix	Type of Squeezing	Occurs for
A	Single One Photon Coherence	$\begin{pmatrix} \bullet & \bullet & 0 \\ \bullet & \bullet & 0 \\ 0 & 0 & \bullet \end{pmatrix}$ or $\begin{pmatrix} \bullet & 0 & 0 \\ 0 & \bullet & \bullet \\ 0 & \bullet & \bullet \end{pmatrix}$	Two Level	Λ,V,Ξ
B	Pair of One Photon Coherences	$\begin{pmatrix} \bullet & \bullet & 0 \\ \bullet & \bullet & \bullet \\ 0 & \bullet & \bullet \end{pmatrix}$	Two Level Pair	Λ,V,Ξ
C	Two Photon Coherence	$\begin{pmatrix} \bullet & 0 & \bullet \\ 0 & \bullet & 0 \\ \bullet & 0 & \bullet \end{pmatrix}$	Three Level	Ξ
D	Two Photon Coherence and Single One Photon Coherence	$\begin{pmatrix} \bullet & \bullet & \bullet \\ \bullet & \bullet & 0 \\ \bullet & 0 & \bullet \end{pmatrix}$ or $\begin{pmatrix} \bullet & 0 & \bullet \\ 0 & \bullet & \bullet \\ \bullet & \bullet & \bullet \end{pmatrix}$	Two Level-Three Level	Λ,V,Ξ
E	General Coherence	$\begin{pmatrix} \bullet & \bullet & \bullet \\ \bullet & \bullet & \bullet \\ \bullet & \bullet & \bullet \end{pmatrix}$	General	Λ,V,Ξ

FIG. 2. The distinct states and types of squeezing for three-level systems. A solid dot indicates a nonzero atomic density matrix element.

As will be seen below, the phase dependence of the variance of the source-field quadrature component depends in general on both one-photon coherences (all three cases) and on two-photon coherences (ladder case only). Various types of source-field squeezing can be distinguished depending on which new coherences are nonzero for the source-field state in question. These are shown in Fig. 2, along with the corresponding atomic states.

B. Squeezing expressions for light from three-level atoms

For the three configurations involved the contributions to the positive frequency component of the source-field expressions for E_S^+ can be obtained from (2.35) and these are listed in Table I. The retarded time $t - R/c$ is implicit in the transition operators, where we have written

TABLE I. Contributions to electric field $\hat{E}_S^+$.

	Λ	V	Ladder
$\hat{E}_S^+$	$K(c\hat{\Lambda}_{32} + s\hat{\Lambda}_{12})$	$K(c\hat{\Lambda}_{23} + s\hat{\Lambda}_{21})$	$K(c\hat{\Lambda}_{23} + s\hat{\Lambda}_{12})$

TABLE II. The quantities λ_3 and λ_1.

	Λ	V	Ladder
λ_3	$\omega_{23}^2 \langle 2 \vert \hat{\mathbf{d}} \cdot \mathbf{e} \vert 3 \rangle$	$\omega_{32}^2 \langle 3 \vert \hat{\mathbf{d}} \cdot \mathbf{e} \vert 2 \rangle$	$\omega_{32}^2 \langle 3 \vert \hat{\mathbf{d}} \cdot \mathbf{e} \vert 2 \rangle$
λ_1	$\omega_{21}^2 \langle 2 \vert \hat{\mathbf{d}} \cdot \mathbf{e} \vert 1 \rangle$	$\omega_{12}^2 \langle 1 \vert \hat{\mathbf{d}} \cdot \mathbf{e} \vert 2 \rangle$	$\omega_{21}^2 \langle 2 \vert \hat{\mathbf{d}} \cdot \mathbf{e} \vert 1 \rangle$

$$K = \sqrt{\lambda_3^2 + \lambda_1^2} / \left(4\pi\epsilon_0 c^2 R\right), \tag{3.4}$$

$$K = \sqrt{2}\, \overline{(\omega^2 \mu)} / \left(4\pi\epsilon_0 c^2 R\right), \tag{3.5}$$

$$c = \lambda_3 / \sqrt{\lambda_3^2 + \lambda_1^2}, \tag{3.6}$$

$$s = \lambda_1 / \sqrt{\lambda_3^2 + \lambda_1^2}, \tag{3.7}$$

$$\overline{(\omega^2 \mu)} = \sqrt{\frac{1}{2}\left(\lambda_3^2 + \lambda_1^2\right)}. \tag{3.8}$$

The quantities λ_1,λ_3 are given in terms of electric dipole matrix elements and atomic transition frequencies in Table II. $\overline{(\omega^2\mu)}$ is the root mean square average of the dipole matrix element times the square of the transition frequency. The atomic states can be chosen to make the dipole matrix elements and hence the quantities λ_1,λ_3, s, c, K all real.

The source-field operators $\hat{E}_S^{\pm}$ and their squares are then expressed in terms of atomic transition and population operators. As only one-time atomic operators are involved, evaluation of the expressions Tr $[\hat{\Omega}(t)\hat{W}(0)]$ will then yield results in terms of the matrix elements ρ_{ij} of the atomic density operator $\hat{\rho}$. Results for the various expectation values of source field operators are given in Table III.

The normally ordered and total variances in the quadrature component $\hat{E}_{\varphi S}$ are given by

(a) the Λ configuration

$$\frac{\left\langle : \Delta \hat{E}_{\phi S}^2 : \right\rangle}{K^2} = 2\rho_{22} - \left[\left(c\rho_{23} + s\rho_{21}\right) e^{i\xi} + \left(c\rho_{32} + s\rho_{12}\right) e^{-i\xi}\right]^2, \tag{3.9}$$

$$\frac{\left\langle \Delta \hat{E}_{\phi S}^2 \right\rangle}{K^2} = \rho_{22} + c^2\rho_{33} + s^2\rho_{11} + cs\left(\rho_{13} + \rho_{31}\right) - \left[\left(c\rho_{23} + s\rho_{21}\right) e^{i\xi} + \left(c\rho_{32} + s\rho_{12}\right) e^{-i\xi}\right]^2; \tag{3.10}$$

(b) the V configuration

$$\frac{\left\langle : \Delta \hat{E}_{\phi S}^2 : \right\rangle}{K^2} = 2[c^2\rho_{33} + cs(\rho_{13} + \rho_{31}) + s^2\rho_{11}] - [(c\rho_{32} + s\rho_{12})e^{i\xi} + (c\rho_{23} + s\rho_{21})e^{-i\xi}]^2, \tag{3.11}$$

$$\frac{\left\langle \Delta \hat{E}_{\phi S}^2 \right\rangle}{K^2} = \rho_{22} + c^2\rho_{33} + s^2\rho_{11} + cs\left(\rho_{13} + \rho_{31}\right) - \left[\left(c\rho_{32} + s\rho_{12}\right) e^{i\xi} + \left(c\rho_{23} + s\rho_{21}\right) e^{-i\xi}\right]^2; \tag{3.12}$$

and (c) the ladder configuration

$$\frac{\left\langle : \Delta \hat{E}_{\phi S}^2 : \right\rangle}{K^2} = 2\left(c^2\rho_{33} + s^2\rho_{22}\right) - \left[\left(c\rho_{32} + s\rho_{21}\right) e^{i\xi} + \left(c\rho_{23} + s\rho_{12}\right) e^{-i\xi}\right]^2 + cs\left(\rho_{31} e^{2i\xi} + \rho_{13} e^{-2i\xi}\right), \tag{3.13}$$

$$\frac{\left\langle \Delta \hat{E}_{\phi S}^2 \right\rangle}{K^2} = \rho_{22} + c^2\rho_{33} + s^2\rho_{11} - [(c\rho_{32} + s\rho_{21})e^{i\xi} + (c\rho_{23} + s\rho_{12})e^{-i\xi}]^2 + sc\left(\rho_{31} e^{2i\xi} + \rho_{13} e^{-2i\xi}\right). \tag{3.14}$$

The quadrature phase dependence of the variance depends on the one-photon atomic coherences ρ_{12},ρ_{21},ρ_{32},ρ_{23} in all cases, but on the two-photon atomic coherences ρ_{13},ρ_{31} only in the ladder case. The result for the optimized choice of normally ordered variance for $\hat{E}_{\phi S}$ can be obtained from (2.82) and that for the corresponding normally ordered variance $\hat{E}_{\phi - \frac{\pi}{2} S}$ from (2.84). The variance is given by (2.85). The quantities α,β,γ can be read off from (3.9)–(3.14).

