

CORRELATED SUPERPOSITION STATES IN TWO-ATOM SYSTEMS

ZBIGNIEW FICEK

*Department of Physics and Centre for Laser Science, The University of
Queensland, Brisbane, Australia*

RYSZARD TANAŚ

*Nonlinear Optics Division, Institute of Physics, Adam Mickiewicz
University, Poznań, Poland*

CONTENTS

- I. Introduction
- II. Master Equation of Two Coupled Atoms
 - A. Atomic System and Hamiltonian
 - B. Master Equation
- III. Collective Atomic States
 - A. Collective States of Two Identical Atoms
 - B. Collective States of Two Nonidentical Atoms
 - C. Maximally Entangled States of Two Nonidentical Atoms
- IV. Selective Excitation of the Collective Atomic States
 - A. Preparation of the Symmetric State by a Pulse Laser
 - B. Preparation of the Antisymmetric State
 - 1. Pulse Laser
 - 2. Indirect Driving through the Symmetric State
 - 3. Atom–Cavity–Field Interaction
 - C. Preparation of a Superposition of Antisymmetric and Ground States
- V. Detection of the Entangled States
 - A. Fluorescence Intensity
 - B. Interference Pattern
- VI. Two-Photon Entangled States

Modern Nonlinear Optics, Part 1, Second Edition, Advances in Chemical Physics, Volume 119,
Edited by Myron W. Evans. Series Editors I. Prigogine and Stuart A. Rice.
ISBN 0-471-38930-7 © 2001 John Wiley & Sons, Inc.

- A. Two Atoms in a Squeezed Vacuum
- B. Steady-State Populations
- C. Effect of the Antisymmetric State on the Purity of the System
- D. Two-Photon Entangled States for Two Nonidentical Atoms
- E. Mapping of the Entanglement of Light on Atoms

Acknowledgments

References

I. INTRODUCTION

The subject of correlated or collective spontaneous emission by a system of a large number of atoms was first proposed by Dicke [1], who introduced the concept of superradiance that the influence on each atomic dipole of the electromagnetic field produced by the other atomic dipoles could, in certain circumstances, cause each atom to decay with an enhanced spontaneous emission rate. The shortening of the atomic lifetime resulting from the interaction between N atoms could involve an enhancement of the intensity of radiation up to N^2 .

The earliest investigations into correlated spontaneous emission from multiatom systems were motivated by attempts to detect coherent effects in the interaction of light with resonant atomic systems [2–4]. Another intrinsic feature of correlated spontaneous emission is that the emitted field exhibits strong nonlinear and directional behavior [5]. Moreover, the interest in correlated spontaneous emission lies in its close connection with the quantum and classical as well as with the spontaneous and stimulated aspects of atomic emission [6].

The phenomenon of collective emission is, in general, characteristic of macroscopic systems with a large number of emitting atoms confined a region much smaller than the optical wavelength. However, to understand collective effects from a macroscopic system of atoms, it is necessary to have a microscopic formulation of the interaction between the atoms and the electromagnetic field. Therefore, some previous work has been devoted to study collective effects in the case of few atoms [7–10]. Although a system of two or three atoms is admittedly an elementary model, it offers some advantages over the multiatom problem. Because of its simplicity, one obtains detailed and almost exact dynamical solutions with a variety of initial conditions. Many of the results predicted for the system of two or three atoms are analogous to phenomena that one could expect in multiatom systems. Early treatments of two or three-atom systems assumed a constant interatomic separation during the radiation process. When averaged over all such possible interatomic separations the collective effects average out, which made them difficult to observe experimentally.

In the 1990s, advances in trapping and cooling of small number of ions and neutral atoms greatly renewed the interest in collective effects in the interaction

of atoms with the electromagnetic field [11–13]. The trapped atoms are essentially motionless and lie at a known and controllable distance from one another, permitting qualitatively new studies of interatomic interactions not accessible in a gas cell or an atomic beam. The advantage of the trapped atoms is that it allows separation of collective effects, arising from the correlations between the atoms, from the single-atom effects. The question of to what extent the interatomic interactions can alter the dynamics of a multiatom system has become of interest as it contains information about the internal structure of the collective system.

A central topic in the current studies of collective effects is the theoretical and experimental investigation of the concept of correlated superposition states (entangled states) of a multiatom system [14]. The entangled states are linear superpositions of the internal states of the system that cannot be separated into product states of the individual atoms. This property is recognized as an entirely quantum-mechanical effect and has played a crucial role in many discussions of the nature of quantum measurements and, in particular, in the development of quantum communications. It has been realized that entangled states can have many practical applications, ranging from quantum computation [15,16], information processing [17,18], and cryptography [19] to atomic spectroscopy [20].

An example of entangled states in a two-atom system are the symmetric and antisymmetric states, which correspond to the symmetric and antisymmetric combinations of the atomic dipole moments, respectively [1,7,21]. These states are created by the dipole–dipole interaction between the atoms and are characterized by different spontaneous decay rates that the symmetric state decays with an enhanced, whereas the antisymmetric state decays with a reduced spontaneous emission rate [7]. For the case of two atoms confined into the region much smaller than the optical wavelength, the antisymmetric state does not decay at all, and therefore can be regarded as a decoherence-free state.

Another particularly interesting entangled states of the two-atom system are two-photon entangled states that are superpositions of only those states of the two-atom system in which both or neither of the atoms are excited. These states have been known for a long time as pairwise atomic states [22] or multiatom squeezed states [23]. The two-photon entangled states cannot be generated by a coherent laser field coupled to the atomic dipole moments. The states can be created by a two-photon excitation process with nonclassical correlations that can transfer the population from the two-atom ground state to the upper state without populating the intermediate one-photon states. An obvious candidate for the creation of the two-photon entangled states is a broadband squeezed vacuum field that is characterized by strong nonclassical two-photon correlations [24,25].

A number of theoretical methods have been proposed to prepare a two-atom system in an entangled state [26–29,31–34], and two-atom entangled states have

already been demonstrated experimentally using ultracold trap ions [35] and cavity quantum electrodynamic (QED) schemes [36]. The preparation of correlated superposition states in multiatom system has been performed using a quantum nondemolition (QND) measurement technique [37]. A mapping of entangled states of light on atoms has also been proposed [38,39] and experimentally demonstrated [40].

In this chapter, we review schemes proposed for the preparation of two 2-level atoms in an entangled state. Since we focus here on basis aspects of the atom-atom entanglement, we begin in Section II with a derivation of the master equation for two nonidentical two-level atoms interacting with the quantized three-dimensional vacuum field and driven by a single-mode coherent laser field. Sections III and IV are concerned mainly with techniques proposed for the preparation of a two-atom system in entangled states. The cases of maximally and nonmaximally entangled states are discussed. In Section V, we discuss methods of detecting of a particular entangled state. In Section VI, we describe the method of preparation of a two-atom system in two-photon entangled states. We also present a method of mapping of the entanglement of light on atoms.

II. MASTER EQUATION OF TWO COUPLED ATOMS

There are several theoretical approaches that can be used to calculate the dynamics and correlation properties of two atoms interacting with the quantized electromagnetic field. One of the methods is the wavefunction approach in which the dynamics are given in terms of the probability amplitudes [9]. Another approach is the Heisenberg equation method, in which equations of motion for the atomic and field operators are found from the Hamiltonian of a given system [10]. The most popular approach is the master equation method, in which the equation of motion is found for the density operator of an atomic system weakly coupled to a system regarded as a reservoir [7,8,41]. There are many possible realizations of reservoirs. The typical reservoir to which atomic systems are coupled is the quantized three-dimensional multimode vacuum field. The major advantage of the master equation is that it allows us to consider the evolution of the atoms plus field system entirely in terms of atomic operators.

A. Atomic System and Hamiltonian

We consider a system of two nonidentical and nonoverlapping atoms at positions \mathbf{r}_1 and \mathbf{r}_2 , coupled to the quantized three-dimensional electromagnetic field. The initial state of the field is the product of a single-mode coherent state of a driving laser field, and the vacuum state of the rest of the modes. Each atom is assumed to have only two levels: the ground level $|g_i\rangle$ and the excited level $|e_i\rangle$ ($i = 1, 2$), separated by an energy $\hbar\omega_i = E_{e_i} - E_{g_i}$, and connected by an

electric dipole transition with the dipole matrix elements μ_1 and μ_2 . The dipole transitions are represented by the dipole raising S_i^+ and lowering S_i^- operators defined as

$$S_i^+ = |e_i\rangle\langle g_i|, \quad S_i^- = |g_i\rangle\langle e_i| \quad (1)$$

and satisfying the relations

$$[S_i^+, S_i^-] = 2S_i^z \delta_{ij}, \quad S_i^+ S_i^- + S_i^- S_i^+ = 1 \quad (2)$$

where $S_i^z = \frac{1}{2}(|e_i\rangle\langle e_i| - |g_i\rangle\langle g_i|)$ is the energy operator of the i th atom. If the i th atom is in its ground state $|g_i\rangle$, then $\langle S_i^z \rangle = -\frac{1}{2}$, whereas $\langle S_i^z \rangle = \frac{1}{2}$ if the atom is in its excited state.

The atoms interact with the quantized three-dimensional vacuum field and are also driven by a single-mode coherent laser field. We express the quantized multimode field in terms of the annihilation and creation operators $\hat{a}_{\mathbf{k}s}$ and $\hat{a}_{\mathbf{k}s}^\dagger$ of field mode $\mathbf{k}s$, which has wavevector \mathbf{k} , frequency ω_k , and polarization $\bar{\mathbf{e}}_{\mathbf{k}s}$. Thus, we write the electric field operator at position \mathbf{r} in the form

$$\hat{\mathbf{E}}(\mathbf{r}, t) = i\hbar \sum_{\mathbf{k}s} \left(\frac{\omega_k}{2\epsilon_0 \hbar V} \right)^{1/2} \bar{\mathbf{e}}_{\mathbf{k}s} (\hat{a}_{\mathbf{k}s} e^{i\mathbf{k}\cdot\mathbf{r}} - \hat{a}_{\mathbf{k}s}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}}) \quad (3)$$

where V is the normalization volume.