The atomic density matrix is parametrized (see Appendix B) in terms of the quantities P_1, P_2, P_3 (popula-

TABLE III. Expectation values of electric field operators.

	Λ	V	Ladder
$\frac{\langle \hat{E}_S^{+2} \rangle}{K^2} = \frac{\langle \hat{E}_S^{-2} \rangle^*}{K^2}$	0	0	$sc\rho_{31}$
$\frac{\langle \hat{E}_S^{+} \rangle}{K} = \frac{\langle \hat{E}_S^{-} \rangle^*}{K}$	$c\rho_{23} + s\rho_{21}$	$c\rho_{32} + s\rho_{12}$	$c\rho_{32} + s\rho_{21}$
$\frac{\langle \hat{E}_S^{-} \hat{E}_S^{+} \rangle}{K^2}$	ρ_{22}	$c^2\rho_{33} + s^2\rho_{11} + cs\left(\rho_{13} + \rho_{31}\right)$	$c^2\rho_{33} + s^2\rho_{22}$
$\frac{\langle [\hat{E}_S^{+}, \hat{E}_S^{-}] \rangle}{K^2}$	$c^2\left(\rho_{33} - \rho_{22}\right) + s^2\left(\rho_{11} - \rho_{22}\right) + cs\left(\rho_{13} + \rho_{31}\right)$	$c^2\left(\rho_{22} - \rho_{33}\right) + s^2\left(\rho_{22} - \rho_{11}\right) - cs\left(\rho_{13} + \rho_{31}\right)$	$\left(c^2 - s^2\right)\rho_{22} - c^2\rho_{33} + s^2\rho_{11}$

tions) and $S_1, S_2, S_3, S_4, S_5, S_6$, (coherences) as

$$[\rho_{ij}] = \begin{pmatrix} P_1 & \frac{1}{2}(S_1 - iS_2) & \frac{1}{2}(S_5 - iS_6) \\ \frac{1}{2}(S_1 + iS_2) & P_2 & \frac{1}{2}(S_3 - iS_4) \\ \frac{1}{2}(S_5 + iS_6) & \frac{1}{2}(S_3 + iS_4) & P_3 \end{pmatrix}. \tag{3.15}$$

These parameters are not independent since the density operator must be Hermitean, have a trace of unity, and be a positive operator. For the case of a three-level system this leads to constraints (see Appendix B)

$$P_1, P_2, P_3, S_1 \cdots S_6 \text{ are real}, \tag{3.16}$$

$$\mathrm{Tr}\hat{\rho} = P_1 + P_2 + P_3 = 1, \tag{3.17}$$

$$\mathrm{Tr}\hat{\rho}^2 = P_1^2 + P_2^2 + P_3^2 + \frac{1}{2}\left(S_1^2 + S_2^2 + \cdots + S_6^2\right) \leq 1, \tag{3.18}$$

$$\begin{aligned} |\hat{\rho}| = {} & P_1P_2P_3 - \frac{1}{4}P_1\left(S_3^2 + S_4^2\right) - \frac{1}{4}P_2\left(S_5^2 + S_6^2\right) \\ & - \frac{1}{4}P_3\left(S_1^2 + S_2^2\right) + \frac{1}{4}\left[S_5\left(S_1S_3 - S_2S_4\right)\right. \\ & \left. + S_6\left(S_1S_4 + S_2S_3\right)\right] \geq 0. \end{aligned} \tag{3.19}$$

The constraints (3.18) and (3.19) are equivalent to the constraints (B11) and (B12), which are necessary if the density operator is to be positive. The special forms of the constraints (3.18) and (3.19) apply to a three-level system. Equations (3.16) and (3.17) ensure that the eigenvalues of $\hat{\rho}$ (λ_1, λ_2, and λ_3) are real and satisfy $\lambda_1 + \lambda_2 + \lambda_3 = 1$. To ensure that $\hat{\rho}$ is positive we then require the eigenvalues to be ≥ 0. Equation (3.18) ensures that $\lambda_1^2 + \lambda_2^2 + \lambda_3^2 \leq 1$, while (3.19) ensures that $\lambda_1\lambda_2\lambda_3 \geq 0$. There is no possibility that three real numbers add up to one, are less than one in magnitude, and have a positive product without them all being between 0 and 1.

C. Optimized squeezing in Λ and V configurations

It is convenient to change to new orthogonal bases of the form given by (3.1) – (3.3) and to write the normally ordered variances in terms of the new density matrix elements identifying combinations of the old density matrix elements that occurred in the original expressions (3.9) – (3.12) for the variances as new density matrix elements. For both Λ and V cases the new states $|1'\rangle$ and $|3'\rangle$ are

$$|1'\rangle = s|1\rangle + c|3\rangle, \tag{3.20}$$

$$|2'\rangle = |2\rangle, \tag{3.21}$$

$$|3'\rangle = -c|1\rangle + s|3\rangle, \tag{3.22}$$

so that

$$\rho'_{12} = s\rho_{12} + c\rho_{32}, \tag{3.23}$$

$$\rho'_{21} = s\rho_{21} + c\rho_{23}, \tag{3.24}$$

$$\rho'_{22} = \rho_{22}, \tag{3.25}$$

$$\rho'_{11} = s^2\rho_{11} + sc\rho_{13} + sc\rho_{31} + c^2\rho_{33}, \tag{3.26}$$

and the normally ordered and total variances are given by, for the Λ configuration,

$$\frac{\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle}{K^2} = 2\rho'_{22} - \left(\rho'_{21}e^{i\xi} + \rho'_{12}e^{-i\xi}\right)^2, \tag{3.27}$$

$$\begin{aligned} \frac{\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle}{K^2} = {} & -\rho'^2_{21}e^{2i\xi} - \rho'^2_{12}e^{-2i\xi} \\ & + 2\left(\rho'_{22} - \rho'_{21}\rho'_{12}\right), \end{aligned} \tag{3.28}$$

$$\frac{\left\langle \Delta\hat{E}^2_{\phi S} \right\rangle}{K^2} = \rho'_{22} + \rho'_{11} - \left(\rho'_{21}e^{i\xi} + \rho'_{12}e^{-i\xi}\right)^2. \tag{3.29}$$

This is the same as for a two-level atom with $|2'\rangle$ as the upper state and $|1'\rangle$ the lower state. For the V configuration

$$\frac{\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle}{K^2} = 2\rho'_{11} - \left(\rho'_{12}e^{i\xi} + \rho'_{21}e^{-i\xi}\right)^2, \tag{3.30}$$

$$\begin{aligned} \frac{\left\langle :\Delta\hat{E}^2_{\phi S}: \right\rangle}{K^2} = {} & -\rho'^2_{12}e^{2i\xi} - \rho'^2_{21}e^{-2i\xi} \\ & + 2\left(\rho'_{11} - \rho'_{12}\rho'_{21}\right), \end{aligned} \tag{3.31}$$

$$\frac{\left\langle \Delta\hat{E}^2_{\phi S} \right\rangle}{K^2} = \rho'_{22} + \rho'_{11} - \left(\rho'_{12}e^{i\xi} + \rho'_{21}e^{-i\xi}\right)^2. \tag{3.32}$$

This is the same as for a two-level atom with $|1'\rangle$ as the upper state and $|2'\rangle$ the lower state. The equivalence of expressions for the variances to those for a two-level atom with states $|1'\rangle$, $|2'\rangle$, and $|3'\rangle$ that occurs for Λ and V systems does not imply that all squeezing is automatically two level (see Fig. 2). Other coherences such as ρ'_{23} or ρ'_{13} could still be nonzero and thereby play a role in the dynamics.

The details of the optimization for the squeezing in the case of Λ and V configurations is given in Appendix C. After setting all the other parameters to zero, the equations of constraints are written in terms of P'_1,P'_2,S'_1,S'_2. Identifying the quantities α,β,γ of (2.78), (2.79), and (2.81), optimizing with respect to the choice of ϕ,ω,t using (2.82), and then using the Lagrange method of undetermined multipliers, we arrive at the optimized choice

TABLE IV. Parameter values for optimum squeezing ($c = s = \frac{1}{\sqrt{2}}$ for the ladder case).