The total Hamiltonian of the interacting systems in the electric dipole and RWA approximations [42] is given by

$$H = H_A + H_F + H_I \quad (4)$$

where

$$H_A = \hbar\omega_1 S_1^z + \hbar\omega_2 S_2^z \quad (5)$$

is the Hamiltonian of the atoms

$$H_F = \sum_{\mathbf{k}s} \hbar\omega_k \left(\hat{a}_{\mathbf{k}s}^\dagger \hat{a}_{\mathbf{k}s} + \frac{1}{2} \right) \quad (6)$$

is the Hamiltonian of the field, and $H_I = H_v + H_L$ is the interaction Hamiltonian composed of two terms:

$$\begin{aligned} H_v &= -i\hbar \sum_{\mathbf{k}s} \{ [\mu_1 \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_1) S_1^+ + \mu_2 \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_2) S_2^+] \hat{a}_{\mathbf{k}s} e^{i\omega_k t} - \text{H.c.} \} \\ H_L &= -\frac{1}{2} i\hbar [(\Omega_1 S_1^+ + \Omega_2 S_2^+) e^{i(\omega_L t + \Phi_L)} - \text{H.c.}] \end{aligned} \quad (7)$$

The first term in Eq. (7) represents the interaction of the atoms with the quantized multimode vacuum field, while the second term is the interaction of the atoms with a classical driving laser field (H.c. denotes Hermitian conjugation). Here, ω_L and ϕ_L are the frequency and the phase of the driving field, respectively

$$\mathbf{g}_{\mathbf{k}s}(\mathbf{r}_i) = \left(\frac{\omega_k}{2\epsilon_0 \hbar V} \right)^{1/2} \bar{\mathbf{e}}_{\mathbf{k}s} e^{i\mathbf{k} \cdot \mathbf{r}_i} \quad (8)$$

is the mode function of the three-dimensional vacuum field, evaluated at the position \mathbf{r}_i of the i th atom, and

$$\Omega_i = \frac{\boldsymbol{\mu}_i \cdot \mathbf{E}_L e^{i\mathbf{k}_L \cdot \mathbf{r}_i}}{\hbar} \quad (9)$$

is the Rabi frequency of the i th atom with \mathbf{E}_L and \mathbf{k}_L denoting the amplitude and the wave vector of the driving field, respectively.

If the dipole moments of the atoms are parallel, the Rabi frequencies Ω_1 and Ω_2 are related by

$$\Omega_2 = \Omega_1 \frac{|\boldsymbol{\mu}_2|}{|\boldsymbol{\mu}_1|} e^{i\mathbf{k}_L \cdot \mathbf{r}_{12}} \quad (10)$$

where $\mathbf{r}_{12} = \mathbf{r}_2 - \mathbf{r}_1$ is the vector in the direction of the interatomic axis and $|\mathbf{r}_{12}| = r_{12}$ is the distance between the atoms. Thus, for two atoms with equal magnitudes of the dipole moments ($|\boldsymbol{\mu}_1| = |\boldsymbol{\mu}_2|$), the Rabi frequencies differ only by the phase factor $\exp(i\mathbf{k}_L \cdot \mathbf{r}_{12})$ arising from different positions of the atoms. However, the phase factor $\exp(i\mathbf{k}_L \cdot \mathbf{r}_{12})$ also depends on the orientation of the interatomic axis in respect to the direction of propagation of the driving field, and $\exp(i\mathbf{k}_L \cdot \mathbf{r}_{12})$ can be equal to one even for large interatomic separations r_{12} . This happens when the direction of propagation of the driving field is perpendicular to the interatomic axis, $\mathbf{k}_L \cdot \mathbf{r}_{12} = 0$. When $\mathbf{k}_L \cdot \mathbf{r}_{12} \neq 0$, the atoms are subject to different Rabi frequencies ($\Omega_1 \neq \Omega_2$).

B. Master Equation

Starting from the Hamiltonian (4), we can write the Schrödinger equation for the density operator ρ_T of the total system, two atoms plus the electromagnetic fields, as

$$\frac{\partial}{\partial t} \rho_T = \frac{1}{i\hbar} [H, \rho_T] \quad (11)$$

We are interested in the interaction of two atoms with the vacuum field, and therefore we transform Eq. (11) into the interaction picture with

$$\tilde{\rho}_T(t) = e^{i(H_A + H_F + H_L)t/\hbar} \rho_T e^{-i(H_A + H_F + H_L)t/\hbar} \quad (12)$$

and find that the transformed density operator satisfies the equation

$$\frac{\partial}{\partial t} \tilde{\rho}_T(t) = \frac{1}{i\hbar} [\tilde{H}_v(t), \tilde{\rho}_T(t)] \quad (13)$$

where

$$\tilde{H}_v(t) = e^{i(H_A+H_F+H_L)t/\hbar} H_v e^{-i(H_A+H_F+H_L)t/\hbar} \quad (14)$$

The master equation involves the so-called reduced density operator ρ describing the system of two atoms, which is obtained from the total density operator ρ_T by tracing over vacuum field (reservoir) states

$$\tilde{\rho}(t) = \text{Tr}_F \tilde{\rho}_T(t) \quad (15)$$

We will assume that the interaction is turned on at $t = 0$, and no correlations exist between the atoms and the vacuum field at this initial time. Hence, we can write the density operator $\tilde{\rho}_T(0)$ as a product of the density operator of the atoms $\tilde{\rho}(0)$ and the density operator of the reservoir $\tilde{\rho}_F(0)$:

$$\tilde{\rho}_T(0) = \tilde{\rho}(0) \tilde{\rho}_F(0) \quad (16)$$

The properties of the vacuum field are specified by the density operator $\tilde{\rho}_F(0)$, from which correlation functions of the field operators can be determined as

$$\begin{aligned} \langle \hat{a}_{\mathbf{k}s} \rangle &= \text{Tr}_F[\rho_F(0) \hat{a}_{\mathbf{k}s}] = 0, & \langle \hat{a}_{\mathbf{k}s}^\dagger \rangle &= \text{Tr}_F[\rho_F(0) \hat{a}_{\mathbf{k}s}^\dagger] = 0 \\ \langle \hat{a}_{\mathbf{k}s} \hat{a}_{\mathbf{k}'s'}^\dagger \rangle &= \text{Tr}_F[\rho_F(0) \hat{a}_{\mathbf{k}s} \hat{a}_{\mathbf{k}'s'}^\dagger] = \delta^3(\mathbf{k} - \mathbf{k}') \delta_{ss'} \\ \langle \hat{a}_{\mathbf{k}s}^\dagger \hat{a}_{\mathbf{k}'s'} \rangle &= \text{Tr}_F[\rho_F(0) \hat{a}_{\mathbf{k}s}^\dagger \hat{a}_{\mathbf{k}'s'}] = 0 \\ \langle \hat{a}_{\mathbf{k}s}^\dagger \hat{a}_{\mathbf{k}'s'}^\dagger \rangle &= \text{Tr}_F[\rho_F(0) \hat{a}_{\mathbf{k}s}^\dagger \hat{a}_{\mathbf{k}'s'}^\dagger] = 0 \\ \langle \hat{a}_{\mathbf{k}s} \hat{a}_{\mathbf{k}'s'} \rangle &= \text{Tr}_F[\rho_F(0) \hat{a}_{\mathbf{k}s} \hat{a}_{\mathbf{k}'s'}] = 0 \end{aligned} \quad (17)$$

We now integrate Eq. (13), substitute the solution for $\tilde{\rho}_T(t)$ inside the commutator on the right-hand side (r.h.s.) of Eq. (13), and after taking the trace over the reservoir states, we find that the reduced density operator $\tilde{\rho}(t)$ satisfies the integrodifferential equation

$$\begin{aligned} \frac{\partial}{\partial t} \tilde{\rho}(t) &= \frac{1}{i\hbar} \text{Tr}_F[\tilde{H}_v(t), \tilde{\rho}(0) \tilde{\rho}_F(0)] \\ &\quad - \frac{1}{\hbar^2} \int_0^t d\tau \text{Tr}_F\{[\tilde{H}_v(t), [\tilde{H}_v(t-\tau), \tilde{\rho}_T(t-\tau)]]\} \end{aligned} \quad (18)$$

In the derivation of Eq. (18), we have assumed that the total density operator $\tilde{\rho}_T(t)$ factorizes at $t = 0$. At later times correlations between the atoms and the field may arise as a result of the coupling through the Hamiltonian H_v . Here, we assume that the interaction between the atoms and the field is weak, which allows us to make the so-called Born approximation that ignores the back-reaction effects of the atoms on the field. Thus, $\tilde{\rho}_T(t - \tau) = \tilde{\rho}(t - \tau) \tilde{\rho}_F(t - \tau) = \tilde{\rho}(t - \tau) \tilde{\rho}_F(0)$ for all times $t - \tau > 0$. Moreover, we make the Markov approximation in which we assume that the correlation time of the field is much shorter than the timescale of radiation processes in the atoms. This allows us to replace $\tilde{\rho}(t - \tau)$ by $\tilde{\rho}(t)$.