	P_1'	P_2'	P_3'	$(S_1'^2 + S_2'^2)$	S_3'	S_4'	S_5'	S_6'	α	β	γ
Λ	$\frac{3}{4}$	$\frac{1}{4}$	0	$\frac{3}{4}$	0	0	0	0	$-\frac{3}{16}$	$\frac{1}{8}$	$\frac{1}{2}$
V	$\frac{1}{4}$	$\frac{3}{4}$	0	$\frac{3}{4}$	0	0	0	0	$-\frac{3}{16}$	$\frac{1}{8}$	$\frac{1}{2}$
	P_1	P_2	P_3	S_1	S_2	S_3	S_4	$(S_5^2 + S_6^2)$	α	β	γ
Ladder	$\frac{1}{2}\left(1 + \frac{1}{\sqrt{2}}\right)$	0	$\frac{1}{2}\left(1 - \frac{1}{\sqrt{2}}\right)$	0	0	0	0	$\frac{1}{2}$	$\frac{\sqrt{2}}{8}e^{i\theta}$	$\frac{1}{2}\left(1 - \frac{1}{\sqrt{2}}\right)$	$\frac{\sqrt{2}}{4}$

of P_1', P_2', S_1', S_2'. The quantities α, β, γ can then be calculated.

The values of the parameters $P_1', P_2', \ldots, S_6'$ and the quantities α, β, γ for the situation of optimized squeezing in the Λ and V configuration cases are given in Table IV. The results for the variances are given in Table V.

For the Λ configuration it is found that the source field is squeezed for the optimum value of the normally ordered variance which is given by

$$\left(\frac{\left\langle : \Delta \hat{E}_{\phi S}^2 : \right\rangle}{K^2}\right)_{\text{opt } \phi} = -\frac{1}{4} \tag{3.33}$$

or

$$\left\langle : \Delta \hat{E}_{\phi S}^2 : \right\rangle_{\text{opt } \phi} = -0.50 \times \left[\overline{(\omega^2 \mu)}/4\pi\epsilon_0 c^2 R\right]^2 . \tag{3.34}$$

The atomic state involved is a pure state and is associated with a state vector given in terms of both the new and original atomic states as

$$|\psi\rangle = \frac{1}{\sqrt{4}}|2'\rangle + \sqrt{\frac{3}{4}}e^{i\theta}|1'\rangle \tag{3.35}$$

$$= \frac{1}{\sqrt{4}}|2\rangle + \sqrt{\frac{3}{4}}e^{i\theta}\left(\frac{\lambda_1}{\sqrt{\lambda_1^2 + \lambda_3^2}}|1\rangle + \frac{\lambda_3}{\sqrt{\lambda_1^2 + \lambda_3^2}}|3\rangle\right), \tag{3.36}$$

where θ is arbitrary. This result corresponds to that of Barnett and Knight [18] for a two-level atom: the Bloch vector is oriented at 120° from the orientation of the upper state. The value of the normally ordered variance also corresponds to previous results. In terms of the new basis set the atomic state is one with a single one-photon coherence $\rho_{12}' = \rho_{21}'^*$ and the situation can be described as that of *two-level squeezing*. The other state $|3'\rangle$ does not become involved. This effective two-level behavior is in spite of the apparent situation of general coherence if the original basis set is used to describe the atomic state.

The condition (2.86) is satisfied and hence in the case of the Λ system the source field is in a MUS. The positive value of γ confirms condition (2.72) that the population is not inverted and is in accord with (2.73) that squeezing is found in the source field (3.33). Equation (2.62) shows that the total electric field is squeezed. The normally ordered variance for the other quadrature component $\hat{E}_{\phi - \frac{\pi}{2}S}$ of the source field and the variance for $\hat{E}_{\phi S}$ itself are given in Table V.

For the V configuration it is found that the source field is also squeezed with the corresponding atomic state a pure state. In this case

$$\left(\frac{\left\langle : \Delta \hat{E}_{\phi S}^2 : \right\rangle}{K^2}\right)_{\text{opt } \phi} = -\frac{1}{4}. \tag{3.37}$$

The corresponding atomic state vector is

$$|\psi\rangle = \frac{1}{\sqrt{4}}|1'\rangle + \sqrt{\frac{3}{4}}e^{i\theta}|2'\rangle \tag{3.38}$$

$$= \frac{1}{\sqrt{4}}\left(\frac{\lambda_1}{\sqrt{\lambda^2 + \lambda_3^2}}|1\rangle + \frac{\lambda_3}{\sqrt{\lambda_1^2 + \lambda_3^2}}|3\rangle\right) + \sqrt{\frac{3}{4}}e^{i\theta}|2\rangle , \tag{3.39}$$

where again θ is arbitrary. The atomic state is again one with a single one-photon coherence $\rho_{12}' = \rho_{21}'^*$ and the situation one of *two level squeezing*. All that differs from the Λ case is that the upper and lower states are relabeled.

In the case of V system (2.86) is satisfied and the source field is again a MUS. The positive value of γ confirms condition (2.72) and in accord with (2.73) that squeezing is found in the source field (3.37). The total electric field is squeezed and the normally ordered variance for $\hat{E}_{\phi - \frac{\pi}{2}S}$

TABLE V. Variances for optimum squeezing ($c = s = \frac{1}{\sqrt{2}}$ for the ladder case).

	$\left(\left\langle : \Delta \hat{E}_{\phi S}^2 : \right\rangle / K^2\right)_{\text{opt } \phi}$	$\left(\left\langle : \Delta \hat{E}_{\phi - \frac{\pi}{2}S} : \right\rangle / K^2\right)_{\text{opt } \phi}$	$\left(\left\langle \Delta \hat{E}_{\phi}^2 \right\rangle / K^2\right)_{\text{opt } \phi}$
Λ	$-\frac{1}{4}$	$\frac{1}{2}$	$\frac{1}{4}$
V	$-\frac{1}{4}$	$\frac{1}{2}$	$\frac{1}{4}$
Ladder	$-\frac{1}{2}\left(\sqrt{2} - 1\right)$	$\frac{1}{2}$	$\frac{1}{2}\left(1 - 1/\sqrt{2}\right)$

and the variance for $\hat{E}_{\phi S}$ itself are given in Table V.

For both Λ and V cases and for the special case where both transitions contribute equally $\left(c = s = 1/\sqrt{2}\right)$ the new state $|1'\rangle$ is $(|1\rangle + |3\rangle)/\sqrt{2}$. The uninvolved state is $(-|1\rangle + |3\rangle)/\sqrt{2}$.

D. Optimized squeezing in ladder configuration

In this case no convenient change of basis is available and we merely optimize the normally ordered variance using the parametrization of the atomic density matrix elements and the equations of constraint given by Eqs. (3.13) and (3.16)–(3.19) using numerical methods. The details of the optimization in the ladder configuration case are given in Appendix C. The quantities α,β,γ of (2.78), (2.79), and (2.81) are first identified and optimization with respect to the choice of ϕ,ω,t, carried out using (2.82). Numerical methods [28] are used to establish that some of the parameters for the minimum are zero and the remainder are then determined using Lagrange undetermined multipliers. The calculations were restricted to the case $c = s = \frac{1}{\sqrt{2}}$, where both transitions contribute equally.

The values of the parameters $P_1,P_2,\ldots,S_6$ and the quantities α,β,γ for the situation of optimum squeezing in the ladder configuration cases are given in Table IV. The results for the variances are given in Table V.

The optimum value for the normally ordered variance was found to correspond to a squeezed state and is given by

$$\left(\frac{\left\langle :\Delta E_{\phi S}^2: \right\rangle}{K^2}\right)_{\text{opt }\phi} = -\frac{1}{2}\left(\sqrt{2}-1\right) \approx -0.20 \qquad \left(c = s = \frac{1}{\sqrt{2}}\right), \quad (3.40)$$

$$\left(\left\langle :\Delta E_{\phi S}^2: \right\rangle\right)_{\text{opt }\phi} \approx -0.41 \times \left[\overline{(\omega^2\mu)}/4\pi\epsilon_0 c^2 R\right]^2 . \quad (3.41)$$

The atomic state involved is a pure state and associated with a state vector

$$|\psi\rangle = \frac{1}{\sqrt{2}}\sqrt{\left(1+\frac{1}{\sqrt{2}}\right)}\,|1\rangle + \frac{1}{\sqrt{2}}\sqrt{\left(1-\frac{1}{\sqrt{2}}\right)}e^{i\theta}\,|3\rangle \qquad \left(c = s = \frac{1}{\sqrt{2}}\right) \quad (3.42)$$

where θ is arbitrary. This state is one with a two-photon coherence $\rho_{13} = \rho_{31}^*$ and the situation can be described as one of *three-level squeezing*. For this state the middle level population and all the one-photon atomic coherences are zero. A similar state occurs at long times for a ladder system in a squeezed reservoir [29].