Substituting Eq. (17) into Eq. (18) and after the Born–Markov approximation, the master equation takes the form

$$\begin{aligned} \frac{\partial}{\partial t} \tilde{\rho}(t) = & -\frac{i}{\hbar} [H_c, \tilde{\rho}(t)] \\ & - \frac{1}{2} \sum_{i,j=1}^2 \Gamma_{ij} (S_i^+ S_j^- \tilde{\rho}(t) + \tilde{\rho}(t) S_i^+ S_j^- - 2S_j^- \tilde{\rho}(t) S_i^+) \end{aligned} \quad (19)$$

where

$$H_c = \hbar \delta_1 S_1^+ S_1^- + \hbar \delta_2 S_2^+ S_2^- + \hbar \Omega_{12} (S_1^+ S_2^- + S_2^+ S_1^-) \quad (20)$$

represents the vacuum-induced shifts of the atomic transition frequencies and the coherent interaction between the atoms. The parameter

$$\Gamma_i \equiv \Gamma_{ii} = \pi \sum_{\mathbf{k}s} |\boldsymbol{\mu}_i \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_i)|^2 \delta^3(\mathbf{k} - \mathbf{k}_0) \quad (i = 1, 2) \quad (21)$$

describes the spontaneous emission rate of the i th atom resulting from the interaction of the individual atoms with the vacuum field, and

$$\Gamma_{ij} = \Gamma_{ji} = \pi \sum_{\mathbf{k}s} [\boldsymbol{\mu}_1 \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_1)] [\boldsymbol{\mu}_2^* \cdot \mathbf{g}_{\mathbf{k}s}^*(\mathbf{r}_2)] \delta^3(\mathbf{k} - \mathbf{k}_0), \quad (i \neq j) \quad (22)$$

are collective spontaneous emission rates arising from the coupling between the atoms through the vacuum field, and $k_0 = (k_1 + k_2)/2$.

The parameters

$$\delta_i = P \sum_{\mathbf{k}s} |\boldsymbol{\mu}_i \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_i)|^2 \left(\frac{1}{\omega_k + \omega_i} - \frac{1}{\omega_k - \omega_i} \right) \quad (23)$$

represent a part of the Lamb shift, induced by the first-order coupling in the Hamiltonian H_v , of the ground and excited states of the atoms, while

$$\Omega_{12} = P \sum_{\mathbf{k}s} [\boldsymbol{\mu}_1 \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_1)] [\boldsymbol{\mu}_2^* \cdot \mathbf{g}_{\mathbf{k}s}^*(\mathbf{r}_2)] \left(\frac{1}{\omega_k + \omega_0} + \frac{1}{\omega_k - \omega_0} \right) \quad (24)$$

represents the vacuum-induced coherent interaction between the atoms, P refers to the Cauchy principal value, and $\omega_0 = (\omega_1 + \omega_2)/2$ is the average frequency of the atomic transitions.

The parameters δ_i are usually considered to be absorbed into the atomic frequencies ω_1 and ω_2 , by redefining the frequencies $\tilde{\omega}_i = \omega_i + \delta_i$ and are not explicitly included in the master equations. However, we are interested in the qualitative effects of the interactions between the atoms, and the role played by Ω_{12} in their dynamics. It is evident from Eq. (20) that the parameter Ω_{12} does not appear as a shift of the energies, but rather as a coherent coupling between the atoms. Thus, the interaction with the vacuum field not only gives rise to the dissipative spontaneous emission but also leads to a coherent coupling between the atoms.

We may find the explicit form of the collective parameters Γ_{12} and Ω_{12} by using the spherical representation of the unit orthogonal polarization vectors [41]

$$\begin{aligned} \bar{\mathbf{e}}_{\mathbf{k}1} &= (-\cos \theta \cos \phi, -\cos \theta \sin \phi, \sin \theta), \\ \bar{\mathbf{e}}_{\mathbf{k}2} &= (\sin \phi, -\cos \phi, 0) \end{aligned} \quad (25)$$

and changing the sum over \mathbf{k} into an integral

$$\frac{1}{V} \sum_{\mathbf{k}s} \longrightarrow \frac{1}{(2\pi c)^3} \sum_{s=1}^2 \int_0^\infty \omega_k^2 d\omega_k \int_0^\pi \sin \theta d\theta \int_0^{2\pi} d\phi \quad (26)$$

where (k, θ, ϕ) denote spherical coordinates.

Substituting Eq. (26) into Eqs. (22) and (24), we obtain the following explicit expressions for the collective spontaneous emission rate

$$\begin{aligned} \Gamma_{12} = \frac{3}{4} \sqrt{\Gamma_1 \Gamma_2} \left\{ [1 - (\bar{\boldsymbol{\mu}} \cdot \bar{\mathbf{r}}_{12})^2] \frac{\sin(k_0 r_{12})}{k_0 r_{12}} \right. \\ \left. + [1 - 3(\bar{\boldsymbol{\mu}} \cdot \bar{\mathbf{r}}_{12})^2] \left[\frac{\cos(k_0 r_{12})}{(k_0 r_{12})^2} - \frac{\sin(k_0 r_{12})}{(k_0 r_{12})^3} \right] \right\} \end{aligned} \quad (27)$$

and the collective coupling between the atoms

$$\Omega_{12} = \frac{3}{4} \sqrt{\Gamma_1 \Gamma_2} \left\{ -[1 - (\bar{\boldsymbol{\mu}} \cdot \bar{\mathbf{r}}_{12})^2] \frac{\cos(k_0 r_{12})}{k_0 r_{12}} + [1 - 3(\bar{\boldsymbol{\mu}} \cdot \bar{\mathbf{r}}_{12})^2] \left[\frac{\sin(k_0 r_{12})}{(k_0 r_{12})^2} + \frac{\cos(k_0 r_{12})}{(k_0 r_{12})^3} \right] \right\} \quad (28)$$

where $\bar{\boldsymbol{\mu}}$ is the unit vector along the dipole moments of the atoms, which we have assumed to be parallel ($\bar{\boldsymbol{\mu}} = \bar{\boldsymbol{\mu}}_1 = \bar{\boldsymbol{\mu}}_2$), and $\bar{\mathbf{r}}_{12}$ is the unit vector along the interatomic axis.

The collective parameters (27) and (28), which both depend on the interatomic separation, determine the collective properties of the two-atom system. The parameter (28) is the familiar retarded dipole-dipole interaction between the atoms [7,9,10,21], while Γ_{12} gives rise to the collective spontaneous emission. In Fig. 1, we plot $\Gamma_{12}/\sqrt{\Gamma_1 \Gamma_2}$ and $\Omega_{12}/\sqrt{\Gamma_1 \Gamma_2}$ as a function of r_{12}/λ_0 , where λ_0 is the resonant wavelength. For large separations ($r_{12} \gg \lambda_0$) the parameters are very small ($\Gamma_{12} = \Omega_{12} \approx 0$). By contrast, for atomic separations much smaller than the resonant wavelength (the small sample model), the parameters reduce to

$$\Gamma_{12} = \sqrt{\Gamma_1 \Gamma_2} \quad (29)$$

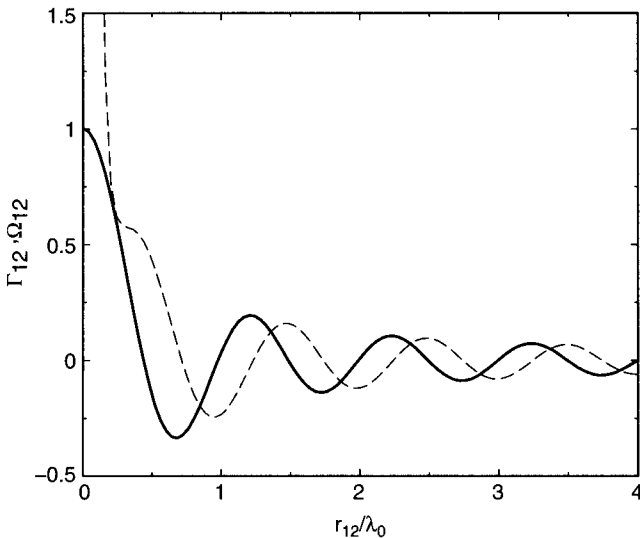


Figure 1. Collective parameters $\Gamma_{12}/\sqrt{\Gamma_1 \Gamma_2}$ (solid line) and $\Omega_{12}/\sqrt{\Gamma_1 \Gamma_2}$ (dashed line) as a function of r_{12}/λ_0 for $\bar{\boldsymbol{\mu}} \perp \bar{\mathbf{r}}_{12}$.

and

$$\Omega_{12} \approx \frac{3\sqrt{\Gamma_1\Gamma_2}}{4(k_0r_{12})^3} [1 - 3(\bar{\boldsymbol{\mu}} \cdot \bar{\mathbf{r}}_{12})^2] \quad (30)$$

For this case Ω_{12} corresponds to the quasistatic dipole-dipole interaction potential.

On transforming Eq. (19) into the Schrödinger picture, the master equation of the two-atom system takes the form

$$\begin{aligned} \frac{\partial}{\partial t} \rho = & -\frac{i}{\hbar} [H', \rho] \\ & -\frac{1}{2} \sum_{i,j=1}^2 \Gamma_{ij} (S_i^+ S_j^- \rho + \rho S_i^+ S_j^- - 2S_j^- \rho S_i^+) \end{aligned} \quad (31)$$

where

$$H' = H_s + H_L + \hbar\Omega_{12}(S_1^+ S_2^- + S_2^+ S_1^-) \quad (32)$$

Equation (31) is the final form of the master equation that gives us an elegant description of the physics involved in the dynamics of two interacting atoms. The collective parameters Γ_{12} and Ω_{12} , which arise from the mutual interaction between the atoms, significantly modify the master equation of a two-atom system. The parameter Γ_{12} introduces a coupling between the atoms through the vacuum field that the spontaneous emission from one of the atoms influences the spontaneous emission from the other. The dipole-dipole interaction Ω_{12} introduces a coherent coupling between the atoms. Owing to the dipole-dipole interaction, the population is coherently transferred back and forth from one atom to the other. Here, the dipole-dipole interaction parameter Ω_{12} plays a role similar to that of the Rabi frequency in the atom-field interaction.

III. COLLECTIVE ATOMIC STATES

The presence of the collective parameters Γ_{12} and Ω_{12} introduces off-diagonal terms in the Hamiltonian H' and in the dissipative part of the master equation. This suggests that in the presence of the interaction between the atoms the bare atomic states are no longer the eigenstates of the two-atom system. We can diagonalize the Hamiltonian (32) with respect to the dipole-dipole interaction and find collective states of the two-atom system.

In the absence of the driving laser field and the dipole-dipole interaction, the basis states of the two-atom system are the four direct products states

$$|g_1\rangle|g_2\rangle, \quad |e_1\rangle|g_2\rangle, \quad |g_1\rangle|e_2\rangle, \quad |e_1\rangle|e_2\rangle \quad (33)$$

In the basis of these states the matrix representation of the Hamiltonian H' , with $\Omega_1 = \Omega_2 = 0$, is given by

$$\frac{H'}{\hbar} = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & \omega_0 - \frac{1}{2}\Delta & \Omega_{12} & 0 \\ 0 & \Omega_{12} & \omega_0 + \frac{1}{2}\Delta & 0 \\ 0 & 0 & 0 & 2\omega_0 \end{pmatrix} \quad (34)$$

where $\omega_0 = \frac{1}{2}(\omega_1 + \omega_2)$ and $\Delta = \omega_2 - \omega_1$.

Evidently, in the presence of the dipole-dipole interaction the matrix (34) is not diagonal, which indicates that the product states (33) are not the eigenstates of the two-atom system. We will diagonalize the matrix (34) for the case of identical ($\Delta = 0$) as well as nonidentical ($\Delta \neq 0$) atoms to find eigenstates of the system and their energies.

A. Collective States of Two Identical Atoms

We begin by studying the collective properties of the system of two identical atoms ($\Delta = 0$). In order to find eigenstates and corresponding energies of the system, we diagonalize the matrix (34), and find that in the case of two identical atoms the eigenstates are given by [1,7]

$$\begin{aligned} |g\rangle &= |g_1\rangle|g_2\rangle \\ |s\rangle &= \frac{1}{\sqrt{2}}(|e_1\rangle|g_2\rangle + |g_1\rangle|e_2\rangle) \\ |a\rangle &= \frac{1}{\sqrt{2}}(|e_1\rangle|g_2\rangle - |g_1\rangle|e_2\rangle) \\ |e\rangle &= |e_1\rangle|e_2\rangle \end{aligned} \quad (35)$$

with corresponding energies

$$\begin{aligned} E_g &= 0 \\ E_s &= \hbar(\omega_0 + \Omega_{12}) \\ E_a &= \hbar(\omega_0 - \Omega_{12}) \\ E_e &= 2\hbar\omega_0 \end{aligned} \quad (36)$$

The eigenstates (35) are the collective states of two interacting atoms and are known in quantum optics as the Dicke states of the two-atom system [1]. We note here that the collective states $|s\rangle$ and $|a\rangle$ are an example of maximally entangled states of the two-atom system that the eigenstates of the system are linear superpositions which cannot be separated into product states of the individual atoms.

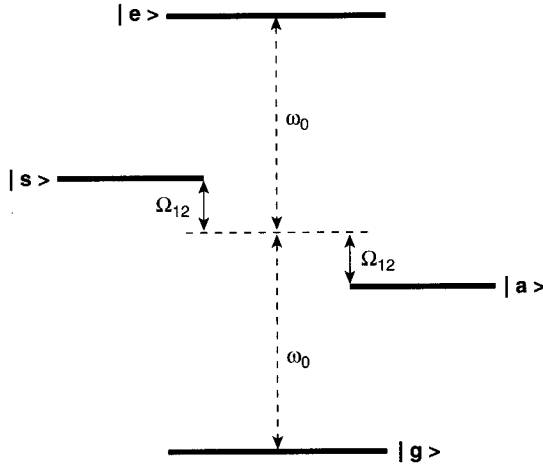


Figure 2. Collective states of two identical atoms. The dipole-dipole interaction Ω_{12} shifts the energies of the symmetric and antisymmetric states in the opposite directions.

The collective states are shown in Fig. 2. It is seen that in the collective states representation, the two-atom system behaves as a single four-level system with the ground state $|g\rangle$, the upper state $|e\rangle$, and two intermediate states: the symmetric state $|s\rangle$ and the antisymmetric state $|a\rangle$. The energies of the intermediate states depend on the dipole-dipole interaction and these states suffer a large shift when the interatomic separation is small.

From Eqs. (1) and (35), we find the following relations between the atomic and collective operators

$$\begin{aligned} S_1^+ &= \frac{1}{\sqrt{2}}(A_{es} - A_{ea} + A_{sg} + A_{ag}) \\ S_2^+ &= \frac{1}{\sqrt{2}}(A_{es} + A_{ea} + A_{sg} - A_{ag}) \end{aligned} \quad (37)$$

where $A_{ij} = |i\rangle\langle j|$, $(i, j = e, a, s, g)$ are the collective operators that represent the energies $(i = j)$ of the collective states and transition dipole moments $(i \neq j)$.

Substituting the relations (37) into Eq. (31), we find that in terms of the collective operators, the master equation is given by

$$\begin{aligned} \frac{\partial}{\partial t} \rho &= -\frac{i}{\hbar}[H_{cs}, \rho] - \frac{1}{2}(\Gamma + \Gamma_{12})\{(A_{ee} + A_{ss})\rho + \rho(A_{ee} + A_{ss}) \\ &\quad - 2(A_{se} + A_{gs})\rho(A_{es} + A_{sg})\} - \frac{1}{2}(\Gamma - \Gamma_{12})\{(A_{ee} + A_{aa})\rho \\ &\quad + \rho(A_{ee} + A_{aa}) - 2(A_{ae} + A_{ga})\rho(A_{ea} + A_{ag})\} \end{aligned} \quad (38)$$

where the Hamiltonian H_{cs} reads

$$\begin{aligned} H_{cs} = & \hbar[2\omega_0 A_{ee} + (\omega_0 + \Omega_{12})A_{ss} + (\omega_0 - \Omega_{12})A_{aa}] \\ & - \frac{\hbar}{2\sqrt{2}} \{ (\Omega_1 + \Omega_2)[(A_{es} + A_{sg})e^{i(\omega_L t + \phi_L)} + \text{H.c.}] \\ & + (\Omega_1 - \Omega_2)[(A_{ea} + A_{ag})e^{i(\omega_L t + \phi_L)} + \text{H.c.}] \} \end{aligned} \quad (39)$$

The master equation (38) provides the simplest example of the effects introduced by the coherent interaction of atoms with the radiation field. These effects include the shifts of the energy levels of the system, produced by the dipole-dipole interaction, and the phenomena of enhanced (superradiant) and reduced (subradiant) spontaneous emission, which appear in the changed damping rates to $\frac{1}{2}(\Gamma + \Gamma_{12})$ and $\frac{1}{2}(\Gamma - \Gamma_{12})$, respectively.

B. Collective States of Two Nonidentical Atoms

The collective states (35) are eigenstates of the system of two identical atoms. If the atoms are not identical, the situation becomes more complicated and we will discuss here some consequences of the fact that the atoms could have different transition frequencies and/or different spontaneous emission rates. When the atoms are nonidentical with different transition frequencies, the states (35) are no longer the eigenstates of the Hamiltonian (32). The diagonalization of the matrix (34) with $\Delta \neq 0$ leads to the following eigenstates [43]

$$\begin{aligned} |g\rangle &= |g_1\rangle|g_2\rangle \\ |s'\rangle &= \beta|e_1\rangle|g_2\rangle + \alpha|g_1\rangle|e_2\rangle \\ |a'\rangle &= \alpha|e_1\rangle|g_2\rangle - \beta|g_1\rangle|e_2\rangle \\ |e\rangle &= |e_1\rangle|e_2\rangle \end{aligned} \quad (40)$$

with energies

$$\begin{aligned} E_g &= 0 \\ E_{s'} &= \hbar \left(\omega_0 + \sqrt{\Omega_{12}^2 + \frac{1}{4}\Delta^2} \right) \\ E_{a'} &= \hbar \left(\omega_0 - \sqrt{\Omega_{12}^2 + \frac{1}{4}\Delta^2} \right) \\ E_e &= 2\hbar\omega_0, \end{aligned} \quad (41)$$

where

$$\alpha = \frac{w}{\sqrt{w^2 + \Omega_{12}^2}}, \quad \beta = \frac{\Omega_{12}}{\sqrt{w^2 + \Omega_{12}^2}} \quad (42)$$

and $w = \frac{1}{2}\Delta + \sqrt{\Omega_{12}^2 + \frac{1}{4}\Delta^2}$.

The energy system of two nonidentical atoms is similar to that of the identical atoms, with the ground state $|g\rangle$, the upper state $|e\rangle$, and two intermediate states $|s'\rangle$ and $|a'\rangle$. It is apparent that the effect of the frequency difference Δ on the collective atomic states is to increase the splitting between the intermediate levels, which now is equal to $\sqrt{\Omega_{12}^2 + \frac{1}{4}\Delta^2}$. However, the most dramatic effect of the detuning Δ is on the degree of entanglement of the states $|s'\rangle$ and $|a'\rangle$ that in the case of nonidentical atoms the states $|s'\rangle$ and $|a'\rangle$ are not maximally entangled states. For $\Delta = 0$ the states $|s'\rangle$ and $|a'\rangle$ reduce to the maximally entangled states $|s\rangle$ and $|a\rangle$, whereas for $\Delta \gg \Omega_{12}$ the entangled states $|s'\rangle$ and $|a'\rangle$ reduce to the product states $|e_1\rangle|g_2\rangle$ and $-|g_1\rangle|e_2\rangle$, respectively.