The condition (2.86) is satisfied and hence the source field is a MUS. The positive value of γ confirms condition (2.72) and in accord with (2.73) that squeezing is found in the source field (3.40). Equation (2.62) shows that the total field is squeezed. The normally ordered variance for the other quadrature component $\hat{E}_{\phi-\frac{\pi}{2}S}$ and the variance for $\hat{E}_{\phi S}$ itself are given in Table V.

IV. SUMMARY AND CONCLUSIONS

Squeezing in the total electric field from a multiatom source has been treated confirming that the criterion for squeezing in the total field is that the normally ordered variance of the quadrature component of the source field is negative, subject to the conditions that the initial (driving) field has zero classical amplitude at the detector and the source atoms are located in a regime small compared to the transition wavelength (Dicke atom source). Previously unexamined interference terms between the free field and the source field are shown to vanish. The concept of optimum squeezing has been introduced and expressions for the normally ordered variance of the source-field quadrature component found for the optimized choice of quadrature phase, frequency, and wave vector.

An examination of squeezing in the total electric field from a single three-level source has been carried out. Five differing types of squeezing have been distinguished for such systems, based on an examination of the various one-photon and two-photon atomic coherences involved. It is found that squeezing occurs for the Λ, V, and ladder configurations and that a minimum value for the normally ordered variance of the quadrature component can be obtained which corresponds to the maximum squeezing that could *ever* be observed in three-level atoms undergoing irreversible decay, irrespective of the choice of quadrature component, quadrature frequency, observation time, *and* specific process in which the three-level atom is involved. The corresponding atomic state is always a pure state and the source field is in a minimum uncertainty state. For the Λ and V configurations, however, the three-level atom is equivalent to a two-level system plus a noninvolved third level and the state for optimum squeezing is one with single one-photon atomic coherence and no two-photon atomic coherences. Λ and V configurations thus involve *two-level squeezing*. For the ladder configuration, on the other hand, the state is one with a two-photon atomic coherence and no one-photon atomic coherences or intermediate level population. Ladder configurations thus involve *three-level squeezing*.

ACKNOWLEDGMENTS

This work was supported in part by the U.K. Science and Engineering Research Council and by the European Community. One of us (Z.F.) acknowledges the support of the Australian Research Council. Thanks are due to Mr. Ian Mortimer for numerical calculations.

APPENDIX A: FREE-FIELD–SOURCE-FIELD COMMUTATORS

Consider the commutator

$$C = \left[\hat{\mathbf{E}}_F^+(\mathbf{R}_1,t_1), \hat{\mathbf{E}}_S^+(\mathbf{R}_2,t_2)\right], \tag{A1}$$

where the free field term is

$$\hat{\mathbf{E}}_F^+(\mathbf{R}_1,t_1) = \sum_\lambda \mathbf{f}_\lambda(\mathbf{R}_1)\,\hat{a}_\lambda(0)\,e^{-i\omega_\lambda t_1} \tag{A2}$$

$$= \sum_\lambda i\sqrt{\frac{\hbar\omega_\lambda}{2\epsilon_0 V}}e^{i\mathbf{k}_\lambda\cdot\mathbf{R}_1}\mathbf{e}_\lambda\hat{a}_\lambda(0)\,e^{-i\omega_\lambda t_1} \tag{A3}$$

and the source field term is

$$\hat{\mathbf{E}}_S^+(\mathbf{R}_2,t_2) = \sum_A \hat{\mathbf{E}}_{SA}^+(\mathbf{R}_2,t_2), \tag{A4}$$

with

$$\hat{\mathbf{E}}_{SA}^+(\mathbf{R}_2,t_2) = \nabla_2\times\left(\nabla_2\times\frac{1}{4\pi\epsilon_0}\frac{\hat{\mathbf{d}}_A^-(t_2-\tau_{A2})}{R_{A2}} \times\theta(t_2-\tau_{A2})\right) \tag{A5}$$

and

$$R_{A2} = |\mathbf{R}_2-\mathbf{R}_A|, \quad \tau_{A2} = R_{A2}/c.$$

Substituting for the source field term we find that

$$C = \sum_A \nabla_2\times\left(\nabla_2\times\frac{1}{4\pi\epsilon_0 R_{A2}} \times\left[\hat{\mathbf{E}}_F(\mathbf{R}_1,t_1), \hat{\mathbf{d}}_A^-(t_2-\tau_{A2})\right]\theta(t_2-\tau_{A2})\right), \tag{A6}$$

where we note that ∇_2 does *not* operate on $\hat{\mathbf{E}}_F^+(\mathbf{R}_1,t_1)$.

Now from the multipolar Hamiltonian and in the electric dipole approximation

$$\dot{\hat{a}}_\lambda = -i\omega_\lambda\hat{a}_\lambda - i\sum_{Aij}\frac{i}{\hbar}\sqrt{\frac{\hbar\omega_\lambda}{2\epsilon_0 V}}e^{-i\mathbf{k}_\lambda\cdot\mathbf{R}_A}\left\langle i\left|\mathbf{e}_\lambda\cdot\hat{\mathbf{d}}\right|j\right\rangle\hat{\Lambda}_{ij}^A$$

$$= -i\omega_\lambda\hat{a}_\lambda + \frac{1}{\hbar}\sum_A\sqrt{\frac{\hbar\omega_\lambda}{2\epsilon_0 V}}\mathbf{e}_\lambda\cdot\hat{\mathbf{d}}_A e^{-i\mathbf{k}_\lambda\cdot\mathbf{R}_A}. \tag{A7}$$

Formally solving leads to

$$\hat{a}_\lambda(t) = \hat{a}_\lambda(0)\,e^{-i\omega_\lambda t} + \frac{1}{\hbar}\sum_A\sqrt{\frac{\hbar\omega_\lambda}{2\epsilon_0 V}}e^{-i\mathbf{k}_\lambda\cdot\mathbf{R}_A}\mathbf{e}_\lambda\cdot e^{-i\omega_\lambda t} \times\int_o^t dt' e^{i\omega_\lambda t'}\hat{\mathbf{d}}_A(t'). \tag{A8}$$

Now

$$\hat{\mathbf{E}}^+(\mathbf{R},t) = \sum_\lambda i\sqrt{\frac{\hbar\omega_\lambda}{2\epsilon_0 V}}\mathbf{e}_\lambda e^{i\mathbf{k}_\lambda\cdot\mathbf{R}}\hat{a}_\lambda(t). \tag{A9}$$

Substituting from (A8) into expression for $\hat{\mathbf{E}}^+(\mathbf{R},t)$ and changing t to $t_2-\tau_{A2}$ we find that

$$\hat{\mathbf{E}}^+(\mathbf{R}_1,t_2-\tau_{A2}) = \sum_\mu i\sqrt{\frac{\hbar\omega_\lambda}{2\epsilon_0 V}}\mathbf{e}_\mu e^{i\mathbf{k}_\mu\cdot\mathbf{R}}\hat{a}_\mu(0)\,e^{-i\omega_\mu(t_2-\tau_{A2})} + \sum_{\mu B}\frac{i}{\hbar}\left(\frac{\hbar\omega_\mu}{2\epsilon_o V}\right)\mathbf{e}_\mu e^{i\mathbf{k}_\mu\cdot(\mathbf{R}-\mathbf{R}_B)} \times^{-i\omega_\mu(t_2-\tau_{A2})}\mathbf{e}_\mu\cdot\int_0^{t_2-\tau_{A2}}dt'\,e^{i\omega_\mu t'}\hat{\mathbf{d}}_B(t'). \tag{A10}$$