We follow exactly the same route as in the preceding section, and rewrite the master equation (31) in terms of the collective operators $A_{ij} = |i\rangle\langle j|$, where now the collective states are given in Eq. (40). First, we find that in the case of nonidentical atoms the atomic and collective operators are related by

$$\begin{aligned} S_1^+ &= \beta A_{es'} - \alpha A_{ea'} + \alpha A_{s'g} + \beta A_{a'g} \\ S_2^+ &= \alpha A_{es'} + \beta A_{ea'} + \beta A_{s'g} - \alpha A_{a'g} \end{aligned} \quad (43)$$

In terms of the collective operators A_{ij} the master equation can be written as

$$\frac{\partial}{\partial t} \rho = -\frac{i}{\hbar} [H_{na}, \rho] - \mathcal{L}_d \rho - \mathcal{L}_{nd} \rho \quad (44)$$

where

$$\begin{aligned} H_{na} &= \hbar \left\{ 2\omega_0 A_{ee} + \left(\omega_0 + \sqrt{\Omega_{12}^2 + \frac{1}{4}\Delta^2} \right) A_{s's'} \right. \\ &\quad \left. + \left(\omega_0 - \sqrt{\Omega_{12}^2 + \frac{1}{4}\Delta^2} \right) A_{a'a'} \right\} \\ &\quad - \frac{\hbar}{2} \{ (\alpha\Omega_1 + \beta\Omega_2) [(A_{es'} + A_{s'g})e^{i(\omega_L t + \phi_L)} + \text{H.c.}] \\ &\quad + (\alpha\Omega_1 - \beta\Omega_2) [(A_{ea'} + A_{a'g})e^{i(\omega_L t + \phi_L)} + \text{H.c.}] \} \end{aligned} \quad (45)$$

is the Hamiltonian of the system in the collective states basis. The diagonal dissipative part of the master equation reads

$$\begin{aligned} \mathcal{L}_d \rho &= -\Gamma_{es'} (A_{ee} \rho + \rho A_{ee} - 2A_{s'e} \rho A_{es'}) \\ &\quad - \Gamma_{s'g} (A_{s's'} \rho + \rho A_{s's'} - 2A_{gs'} \rho A_{s'g}) \\ &\quad - \Gamma_{a'g} (A_{a'a'} \rho + \rho A_{a'a'} - 2A_{ga'} \rho A_{a'g}) \\ &\quad - \Gamma_{ea'} (A_{ee} \rho + \rho A_{ee} - 2A_{a'e} \rho A_{ea'}) \end{aligned} \quad (46)$$

while the off-diagonal is given by

$$\begin{aligned}
 \mathcal{L}_{nd}\rho = & -\Gamma_{a's'}\{(A_{a's'} + A_{s'a'})\rho + \rho(A_{a's'} + A_{s'a'}) \\
 & - 2A_{ga'}\rho A_{s'g} - 2A_{gs'}\rho A_{a'g}\} \\
 & - [\alpha\beta(\Gamma_1 + \Gamma_2) + \Gamma_{12}](A_{s'e}\rho A_{s'g} + A_{gs'}\rho A_{es'}) \\
 & + [\alpha\beta(\Gamma_1 + \Gamma_2) - \Gamma_{12}](A_{a'e}\rho A_{a'g} + A_{ga'}\rho A_{ea'}) \\
 & + [\Gamma_{a's'} - 2(\beta^2 - \alpha^2)\Gamma_{12}](A_{s'e}\rho A_{ea'} + A_{a'e}\rho A_{es'}) \\
 & + (\alpha^2\Gamma_1 - \beta^2\Gamma_2)(A_{a'e}\rho A_{s'g} + A_{gs'}\rho A_{ea'}) \\
 & - (\beta^2\Gamma_1 - \alpha^2\Gamma_2)(A_{s'e}\rho A_{a'g} + A_{ga'}\rho A_{es'})
 \end{aligned} \tag{47}$$

with the coefficients

$$\begin{aligned}
 \Gamma_{es'} &= \frac{1}{2}(\beta^2\Gamma_1 + \alpha^2\Gamma_2 + 2\alpha\beta\Gamma_{12}) \\
 \Gamma_{s'g} &= \frac{1}{2}(\alpha^2\Gamma_1 + \beta^2\Gamma_2 + 2\alpha\beta\Gamma_{12}) \\
 \Gamma_{ea'} &= \frac{1}{2}(\alpha^2\Gamma_1 + \beta^2\Gamma_2 - 2\alpha\beta\Gamma_{12}) \\
 \Gamma_{a'g} &= \frac{1}{2}(\beta^2\Gamma_1 + \alpha^2\Gamma_2 - 2\alpha\beta\Gamma_{12}) \\
 \Gamma_{a's'} &= \frac{1}{2}[\alpha\beta(\Gamma_1 - \Gamma_2) + (\beta^2 - \alpha^2)\Gamma_{12}]
 \end{aligned} \tag{48}$$

In the absence of the driving field, the Hamiltonian (45) has a simple diagonal form, where the different terms represent energies of the collective states. In contrast, the dissipative part of the master equation is very extensive and complicated and unlike the case of identical atoms, is not diagonal. The diagonal dissipative part of the master equation, Eq. (46), contains the familiar relaxation terms corresponding to spontaneous transitions between the collective states, and the coefficients $\Gamma_{es'}$, $\Gamma_{s'g}$, $\Gamma_{ea'}$, and $\Gamma_{a'g}$ are the spontaneous emission rates of the transitions. The off-diagonal part, Eq. (47), contains spontaneously induced coherences between the transitions. They are of importance only in systems of atoms with different transition frequencies ($\Delta \neq 0$).

Similar to the case of identical atoms, there are two channels of transitions $|e\rangle \rightarrow |s'\rangle \rightarrow |g\rangle$ and $|e\rangle \rightarrow |a'\rangle \rightarrow |g\rangle$ which decay with the rates $\Gamma_{es'}$, $\Gamma_{s'g}$ and $\Gamma_{ea'}$, $\Gamma_{a'g}$, respectively. However, in contrast to the case of identical atoms, these two channels of transitions are not independent and their decays are correlated through various off-diagonal terms. The decay rates $\Gamma_{ea'}$ and $\Gamma_{a'g}$ are much smaller than the decay rates $\Gamma_{es'}$, $\Gamma_{s'g}$ involving the symmetric state and can be reduced to zero. This happens only for atomic separations much smaller than the optical wavelength (the small sample model). In particular, the decay rate $\Gamma_{a'g}$

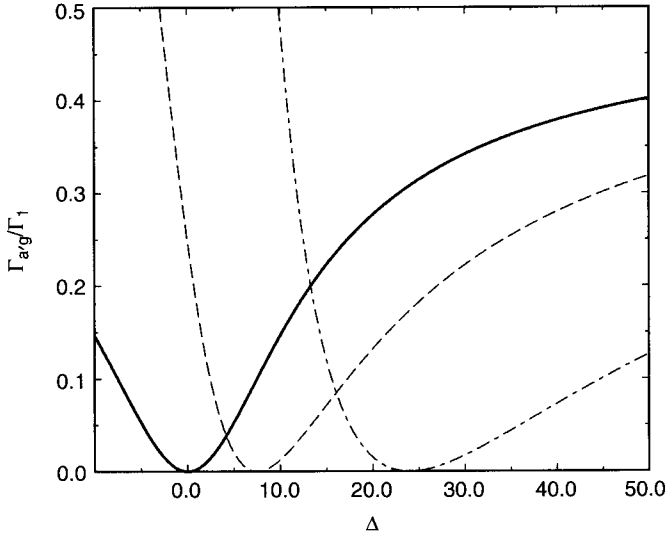


Figure 3. The spontaneous emission damping rate $\Gamma_{a'g}$ as a function of Δ for $\Omega_{12} = 5\Gamma_1$, and different Γ_2 : $\Gamma_2 = \Gamma_1$ (solid line), $\Gamma_2 = 2\Gamma_1$ (dashed line), $\Gamma_2 = 5\Gamma_1$ (dashed-dotted line).

of the antisymmetric state to the ground state, shown in Fig. 3, vanishes when [43]

$$\Gamma_{12} = \sqrt{\Gamma_1 \Gamma_2} \quad \text{and} \quad \frac{\beta}{\alpha} = \sqrt{\frac{\Gamma_2}{\Gamma_1}} \quad (49)$$

The first condition, $\Gamma_{12} = \sqrt{\Gamma_1 \Gamma_2}$, is satisfied when the atoms are separated by distances much smaller than the optical wavelength. The second condition is satisfied when

$$\Delta = -\frac{\Omega_{12}(\Gamma_1 - \Gamma_2)}{2\sqrt{\Gamma_1 \Gamma_2}} \quad (50)$$

Thus, with the condition (49) the antisymmetric state does not decay to the ground state. Moreover, at the condition (49) the interference term vanishes, $\Gamma_{a's'} = 0$. Since in the trapping condition (49) the state $|a'\rangle$ is also decoupled from the interaction with the laser field, the only way to populate this state is by spontaneous emission from the upper state $|e\rangle$.

The decoupling of the antisymmetric state $|a'\rangle$ from the coherent field prevents the state from the external coherent interactions. This is not, however, a useful property in terms of quantum computation, where it is required to

prepare entangled states which are decoupled from the external environment and simultaneously should be accessible by coherent processes.

C. Maximally Entangled States of Two Nonidentical Atoms

The choice of the collective states (40) as a basis leads to a complicated master equation whose physical properties are tractable only for very specific values of the parameters involved. A different choice of basis collective states is proposed here, which allows us to obtain a simple master equation of the system of two nonidentical atoms. Moreover, we will show that it is possible to create a maximally entangled state in the system of two nonidentical atoms that can be decoupled from the external environment and, at the same time, the state exhibits a strong coherent coupling with the remaining states.

In order to show this, we introduce superposition operators S_s^\pm and S_a^\pm , which are linear combinations of the atomic operators S_1^\pm and S_2^\pm , as

$$\begin{aligned} S_s^+ &= uS_1^+ + vS_2^+, & S_s^- &= u^*S_1^- + v^*S_2^- \\ S_a^+ &= vS_1^+ - uS_2^+, & S_a^- &= v^*S_1^- - u^*S_2^- \end{aligned} \quad (51)$$

where the parameters u and v are in general complex numbers such that

$$|u|^2 + |v|^2 = 1 \quad (52)$$

The operators S_s^\pm and S_a^\pm represent, respectively, symmetric and antisymmetric superpositions of the atomic dipole operators. In terms of the operators (51), we can rewrite the dissipative part $\mathcal{L}\rho$ of the master equation as

$$\begin{aligned} \mathcal{L}\rho &= -\Gamma_{ss}(S_s^+ S_s^- \rho + \rho S_s^+ S_s^- - 2S_s^- \rho S_s^+) \\ &\quad - \Gamma_{aa}(S_a^+ S_a^- \rho + \rho S_a^+ S_a^- - 2S_a^- \rho S_a^+) \\ &\quad - \Gamma_{sa}(S_s^+ S_a^- \rho + \rho S_s^+ S_a^- - 2S_a^- \rho S_s^+) \\ &\quad - \Gamma_{as}(S_a^+ S_s^- \rho + \rho S_a^+ S_s^- - 2S_s^- \rho S_a^+) \end{aligned} \quad (53)$$

where the coefficients Γ_{mn} are

$$\begin{aligned} \Gamma_{ss} &= |u|^2\Gamma_1 + |v|^2\Gamma_2 + (uv^* + u^*v)\Gamma_{12} \\ \Gamma_{aa} &= |v|^2\Gamma_1 + |u|^2\Gamma_2 - (uv^* + u^*v)\Gamma_{12} \\ \Gamma_{as} &= uv^*\Gamma_1 - u^*v\Gamma_2 - (|u|^2 - |v|^2)\Gamma_{12} \\ \Gamma_{sa} &= u^*v\Gamma_1 - uv^*\Gamma_2 - (|u|^2 - |v|^2)\Gamma_{12} \end{aligned} \quad (54)$$

The first two terms in Eq. (53) are familiar spontaneous transitions terms and the parameters Γ_{ss} and Γ_{aa} are spontaneous emission rates of the symmetric and antisymmetric superpositions, respectively. The last two terms are due to

coherence between the superposition states and the parameters Γ_{as} and Γ_{sa} appear as cross-damping rates between the superpositions.

If we make the identification

$$u = \sqrt{\frac{\Gamma_1}{\Gamma_1 + \Gamma_2}}, \quad v = \sqrt{\frac{\Gamma_2}{\Gamma_1 + \Gamma_2}}, \quad (55)$$

then the parameters (54) simplify to

$$\begin{aligned} \Gamma_{ss} &= \frac{1}{2}(\Gamma_1 + \Gamma_2) + \frac{\sqrt{\Gamma_1\Gamma_2}(\Gamma_{12} - \sqrt{\Gamma_1\Gamma_2})}{\Gamma_1 + \Gamma_2} \\ \Gamma_{aa} &= \frac{(\sqrt{\Gamma_1\Gamma_2} - \Gamma_{12})\sqrt{\Gamma_1\Gamma_2}}{\Gamma_1 + \Gamma_2} \\ \Gamma_{sa} = \Gamma_{as} &= \frac{1}{2} \frac{(\Gamma_1 - \Gamma_2)(\sqrt{\Gamma_1\Gamma_2} - \Gamma_{12})}{\Gamma_1 + \Gamma_2} \end{aligned} \quad (56)$$

Clearly, the cross-damping terms Γ_{as} and Γ_{sa} vanish when the damping rates of the atoms are equal ($\Gamma_1 = \Gamma_2$). Furthermore, if $\Gamma_{12} = \sqrt{\Gamma_1\Gamma_2}$, then the spontaneous emission rates Γ_{aa} , Γ_{as} , and Γ_{sa} vanish regardless of the ratio between the Γ_1 and Γ_2 . In this limit, which corresponds to the case of the atoms confined to the region much smaller than the optical wavelength, the antisymmetric superposition does not decay and also decouples from the symmetric superposition.

An interesting question arises as to whether the nondecaying antisymmetric superposition can still be coupled to the symmetric superposition through the coherent terms contained in the Hamiltonian H' . To check it, we first transform the Hamiltonian (32) into the interaction picture and next rewrite the transformed Hamiltonian in terms of the S_s^\pm and S_a^\pm operators as

$$\begin{aligned} H' = -\hbar \bigg\{ & \left[\Delta_L - \frac{1}{2}(u^2 - v^2)\Delta \right] S_s^+ S_s^- + \left[\Delta_L + \frac{1}{2}(u^2 - v^2)\Delta \right] S_a^+ S_a^- \\ & - \Delta uv (S_s^+ S_a^- + S_a^+ S_s^-) \bigg\} + \hbar \Omega_{12} [2uv (S_s^+ S_s^- - S_a^+ S_a^-) \\ & + (u^2 - v^2) (S_s^+ S_a^- + S_a^+ S_s^-)] - \frac{\hbar}{2} \{ (u\Omega_1 + v\Omega_2) S_s^+ \\ & + (v\Omega_1 - u\Omega_2) S_a^+ + \text{H.c.} \} \end{aligned} \quad (57)$$

where $\Delta_L = \omega_0 - \omega_L$.

In Eq. (57), the first term arises from the Hamiltonian H_A and shows that the energies of the symmetric and antisymmetric superpositions depend on the energy difference Δ between the atomic transition frequencies and the spontaneous emission rates Γ_i . It is interesting to note that the energy difference Δ

introduces a coherent coupling between the superpositions. If the atoms are identical, $\Delta = 0$, $\Gamma_1 = \Gamma_2$, and then the superpositions have the same energies and there is no contribution to the coherent interaction from the Hamiltonian H_A .

The second term in Eq. (57), proportional to the dipole-dipole interaction between the atoms, has two effects on the dynamics of the symmetric and antisymmetric superpositions. The first is a shift of the energies, and the second is the coherent interaction between the superpositions. It is seen from Eq. (57) that the contribution of Ω_{12} to the coherent interaction between the superpositions vanishes for $\Gamma_1 = \Gamma_2$, and then the effect of Ω_{12} is only the shift of the energies from their unperturbed values. Note that the dipole-dipole interaction Ω_{12} shifts the energies in the opposite directions.

The third term in Eq. (57) represents the interaction of the superpositions with the driving laser field. We see that the symmetric superposition couples to the laser field with an enhanced Rabi frequency proportional to $u\Omega_1 + v\Omega_2$, whereas the Rabi frequency of the antisymmetric superposition is proportional to $v\Omega_1 - u\Omega_2$ and vanishes for $v\Omega_1 = u\Omega_2$.

We may rewrite the Hamiltonian (57) in a physically transparent form that shows explicitly the presence of the coherent coupling between the superpositions

$$\begin{aligned}
 H' = & -\hbar \left\{ \left(\Delta_L - \frac{1}{2} \Delta' \right) S_s^+ S_s^- + \left(\Delta_L + \frac{1}{2} \Delta' \right) S_a^+ S_a^- \right. \\
 & \left. + \Delta_c (S_s^+ S_a^- + S_a^+ S_s^-) \right\} - \frac{\hbar}{2} \{ (u\Omega_1 + v\Omega_2) S_s^+ \\
 & + (v\Omega_1 - u\Omega_2) S_a^+ + \text{H.c.} \}
 \end{aligned} \tag{58}$$

where Δ' and Δ_c are given by

$$\begin{aligned}
 \Delta' &= [(u^2 - v^2)\Delta + 4\Omega_{12}uv] \\
 \Delta_c &= [(u^2 - v^2)\Omega_{12} - \Delta uv]
 \end{aligned} \tag{59}$$

The parameters Δ' and Δ_c allow us to gain physical insight into how the dipole-dipole interaction Ω_{12} and the frequency difference Δ can modify the dynamics of the two-atom system. The parameter Δ' appears as a shift of the energies of the superposition systems, while Δ_c determines the magnitude of the coherent interaction between the superpositions. For $\Omega_{12} \neq 0$ and identical atoms the shift $\Delta' \neq 0$, but can vanish for nonidentical atoms. This occurs for

$$\Omega_{12} = -\frac{1}{4} \frac{(\Gamma_1 - \Gamma_2)\Delta}{\sqrt{\Gamma_1\Gamma_2}} \tag{60}$$

In contrast to the shift Δ' , which is different from zero for identical atoms, the coherent coupling Δ_c can be different from zero only for nonidentical atoms. However, even in this case the coupling can vanish, which happens for

$$\Delta = \frac{\Omega_{12}(\Gamma_1 - \Gamma_2)}{\sqrt{\Gamma_1\Gamma_2}} \quad (61)$$

Obviously, with the condition (61) and $\Gamma_{12} = \sqrt{\Gamma_1\Gamma_2}$ the antisymmetric superposition of two nonidentical atoms completely decouples from the interactions.

Thus, the condition $\Gamma_{12} = \sqrt{\Gamma_1\Gamma_2}$ for suppression of spontaneous emission from the antisymmetric state is valid for identical as well as nonidentical atoms, whereas the coherent interaction between the superpositions appears only for nonidentical atoms with different transition frequencies and/or spontaneous damping rates.