Multiply each side of (A10) by $\frac{1}{V}\,e^{i\mathbf{k}_\lambda\cdot\mathbf{R}}\mathbf{e}_\lambda$, integrate over $\mathbf{R}$, and use

$$\frac{1}{V}\int d^3\mathbf{R}e^{-i\mathbf{k}_\lambda\cdot\mathbf{R}}\mathbf{e}^{i\mathbf{k}_\mu\cdot\mathbf{R}} = \delta_{\mathbf{k}_\lambda\mathbf{k}_\mu}; \tag{A11}$$

thus we obtain for $\hat{a}_\lambda(0)$

$$i\sqrt{\frac{\hbar\omega_\lambda}{2\epsilon_0 V}}\hat{a}_\lambda(0) = e^{i\omega_\lambda(t_2-\tau_{A2})}\frac{1}{V}\int d^3\mathbf{R}\,e^{-i\mathbf{k}_\lambda\cdot\mathbf{R}}\mathbf{e}_\lambda\cdot\hat{\mathbf{E}}^+(\mathbf{R},t_2-\tau_{A2}) - \frac{i}{\hbar}\sum_B\left(\frac{\hbar\omega_\lambda}{2\epsilon_0 V}\right) \times e^{-i\mathbf{k}_\lambda\cdot\mathbf{R}_B}\mathbf{e}_\lambda\cdot\int_0^{t_2-\tau_{A2}}dt'\,e^{i\omega_\lambda t'}\hat{\mathbf{d}}_B(t'). \tag{A12}$$

After this the free-field term can be written

$$\hat{\mathbf{E}}_F^+(\mathbf{R}_1,t_1) = \sum_\lambda\mathbf{e}_\lambda\frac{1}{V}\int d^3\mathbf{R}\,e^{i\mathbf{k}_\lambda\cdot(\mathbf{R}_1-\mathbf{R})}\mathbf{e}_\lambda\cdot\hat{\mathbf{E}}^+(\mathbf{R},t_2-\tau_{A2})\,e^{i\omega_\lambda(t_2-t_1-\tau_{A2})} -\frac{i}{\hbar}\sum_{\lambda B}\left(\frac{\hbar\omega_\lambda}{2\epsilon_0 V}\right)\mathbf{e}_\lambda\,e^{i\mathbf{k}_\lambda\cdot(\mathbf{R}_1-\mathbf{R}_B)}e^{-i\omega_\lambda t_1}\int_0^{t_2-\tau_{A2}}dt'\,e^{i\omega_\lambda t'}\mathbf{e}_\lambda\cdot\hat{\mathbf{d}}_B(t'), \tag{A13}$$

which involves the total field $\hat{\mathbf{E}}^+$ and the dipole operator $\hat{\mathbf{d}}_B$.

Substituting $\hat{\mathbf{E}}_F^+(\mathbf{R}_1,t_1)$ into the expression for the commutator we find that

$$C=\sum_A \nabla_2\times\left(\nabla_2\times\frac{1}{4\pi\epsilon_0 R_{A2}}\sum_\lambda \mathbf{e}_\lambda\frac{1}{V}\int d^3\mathbf{R}e^{i\mathbf{k}_\lambda\cdot(\mathbf{R}_1-\mathbf{R})}e^{i\omega_\lambda(t_2-t_1-\tau_{A2})}\right.$$
$$\left.\times\theta(t_2-\tau_{A2})[\mathbf{e}_\lambda\cdot\hat{\mathbf{E}}^+(\mathbf{R},t_2-\tau_{A2}),\hat{\mathbf{d}}_A^-(t_2-\tau_{A2})]\right)$$
$$+\sum_A \nabla_2\times\left[\nabla_2\times\frac{1}{4\pi\epsilon_0 R_{A2}}\left(-\frac{i}{\hbar}\right)\sum_{\lambda B}\mathbf{e}_\lambda\frac{\hbar\omega_\lambda}{2\epsilon_0 V}e^{i\mathbf{k}_\lambda\cdot(\mathbf{R}_1-\mathbf{R}_B)}e^{-i\omega_\lambda t_1}\right.$$
$$\left.\times\,\theta(t_2-\tau_{A2})\int_0^{t_2-\tau_{A2}}dt'\,e^{i\omega_\lambda t'}[\mathbf{e}_\lambda\cdot\hat{\mathbf{d}}_B(t'),\hat{\mathbf{d}}_A^-(t_2-\tau_{A2})]\right]. \tag{A14}$$

The first term is zero, since as $\hat{\mathbf{E}}^+(\mathbf{R},0)=i\sum\mathbf{e}_\lambda e^{i\mathbf{k}_\lambda\cdot\mathbf{R}}\hat{a}_\lambda(0)$ commutes with $\hat{\mathbf{d}}_A^-(0)$, then $\hat{\mathbf{E}}^+(\mathbf{R},t_2-\tau_{A2})$ will commute with $\hat{\mathbf{d}}_A^-(t_2-\tau_{A2})$. Hence

$$C=\sum_{AB}\nabla_2\times\left(\nabla_2\times\frac{1}{4\pi\epsilon_0 R_{A2}}\left(\frac{-i}{2\epsilon_0}\right)\right.$$
$$\times\int_0^{t_2-\tau_{A2}}dt'\,\frac{1}{V}\sum_\lambda e^{i\mathbf{k}_\lambda\cdot(\mathbf{R}_1-\mathbf{R}_B)}$$
$$\left.\times e^{i\omega_\lambda(t'-t_1)}\omega_\lambda\mathbf{e}_\lambda[\mathbf{e}_\lambda\cdot\hat{\mathbf{d}}_B(t'),\hat{\mathbf{d}}_A^-(t_2-\tau_{A2})]\right.$$
$$\left.\times\theta(t_2-\tau_{A2})\right]. \tag{A15}$$

Now

$$\frac{1}{V}\sum_\lambda=\frac{1}{(2\pi)^3}\int d^3\mathbf{k}_\lambda\sum_{\text{pol}}=\frac{1}{(2\pi)^3}\int\int d\Omega_\lambda k_\lambda^2 dk_\lambda\sum_{\text{pol}}, \tag{A16}$$

and with

$$\sum_{\text{pol}}\mathbf{e}_\lambda\left[\mathbf{e}_\lambda\cdot\hat{\mathbf{d}}_B(t')\right]=-\hat{\mathbf{k}}_\lambda\times\left[\hat{\mathbf{k}}_\lambda\times\hat{\mathbf{d}}_B(t')\right]$$
$$=-\frac{1}{k_\lambda^2}\mathbf{k}_\lambda\times\left[\mathbf{k}_\lambda\times\hat{\mathbf{d}}_B(t')\right], \tag{A17}$$

we then have

$$\sum_{\text{pol}}\mathbf{e}_\lambda\left[\mathbf{e}_\lambda\cdot\hat{\mathbf{d}}_B(t')\right]e^{i\mathbf{k}_\lambda\cdot(\mathbf{R}_1-\mathbf{R}_B)}$$
$$=\frac{1}{k_\lambda^2}\nabla_1\times\left[\nabla_1\times e^{i\mathbf{k}_\lambda\cdot(\mathbf{R}_1-\mathbf{R}_B)}\hat{\mathbf{d}}_B(t')\right]. \tag{A18}$$

Hence we obtain

$$C=\sum_{AB}\nabla_2\times\left[\nabla_2\times\frac{1}{4\pi\epsilon_0 R_{A2}}\left(\frac{-i}{2\epsilon_0}\right)\int_0^{t_2-\tau_{A2}}dt'\frac{1}{(2\pi)^3}\right.$$
$$\times\int\int\int k_\lambda^2\sin\theta_\lambda\,d\theta_\lambda\,d\varphi_\lambda dk_\lambda$$
$$\times\frac{\omega_\lambda}{k_\lambda^2}e^{i\omega_\lambda(t'-t_1)}\nabla_1\times\{\nabla_1\times e^{i\mathbf{k}_\lambda\cdot(\mathbf{R}_1-\mathbf{R}_B)}$$
$$\left.\times[\hat{\mathbf{d}}_B(t'),\hat{\mathbf{d}}_A^-(t_2-\tau_{A2})]\theta(t_2-\tau_{A2})\}\right]. \tag{A19}$$