The symmetric and antisymmetric superpositions (51) can be represented by collective states of the system

$$\begin{aligned} |e\rangle &= |e_1\rangle|e_2\rangle \\ |+\rangle &= u|e_1\rangle|g_2\rangle + v|g_1\rangle|e_2\rangle \\ |-\rangle &= v|e_1\rangle|g_2\rangle - u|g_1\rangle|e_2\rangle \\ |g\rangle &= |g_1\rangle|g_2\rangle \end{aligned} \quad (62)$$

In the general case of $\Gamma_1 \neq \Gamma_2$, the superposition states $|+\rangle$ and $|-\rangle$ are non-maximally entangled states. However, the states $|+\rangle$ and $|-\rangle$ can be represented by linear superpositions of the maximally entangled states of two identical atoms as

$$\begin{aligned} |+\rangle &= (u+v)|s\rangle + (u-v)|a\rangle \\ |-\rangle &= (u+v)|a\rangle - (u-v)|s\rangle \end{aligned} \quad (63)$$

The entangled states $|+\rangle$ and $|-\rangle$ are independent of Δ , but depend on the damping rates Γ_1 and Γ_2 . For $\Gamma_1 = \Gamma_2$ ($u = v$) the states are maximally entangled, whereas for either $\Gamma_1 \ll \Gamma_2$ or $\Gamma_1 \gg \Gamma_2$ the entangled states reduce to the product states.

IV. SELECTIVE EXCITATION OF THE COLLECTIVE ATOMIC STATES

We now consider excitation and population transfer processes that can lead to a preparation of the two-atom system in one of the collective states. In particular, we will focus on processes that can prepare the two-atom system in the

entangled symmetric state $|s\rangle$. Our main interest, however, is in the preparation of the system in the maximally entangled antisymmetric state $|a\rangle$, which, under the condition $\Gamma_{12} = \sqrt{\Gamma_1\Gamma_2}$, is a decoherence-free state.

A. Preparation of the Symmetric State by a Pulse Laser

It has been shown [31] that a system of two identical two-level atoms may be prepared in the symmetric state $|s\rangle$ by a short laser pulse. The conditions for a selective excitation of the collective atomic states can be analyzed from the interaction Hamiltonian of the laser field with the two-atom system. We make the unitary transformation

$$\tilde{H}_L = e^{iH_0t/\hbar} H' e^{-iH_0t/\hbar} \quad (64)$$

where

$$H_0 = \hbar\{2\Delta_L|e\rangle\langle e| + (\Delta_L + \Omega_{12})|s\rangle\langle s| + (\Delta_L - \Omega_{12})|a\rangle\langle a|\} \quad (65)$$

and find that in the case of identical atoms, $\Gamma_1 = \Gamma_2$ and $\Delta = 0$, the transformed interaction Hamiltonian \tilde{H}_L is given by

$$\begin{aligned} \tilde{H}_L = & -\frac{\hbar}{2\sqrt{2}}\{(\Omega_1 + \Omega_2)(S_{es}^+ e^{i(\Delta_L - \Omega_{12})t} + S_{sg}^+ e^{i(\Delta_L + \Omega_{12})t}) \\ & + (\Omega_1 - \Omega_2)(S_{ag}^+ e^{i(\Delta_L - \Omega_{12})t} + S_{ea}^+ e^{i(\Delta_L + \Omega_{12})t}) + \text{H.c.}\} \end{aligned} \quad (66)$$

where Hamiltonian represents the interaction of the laser field with the collective two-atom system, and in the transformed form contains terms oscillating at frequencies $(\Delta_L \pm \Omega_{12})$, which correspond to the two separate groups of transitions between the collective atomic states at frequencies $\omega_L = \omega_0 + \Omega_{12}$ and $\omega_L = \omega_0 - \Omega_{12}$. The $\Delta_L + \Omega_{12}$ frequencies are separated from $\Delta_L - \Omega_{12}$ frequencies by $2\Omega_{12}$, and hence the two groups of the transitions evolve separately when $\Omega_{12} \gg \Gamma$. Depending on the frequency, the laser can be selectively tuned to one of the two groups of the transitions. When $\omega_L = \omega_0 + \Omega_{12}$ ($\Delta_L + \Omega_{12} = 0$), the laser is tuned to exact resonance with the $|e\rangle - |a\rangle$ and $|g\rangle - |s\rangle$ transitions, and then the terms appearing in the Hamiltonian (66) and corresponding to these transitions have no explicit time dependence. In contrast, the $|g\rangle - |a\rangle$ and $|e\rangle - |s\rangle$ transitions are off-resonance and the terms corresponding to these transitions have an explicit time dependence $\exp(\pm 2i\Omega_{12}t)$. If $\Omega_{12} \gg \Gamma$, the off-resonance terms rapidly oscillate with the frequency $2\Omega_{12}$, and then we can make a secular approximation in which we neglect all those rapidly oscillating terms. The interaction Hamiltonian can then be written in the simplified form:

$$\tilde{H}_L = -\frac{\hbar}{2\sqrt{2}}[(\Omega_1 + \Omega_2)S_{sg}^+ + (\Omega_1 - \Omega_2)S_{ea}^+ + \text{H.c.}] \quad (67)$$

It is seen that the laser field couples to the transitions with significantly different Rabi frequencies. The coupling strength of the laser to the $|g\rangle - |s\rangle$ transition is proportional to the sum of the Rabi frequencies $\Omega_1 + \Omega_2$, whereas the coupling strength of the laser to the $|a\rangle - |e\rangle$ transition is proportional to the difference of the Rabi frequencies $\Omega_1 - \Omega_2$. According to Eq. (10), the Rabi frequencies Ω_1 and Ω_2 of two identical atoms differ only by the phase factor $\exp(i\mathbf{k}_L \cdot \mathbf{r}_{12})$. Thus, in order to selectively excite the $|g\rangle - |s\rangle$ transition, the driving laser field should be in phase with both atoms: $\Omega_1 = \Omega_2$. This can be achieved by choosing the propagation vector \mathbf{k}_L of the laser orthogonal to the line joining the atoms. Under this condition we can make a further simplification and truncate the state vector of the system into two states $|g\rangle$ and $|s\rangle$. In this two-state approximation we find from the Schrödinger equation the time evolution of the population $P_s(t)$ of the state $|s\rangle$ as

$$P_s(t) = \sin^2\left(\frac{1}{\sqrt{2}}\Omega t\right) \quad (68)$$

where $\Omega = \Omega_1 = \Omega_2$.

The population oscillates with the Rabi frequency of the $|g\rangle - |s\rangle$ transition and at certain times $P_s(t) = 1$, indicating that all the population is in the symmetric state. This happens at times

$$T_n = (2n + 1) \frac{\pi}{\sqrt{2}\Omega}, \quad n = 0, 1, \dots \quad (69)$$

Hence, the system can be prepared in the state $|s\rangle$ by simply applying a laser pulse, for example, with the duration T_0 , that is a standard π pulse.

The two-state approximation is of course an idealization, and a possibility that all the transitions can be driven by the laser imposes significant limits on the Rabi frequency and the duration of the pulse. Namely, the Rabi frequency cannot be too strong in order to avoid the coupling of the laser to the $|s\rangle - |e\rangle$ transition, which could lead to a slight pumping of the population to the state $|e\rangle$. On the other hand, the Rabi frequency cannot be too small as for a small Ω the duration of the pulse, required for the complete transfer of the population into the state $|s\rangle$, becomes longer and then spontaneous emission can occur during the excitation process. Therefore, the transfer of the population to the state $|s\rangle$ cannot be made arbitrarily fast and, in addition, requires a careful estimation of the optimal Rabi frequency, which could be difficult to achieve in a real experimental situation.

B. Preparation of the Antisymmetric State

1. Pulse Laser

If we choose the laser frequency such that $\Delta_L - \Omega_{12} = 0$, the laser field is then resonant to the $|a\rangle - |g\rangle$ and $|e\rangle - |s\rangle$ transitions and, after the secular

approximation, the Hamiltonian (66) reduces to

$$\tilde{H}_L = -\frac{\hbar}{2\sqrt{2}}[(\Omega_1 - \Omega_2)S_{ag}^+ + (\Omega_1 + \Omega_2)S_{es}^+ + \text{H.c.}] \quad (70)$$

Clearly, for $\Omega_1 = -\Omega_2$ the laser couples only to the $|a\rangle - |g\rangle$ transition. Thus, in order to selectively excite the $|g\rangle - |a\rangle$ transition, the atoms should experience opposite phases of the laser field. This can be achieved by choosing the propagation vector \mathbf{k}_L of the laser along the interatomic axis, and the atomic separations such that

$$\mathbf{k}_L \cdot \mathbf{r}_{12} = (2n + 1)\pi, \quad n = 0, 1, 2, \dots \quad (71)$$

which corresponds to a situation that the atoms are separated by a distance $r_{12} = (2n + 1)\lambda_0/2$.

The smallest distance at which the atoms could experience opposite phases corresponds to $r_{12} = \lambda_0/2$. However, at this particular separation the dipole-dipole interaction parameter Ω_{12} is small (see Fig. 1), and then all of the transitions between the collective states occur at approximately the same frequency. In this case the secular approximation is not valid, and we cannot separate the transitions at $\Delta_L + \Omega_{12}$ from the transitions at $\Delta_L - \Omega_{12}$.