We choose the z axis for $\mathbf{k}_\lambda$ along $\mathbf{R}_1-\mathbf{R}_B$. The ϕ_λ integral yields 2π. The θ_λ integral can be obtained as

$$\int_0^\pi d\theta_\lambda\sin\theta_\lambda e^{ik_\lambda R_{B1}\sin\theta_\lambda}=\frac{2c\sin(\omega_\lambda\tau_{B1})}{\omega_\lambda R_{B1}}, \tag{A20}$$

where

$$R_{B1}=|\mathbf{R}_1-\mathbf{R}_B|,\ \tau_{B1}=R_{B1}/c. \tag{A21}$$

Also $dk_\lambda=d\omega_\lambda/c$ so that

$$C=\sum_{AB}\nabla_2\times\left[\nabla_2\times\frac{1}{4\pi\epsilon_0 R_{A2}}\left(\nabla_1\times\left\{\nabla_1\times\frac{1}{4\pi\epsilon_0 R_{B1}}\right.\right.\right.$$
$$\times\left[\left(\frac{-i}{\pi}\right)\int_0^{t_2-\tau_{A2}}dt'\right.$$
$$\times\int_0^\infty d\omega_\lambda e^{i\omega_\lambda(t'-t_1)}\sin(\omega_\lambda\tau_{B1})$$
$$\left.\left.\left.\times[\hat{\mathbf{d}}_B(t'),\hat{\mathbf{d}}_A^-(t_2-\tau_{A2})]\right]\theta(t_2-\tau_{A2})\right\}\right)\right]. \tag{A22}$$

Designating the Laplace transform of the dipole operator $\hat{\mathbf{d}}(t)$ as $\hat{\mathbf{D}}(s)$ and using the inverse Laplace transform formula

$$\hat{\mathbf{d}}_B(t')=\frac{1}{2\pi i}\int_c ds\,e^{st'}\hat{\mathbf{D}}_B(s), \tag{A23}$$

the quantity in the inner large square brackets in (A22) is

$$I=\left(\frac{-i}{\pi}\right)\int_0^{t_2-\tau_{A2}}dt'\int_0^\infty d\omega_\lambda e^{i\omega_\lambda(t'-t_1)}$$
$$\times\sin(\omega_\lambda\tau_{B1})\,\hat{\mathbf{d}}_B(t')$$
$$=\left(\frac{-i}{\pi}\right)\frac{1}{2\pi i}\int_c ds\int_0^{t_2-\tau_{A2}}dt'\int_0^\infty d\omega_\lambda e^{i\omega_\lambda(t'-t_1)}$$
$$\times\sin(\omega_\lambda\tau_{B1})\,\hat{\mathbf{D}}_B(s)\,e^{st'}. \tag{A24}$$

Performing the t' integration we find

$$I=\frac{-1}{2\pi}\frac{1}{2\pi i}\int_c ds\int_0^\infty d\omega_\lambda\,\hat{\mathbf{D}}_B(s)\left\{\frac{e^{i\omega_\lambda(t_2-\tau_{A2}-t_1+\tau_{B1})}}{(s+i\omega_\lambda)}\right.$$
$$\times e^{s(t_2-\tau_{A2})}-\frac{e^{-i\omega_\lambda(t_1-\tau_{B1})}}{(s+i\omega_\lambda)}-\frac{e^{i\omega_\lambda(t_2-\tau_{A2}-t_1-\tau_{B1})}}{(s+i\omega_\lambda)}$$
$$\left.\times e^{s(t_2-\tau_{A2})}+\frac{e^{-i\omega_\lambda(t_1+\tau_{B1})}}{(s+i\omega_\lambda)}\right\}. \tag{A25}$$

Now

$$\hat{\mathbf{D}}_B(s)=\hat{\mathbf{D}}_B^+(s)+\hat{\mathbf{D}}_B^-(s) \tag{A26}$$

and $\hat{\mathbf{D}}_B^+(s)$ is only large near $s=+i\omega_0$ whereas $\hat{\mathbf{D}}_B^-(s)$ is only large near $s=-i\omega_0$. Due to the $(s+i\omega_\lambda)$ term the ω_λ integral is large near $\omega_\lambda=+is$, so for the $\hat{\mathbf{D}}_B^+(s)$ term this is near $\omega_\lambda=-\omega_0$, which is outside the range $0-\infty$ for the values of ω_λ. On the other hand, the $\hat{\mathbf{D}}_B^-(s)$ term gives significant contributions near $\omega_\lambda=+\omega_0$, which is inside the range. Assuming that ω_0 is large compared to the decay rate, the $\hat{\mathbf{D}}_B^+(s)$ contribution can be ignored and

$$I=\left(\frac{-1}{2\pi}\right)\frac{1}{2\pi i}\int_c ds\int_0^\infty d\omega_\lambda\hat{\mathbf{D}}_B^-(s)\{\cdots\}, \tag{A27}$$

where $\{\cdots\}$ is the term in large curly brackets in Eq. (A25). With $\hat{\mathbf{D}}_B^-(s)$ only being significant for s near $-i\omega_0$ and the ω_λ integral only large near $\omega_\lambda=\omega_0$ we can then arbitrarily extend the ω_λ integral to $-\infty$ with little error to give

$$I=\left(\frac{i}{2\pi}\right)\frac{1}{2\pi i}\int_c ds\int_{-\infty}^\infty d\omega_\lambda\hat{\mathbf{D}}_B^-(s)\left(\frac{e^{i\omega_\lambda(t_2-\tau_{A2}-t_1+\tau_{B1})}}{(\omega_\lambda-is)}e^{s(t_2-\tau_{A2})}-\frac{e^{-i\omega_\lambda(t_1-\tau_{B1})}}{(\omega_\lambda-is)}-\frac{e^{i\omega_\lambda(t_2-\tau_{A2}-t_1-\tau_{B1})}}{(\omega_\lambda-is)}\right.$$
$$\left.\times e^{s(t_2-\tau_{A2})}+\frac{e^{-i\omega_\lambda(t_1+\tau_{B1})}}{(\omega_\lambda-is)}\right). \tag{A28}$$

The ω_λ integrals are evaluated using contour integration. For all terms in (A28) the pole is at $\omega_\lambda=is=i\in -y$ in the upper half plane and we find that

$$I=\frac{i}{2\pi}\int_c ds\,\hat{\mathbf{D}}_B^-(s)\{\theta(t_2-\tau_{A2}-t_1+\tau_{B1})e^{-s(t_2-\tau_{A2}-t_1+\tau_{B1})}e^{s(t_2-\tau_{A2})}-\theta(\tau_{B1}-t_1)e^{s(t_1-\tau_{B1})}$$
$$-\theta(t_2-\tau_{A2}-t_1-\tau_{B1})e^{-s(t_2-\tau_{A2}-t_1-\tau_{B1})}e^{s(t_2-t_{A2})}+\theta(-\tau_{B1}-t_1)e^{s(t_1+\tau_{B1})}\}. \tag{A29}$$

From the inverse Laplace transforms

$$I=-\{\hat{\mathbf{d}}_B^-(t_1-\tau_{B1})\theta((t_2-\tau_{A2})-(t_1-\tau_{B1}))\theta(t_1-\tau_{B1})-\hat{\mathbf{d}}_B^-(t_1-\tau_{B1})\theta(\tau_{B1}-t_1)\theta(t_1-\tau_{B1})$$
$$-\hat{\mathbf{d}}_B^-(t_1+\tau_{B1})\theta((t_2-\tau_{A2})-(t_1+\tau_{B1}))\theta(t_1+\tau_{B1})+\hat{\mathbf{d}}_B^-(t_1+\tau_{B1})\theta(-\tau_{B1}-t_1)\theta(t_1+\tau_{B1})\}. \tag{A30}$$

The second and fourth terms are zero.

Finally from (A22) the commutator is

$$C=\left[\hat{\mathbf{E}}_F^+(\mathbf{R}_1,t_1),\hat{\mathbf{E}}_S^+(\mathbf{R}_2,t_2)\right]$$
$$=-\sum_{AB}\nabla_2\times\left\{\nabla_2\times\frac{1}{4\pi\epsilon_0 R_{A2}}\left[\nabla_1\times\left(\nabla_1\times\frac{1}{4\pi\epsilon_o R_{B1}}\hat{K}\right)\right]\right\}, \tag{A31}$$

where

$$\hat{K}=[\hat{\mathbf{d}}_B^-(t_1-\tau_{B1}),\hat{\mathbf{d}}_A^-(t_2-\tau_{A2})]\theta(t_1-\tau_{B1})\theta(t_2-\tau_{A2})\theta((t_2-\tau_{A2})-(t_1-\tau_{B1}))$$
$$-[\hat{\mathbf{d}}_B^-(t_1+\tau_{B1}),\hat{\mathbf{d}}_A^-(t_2-\tau_{A2})]\theta(t_1+\tau_{B1})\theta(t_2-\tau_{A2})\theta((t_2-\tau_{A2})-(t_1+\tau_{B1})). \tag{A32}$$

The result may be compared to that given by Cresser [20] for the case of a single atom at $\mathbf{R}=\mathbf{0}$.

APPENDIX B: GENERAL PROPERTIES OF DENSITY OPERATORS

The density operator $\hat{\rho}$ for any quantum system can also be defined by its matrix elements ρ_{ij} for a suitable orthogonal basis $\langle i\mid j\rangle=\delta_{ij}$, where $\rho_{ij}=\langle i\mid\hat{\rho}\mid j\rangle$. The three basic conditions that the density operator must satisfy are that (a) $\hat{\rho}$ is Hermitean

$$\hat{\rho}=\hat{\rho}^\dagger,\quad \rho_{ij}=\rho_{ji}^*, \tag{B1}$$

(b) $\hat{\rho}$ has unit trace

$$\mathrm{Tr}\hat{\rho}=1,\qquad \sum_i\hat{\rho}_{ii}=1, \tag{B2}$$

and (c) $\hat{\rho}$ is positive. Thus for any state $|\psi\rangle$

$$\langle\psi\mid\hat{\rho}\mid\psi\rangle\geq 0. \tag{B3}$$

It is useful to consider the eigenvalues λ_i and normalized eigenvectors $|\psi_i\rangle$ of $\hat{\rho}$

$$\hat{\rho}\,|\lambda_i\rangle=\lambda_i\,|\lambda_i\rangle, \tag{B4}$$

$$\hat{\rho}=\sum_i\lambda_i\,|\lambda_i\rangle\langle\lambda_i|. \tag{B5}$$

The density matrix elements can be written in terms of parameters P_i, s_{ij}, d_{ij} as

$$[\rho] = \begin{pmatrix} P_1 & \frac{1}{2}(s_{12}-id_{12}) & \frac{1}{2}(s_{13}-id_{13}) & \cdots & \cdots \\ \frac{1}{2}(s_{12}+id_{12}) & P_2 & \frac{1}{2}(s_{23}-id_{23}) & \cdots & \cdots \\ \frac{1}{2}(s_{13}+id_{13}) & \frac{1}{2}(s_{23}+id_{23}) & P_3 & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots & \cdots \end{pmatrix}. \tag{B6}$$

From the three basic conditions it is then easy to show that necessary and sufficient conditions for (B1)–(B3) to hold are as follows [the conditions (i) and (ii) are alternative]: (a)

$$\text{(i) } \lambda_i \text{ are real,} \tag{B7}$$

$$\text{(ii) } P_i\text{(populations), } S_{ij} \text{ and } d_{ij}\text{(coherences) are real;} \tag{B8}$$

(b)

$$\text{(i) } \sum_i \lambda_i = 1, \tag{B9}$$

$$\text{(ii) } \sum_i P_i = 1; \tag{B10}$$

(c)

$$\text{(i) } \lambda_i \text{ are positive,} \tag{B11}$$

(ii) the determinant of $[\rho]$ and all principle minors are positive

$$|\rho| \geq 0, \quad \begin{vmatrix} P_1 & \frac{1}{2}(s_{12}-id_{12}) \\ \frac{1}{2}(s_{12}+id_{12}) & P_2 \end{vmatrix} \geq 0 \cdots . \tag{B12}$$

Other results also follow: (d)

$$\text{(i) } 1 \geq \lambda_i \geq 0, \tag{B13}$$

$$\text{(ii) } \mathrm{Tr}\hat{\rho}^2 = \sum_i \lambda_i^2 \leq 1, \tag{B14}$$

$$\text{(iii) } P_1^2 + P_2^2 + \cdots + \frac{1}{2}\left(s_{12}^2 + d_{12}^2 + s_{13}^2 + d_{13}^2 + \cdots\right) \leq 1, \tag{B15}$$

(e)

$$\langle \psi \mid \hat{\rho} \mid \psi \rangle \leq 1 \qquad (|\psi\rangle \text{ normalized to unity}). \tag{B16}$$

One possible situation is to have all the eigenvalues of $\hat{\rho}$ equal to zero, except one which is unity. In this case the system is in a *pure state* such that

$$\hat{\rho} = |\lambda\rangle\langle\lambda|, \tag{B17}$$

$$\hat{\rho} = \hat{\rho}^2, \tag{B18}$$

and

$$\mathrm{Tr}\hat{\rho}^2 = 1. \tag{B19}$$

If we deal with a system consisting of two parts we often wish to describe one subsystem S by a reduced density operator $\hat{\rho}$. Assuming that $\hat{W}$ is the density operator for the combined system, with orthogonal states as products $|A\rangle|i\rangle$ (where $|A\rangle$ are the orthogonal states $\langle A \mid B\rangle = \delta_{AB}$ for the other subsystem R), the reduced density operator is given by

$$\hat{\rho} = \mathrm{Tr}_R \hat{W}, \tag{B20}$$

or in matrix elements

$$\rho_{ij} = \sum_A W_{Ai;Aj}. \tag{B21}$$

It is easily shown that if $\hat{W}$ is a density operator satisfying (B1)–(B3), then $\hat{\rho}$ is also a density operator satisfying these (and all the other) conditions.

Often we wish to change to another orthogonal basis set via

$$|\alpha\rangle = \sum_i C_{i\alpha}|i\rangle. \tag{B22}$$

In terms of the new basis states the density matrix elements are given as

$$\rho'_{\alpha\beta} = \langle\alpha|\hat{\rho}|\beta\rangle, \tag{B23}$$

$$\rho'_{\alpha\beta} = \sum_{ij} C^*_{i\alpha}\rho_{ij}C_{j\beta}. \tag{B24}$$

APPENDIX C: DETAILS OF OPTIMIZED SQUEEZING CALCULATIONS

1. Case of Λ and V configurations

Parametrizing the atomic density matrix in terms of new variables $P'_1, P'_2, P'_3, S'_1, ..., S'_6$ as in Eq. (3.15), the constraints take the same form as (3.16)–(3.19). The

normally ordered variances (3.28) and (3.31) now only involve P_1' or P_2', S_1', S_2' so that we may set the other parameters to zero

$$P_3' = 0, \tag{C1}$$

$$S_3' = S_4' = S_5' = S_6' = 0 \tag{C2}$$

in order to determine the situation of optimum squeezing.

Using (3.33) and (3.34) the equations of constraint reduce to

$$\mathrm{Tr}\hat{\rho} = P_1' + P_2' = G = 1, \tag{C3}$$

$$\mathrm{Tr}\hat{\rho}^2 = P_1'^2 + P_2'^2 + \frac{1}{2}\left(S_1'^2 + S_2'^2\right) = H \leq 1. \tag{C4}$$

The third equation of constraint $|\rho| \geq 0$ involves a left hand side which is identically zero.

For the two cases we have, for the parameters α,β,γ of (2.78), (2.79) and (2.81),

$$\alpha = -\rho_{21}'^2, \tag{C5}$$

$$\beta = 2\left(\rho_{22}' - \rho_{21}'\rho_{12}'\right), \tag{C6}$$

$$\gamma = \rho_{11}' - \rho_{22}' \tag{C7}$$

for the Λ configuration and

$$\alpha = -\rho_{12}'^2, \tag{C8}$$

$$\beta = 2\left(\rho_{11}' - \rho_{12}'\rho_{21}'\right), \tag{C9}$$

$$\gamma = \rho_{22}' - \rho_{11}', \tag{C10}$$

for the V configuration, so that after optimizing the choice of ϕ, ω, t we have from (2.82)

$$f = \left(\frac{\left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle}{K^2}\right)_{\mathrm{opt}\,\phi} = 2P_2' - \left(S_1'^2 + S_2'^2\right), \tag{C11}$$

$$f = \left(\frac{\left\langle : \Delta\hat{E}_{\phi S}^2 : \right\rangle}{K^2}\right)_{\mathrm{opt}\,\phi} = 2P_1' - \left(S_1'^2 + S_2'^2\right), \tag{C12}$$

for the Λ and V configurations, respectively.

The Lagrange method of undetermined multipliers involves consideration of the function $F = f - \ell G - mH$, where ℓ, m are undetermined multipliers. For the Λ case the method gives

$$\frac{\partial F}{\partial P_1'} = -\ell - m2P_1' = 0, \tag{C13}$$

$$\frac{\partial F}{\partial P_2'} = 2 - \ell - m2P_2' = 0, \tag{C14}$$

$$\frac{\partial F}{\partial S_1'} = -2S_1' - mS_1' = 0, \tag{C15}$$

$$\frac{\partial F}{\partial S_2'} = -2S_2' - mS_2' = 0. \tag{C16}$$

Solving for ℓ from (C13) and substituting into (C14) we get

$$0 = 1 + m(P_1' - P_2'), \tag{C17}$$

while (C14) and (C16) give

$$S_1' = S_2' = 0 \tag{C18}$$

and/or

$$m = -2. \tag{C19}$$

The solution (C18) cannot lead to squeezing as (C11) shows f to be positive. The solution (C19) together with (C17), (C3), and (C4) gives

$$P_1' = \frac{3}{4}, \qquad P_2' = \frac{1}{4}, \tag{C20}$$

$$S_1'^2 + S_2'^2 = 2H - \frac{5}{4}, \tag{C21}$$

$$f = \frac{7}{4} - 2H. \tag{C22}$$

The smallest value for f corresponds to choosing the largest value possible for H which is unity, corresponding to a pure state.

The quantities α, β, and γ can be obtained from (C5)–(C7) in the Λ case.

The details of the calculation for the V configuration are similar. The quantities α, β, and γ are obtained from (C8)–(C10).

2. Case of ladder configuration

For the *ladder* case

$$\alpha = sc\rho_{31} - \left(c\rho_{32} + s\rho_{21}\right)^2, \tag{C23}$$

$$\beta = 2\left[c^2\rho_{33} + s^2\rho_{22} - \left(c\rho_{32} + s\rho_{21}\right)\left(c\rho_{23} + s\rho_{12}\right)\right], \tag{C24}$$

$$\gamma = \left(c^2 - s^2\right)\rho_{22} - c^2\rho_{33} + s^2\rho_{11}. \tag{C25}$$

The minimization was carried out numerically using a sequential quadrature programming algorithm [28] in which the search direction involves the solution of a quadratic equation. The method first determines a point that satisfies the constraints. From that point an interactive search for the minimum point is made. Each iteration step includes (a) the solution of a quadratic equa-

tion involving derivatives of the function to be minimized and (b) a line search with an augmented Lagrangian merit function involving the equations of constraint and a quasi-Newton update of the approximate Hessian of the function; the Hessian is zero at the minimum point. Calculations were restricted to the special case where both transitions contribute equally $c = s = 1/\sqrt{2}$. The numerical approach first established that the minimum was for zero values of P_2, S_1, S_2, S_3, and S_4. The values for P_1, P_3, S_5, and S_6 are then established analytically using Lagrangian undetermined multipliers. The values of α,β,γ are calculated from (C23)–(C25).

[1] (a) Special Issue, J. Mod. Opt. **34** (6/7) 1987; (b) Special Issue, J. Opt. Soc. Am. B **4** (10) 1987; (c) K. Zaheer and M. S. Zubairy, Adv. At. Mol. Phys. **28**, 142 (1991).

[2] H. Takahashi, Adv. Commun. Syst. **1**, 227 (1965); G. J. Milburn and D. F. Walls, Opt. Commun. **39**, 401 (1981).

[3] L. A. Lugiato, G. Strini, and F. De Martini, Opt. Lett. **8**, 256 (1983); P. D. Drummond, K. J. McNeil, and D. F. Walls, Opt. Acta **28**, 211 (1981).

[4] H. P. Yuen and J. H. Shapiro, Opt. Lett. **4**, 434 (1979); G. J. Milburn, D. F. Walls, and M. D. Levenson, J. Opt. Soc. Am. B **1**, 390 (1984).

[5] M. O. Scully, Phys. Rev. Lett. **55**, 2808 (1985); N. A. Ansari, J. Gea-Banacloche, and M. S. Zubairy, Phys. Rev. A **41**, 5179 (1990); M. O. Scully, K. Wodkiewicz, J. Bergou, N. Lu, J. Meyer Tervehn, and M. S. Zubairy, Phys. Rev. Lett. **60**, 1832 (1988); M. Orszag, J. Bergou, W. Schleich, and M. O. Scully, in *Squeezed and Nonclassical Light*, Vol. 190 of *NATO Advanced Study Institute, Series B: Physics,* edited by P. Tombesi and E. R. Pike (Plenum, New York, 1988), p. 287; B. J. Dalton, in *New Frontiers in Quantum Electrodynamics and Quantum Optics*, Vol. 232 of *NATO Advanced Study Institute, Series B: Physics*, edited by A. O. Barut (Plenum, New York, 1990), p. 167.

[6] B. J. Dalton, in *Quantum Optics IV*, edited by J. D. Harvey and D. F. Walls (Springer-Verlag, Berlin, 1986), p. 99; L. A. Lugiato, Phys. Rev. A **33**, 4079 (1986).

[7] M. D. Reid and D. F. Walls, Phys. Rev. A **32**, 392 (1985); M. Sargent, D. A. Holm, and M. S. Zubairy, *ibid.* **31**, 3112 (1985).

[8] (a) P. Meystre and M. S. Zubairy, Phys. Lett. **89A**, 390 (1982); (b) K. Wodkiewicz, P. L. Knight, S. J. Buckle, and S. M. Barnett, Phys. Rev. A **35**, 2567 (1987); (c) S. M. Barnett and M. A. Dupertius, J. Opt. Soc. Am. B **4**, 505 (1987); (d) A. Heidmann, J. M. Raimond, and S. Reynaud, Phys. Rev. Lett. **54**, 326 (1985); (e) A. Heidmann, J. M. Raimond, S. Reynaud, and N. Zagury, Opt. Commun. **54**, 189 (1985); (f) M. Butler and P. D. Drummond, Opt. Acta **33**, 1 (1986).

[9] G. S. Agarwal, in *Quantum Optics*, edited by G. Höhler Springer Tracts in Modern Physics Vol. 70 (Springer-Verlag, Berlin, 1974).

[10] D. F. Walls and P. Zoller, Phys. Rev. Lett. **47**, 709 (1981).

[11] (a) Z. Ficek, R. Tanas, and S. Kielich, Phys. Rev. A **29**, 2004 (1984); (b) J. Opt. Soc. Am. B **1**, 882 (1984).

[12] A. Heidmann and S. Reynaud, J. Phys. (Paris) **46**, 1937 (1985).

[13] (a) W. Vogel and R. Blatt, Phys. Rev. A **45**, 3319 (1992); (b) F. A. M. DeOliveira, B. J. Dalton, and P. L. Knight, J. Opt. Soc. Am. B **4**, 1558 (1987).

[14] (a) F. Diedrich and H. Walther, Phys. Rev. Lett. **58**, 203 (1987); (b) I. Siemers, M. Schubert, R. Blatt, W. Neuhauser, and P. E. Toschek, Europhys. Lett. **18**, 139 (1992).

[15] M. J. Collett and D. F. Walls, Phys. Rev. A **32**, 2887 (1985).

[16] S. M. Barnett and C. R. Gilson, Europhys. Lett. **12**, 325 (1990).

[17] E. S. Polzik, J. Carri, and H. J. Kimble, Phys. Rev. Lett. **68**, 3020 (1992).

[18] S. M. Barnett and P. L. Knight, Phys. Scr. **T21**, 5 (1988).

[19] H. J. Kimble and L. Mandel, Phys. Rev. A **13**, 2123 (1976); J. R. Ackerhalt and J. H. Eberly, Phys. Rev. D **10**, 3350 (1974).

[20] J. D. Cresser, Phys. Rep. A **94**, 47 (1983).

[21] B. Renaud, R. M. Whitley, and C. R. Stroud, Jr., J. Phys. B **9**, L19 (1976); B. R. Mollow, J. Phys. A **8**, L13 (1975).

[22] J. D. Cresser, Phys. Rev. A **29**, 1984 (1984).

[23] E. A. Power and S. Zienau, Philos. Trans. R. Soc. London Ser. A **251**, 427 (1959); W. P. Healy, J. Phys. A **10**, 279 (1977).

[24] S. M. Barnett, Opt. Commun. **61**, 432 (1987).

[25] C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 345 (1977).

[26] S. M. Barnett and D. P. Pegg, J. Phys. A **19**, 3849 (1986).

[27] B. J. Dalton and P. L. Knight, Phys. Rev. A **42**, 3034 (1990).

[28] P. E. Gill, W. Murray, and M. H. Wright, *Practical Optimization* (Academic, New York, 1981).

[29] V. Buzek, P. L. Knight, and J. K. Kudryavtsev, Phys. Rev. A **44**, 1931 (1991).