One possible solution to the problem of the selective excitation with opposite phases is to use a standing laser field instead of the running wave field. If the laser amplitudes differ by the sign, namely, $\mathbf{E}_{L_1} = -\mathbf{E}_{L_2} = \mathbf{E}_0$, and $\mathbf{k}_{L_1} \cdot \mathbf{r}_1 = -\mathbf{k}_{L_2} \cdot \mathbf{r}_2$, the Rabi frequencies experienced by the atoms are

$$\begin{aligned} \Omega_1 &= \frac{2i}{\hbar} \boldsymbol{\mu}_1 \cdot \mathbf{E}_0 \sin\left(\frac{1}{2} \mathbf{k}_L \cdot \mathbf{r}_{12}\right) \\ \Omega_2 &= -\frac{2i}{\hbar} \boldsymbol{\mu}_2 \cdot \mathbf{E}_0 \sin\left(\frac{1}{2} \mathbf{k}_L \cdot \mathbf{r}_{12}\right) \end{aligned} \quad (72)$$

where $\mathbf{k}_L = \mathbf{k}_{L_1} = \mathbf{k}_{L_2}$ and we have chosen the reference frame such that $\mathbf{r}_1 = \frac{1}{2}\mathbf{r}_{12}$ and $\mathbf{r}_2 = -\frac{1}{2}\mathbf{r}_{12}$. It follows from Eq. (72) that the Rabi frequencies oscillate with opposite phases independent of the separation between the atoms. However, the magnitude of the Rabi frequencies decreases with decreasing r_{12} .

2. Indirect Driving through the Symmetric State

We now turn to the situation of nonidentical atoms and consider different possible processes of the population transfer to the antisymmetric state that could be present even if the antisymmetric state does not decay to the ground level. This can happen when $\Gamma_{12} = \sqrt{\Gamma_1 \Gamma_2}$, that is, when the separation between the atoms is negligible small. Under this condition the antisymmetric state is also decoupled from the driving field. According to Eq. (58), the antisymmetric

state can still be coupled, through the coherent interaction Δ_c , to the symmetric state $|+\rangle$. However, this coupling appears only for nonidentical atoms.

From the master equation (31), we find that under the condition $\Gamma_{12} = \sqrt{\Gamma_1 \Gamma_2}$ the equation of motion for the population of the state $|-\rangle$ is given by [33]

$$\begin{aligned} \dot{\rho}_{--} = & \frac{(\Gamma_1 - \Gamma_2)^2}{\Gamma_1 + \Gamma_2} \rho_{ee} + i\Delta_c(\rho_{+-} - \rho_{-+}) \\ & - \frac{1}{2} i\Omega \frac{(\Gamma_1 - \Gamma_2)}{\sqrt{\Gamma_1^2 + \Gamma_2^2}} (\rho_{e-} - \rho_{-e}) \end{aligned} \quad (73)$$

This equation shows that the nondecaying antisymmetric state $|-\rangle$ can be populated by spontaneous emission from the upper state $|e\rangle$ and also by the coherent interaction with the state $|+\rangle$. The first condition is satisfied only when $\Gamma_1 \neq \Gamma_2$, while the other condition is satisfied only when $\Delta_c \neq 0$. Thus, the transfer of population to the state $|-\rangle$ from the upper state $|e\rangle$ and the symmetric state $|s\rangle$ does not appear when the atoms are identical, but is possible for nonidentical atoms.

We illustrate this effect in Fig. 4, where we plot the steady-state population of the state $|-\rangle$ as a function of Δ_L for two different types of nonidentical atoms. In the first case the atoms have the same damping rates ($\Gamma_1 = \Gamma_2$) but different transition frequencies ($\Delta \neq 0$), while in the second case the atoms have the same frequencies ($\Delta = 0$) but different damping rates ($\Gamma_1 \neq \Gamma_2$). It is

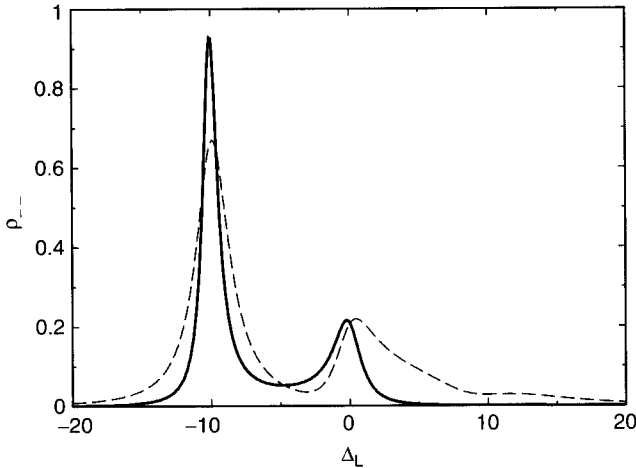


Figure 4. The steady-state population of the antisymmetric state $|-\rangle$ for $\Omega = 5\Gamma_1$, $\Omega_{12} = 10\Gamma_1$ and $\Gamma_2 = \Gamma_1$, $\Delta = \Gamma_1$ (solid line), $\Gamma_2 = 2\Gamma_1$, $\Delta = 0$ (dashed line).

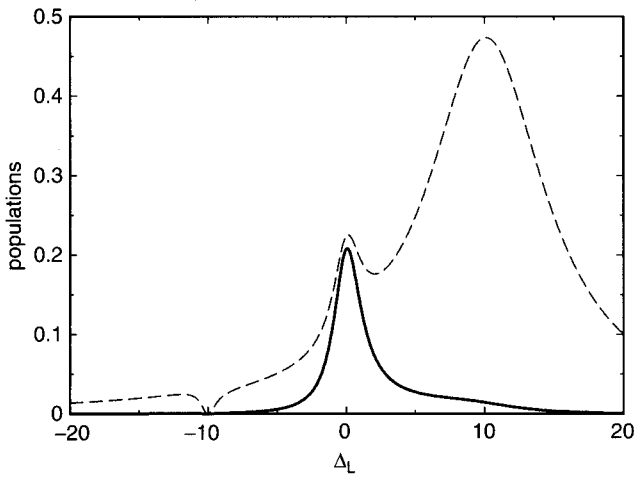


Figure 5. The steady-state populations of the upper state $|e\rangle$ (solid line) and the symmetric state $|+\rangle$ (dashed line) for $\Gamma_2 = \Gamma_1$, $\Omega = 5\Gamma_1$, $\Omega_{12} = 10\Gamma_1$ and $\Delta = \Gamma_1$.

seen from Fig. 4 that in both cases the antisymmetric state can be populated even if is not directly driven from the ground state. The population is transferred to $|-\rangle$ through the coherent interaction Δ_c , which leaves the other excited states completely unpopulated. This is shown in Fig. 5, where we plot the steady-state populations ρ_{++} and ρ_{ee} of the states $|+\rangle$ and $|e\rangle$. It is apparent from Fig. 5 that at $\Delta_L = -\Omega_{12}$ the states $|+\rangle$ and $|e\rangle$ are not populated. However, the population is not entirely trapped in the antisymmetric state $|-\rangle$, but rather in a linear superposition of the antisymmetric and ground states. This is shown in Fig. 6, where we plot the steady-state population ρ_{--} for the same parameters as in Fig. 5, but different Ω . Clearly, for a small Ω the steady-state population $\rho_{--} \approx \frac{1}{2}$, and the amount of the population increases with increasing Ω . The population ρ_{--} attains the maximum value $\rho_{--} \approx 1$ for a very strong driving field.

This result shows that we can relatively easily prepare two nonidentical atoms in the maximally entangled antisymmetric state. The closeness of the prepared state to the ideal one is measured by the fidelity F . Here F is equal to the obtained maximum population in the state $|-\rangle$. For $\Omega \gg \Gamma$ the fidelity of the prepared state is maximal, equal to 1. As we have already mentioned, the system has the advantage that the maximally entangled state $|-\rangle$ does not decay, that is, is a decoherence-free state.

3. Atom-Cavity-Field Interaction

There have been several proposals to generate the antisymmetric state $|a\rangle$ in a system of two identical atoms interacting with a single-mode cavity field. For

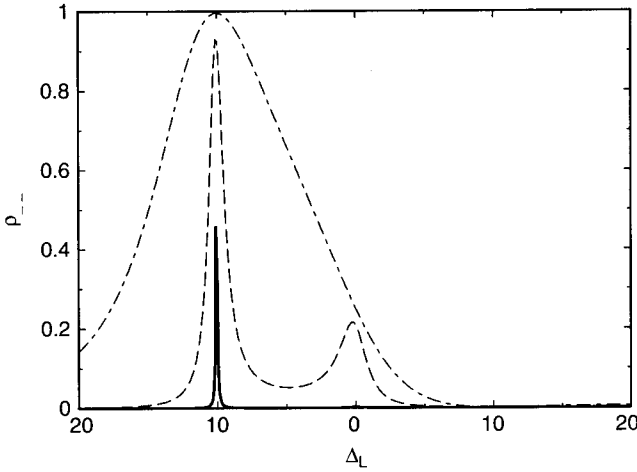


Figure 6. The Steady-state population of the antisymmetric state $|-\rangle$ for $\Gamma_2 = \Gamma_1, \Omega_{12} = 10\Gamma_1, \Delta = \Gamma_1$ and different Ω : $\Omega = \Gamma_1$ (solid line), $\Omega = 5\Gamma_1$ (dashed line), $\Omega = 20\Gamma_1$ (dashed-dotted line).

example, Plenio et al. [30] have considered a system of two atoms trapped inside an optical cavity and separated by a distance much larger than the optical wavelength. This allows for the selective excitation of only one of the atoms. In this scheme the generation of the antisymmetric state relies on the concept of conditional dynamics due to continuous observation of the cavity field. If only one atom is excited and no photon is detected outside the cavity, the atoms are prepared in a dark state [44], which is equivalent to the antisymmetric state $|a\rangle$.

Several investigators [26–28] have analyzed two-atom Jaynes-Cummings models for a violation of Bell's inequality, and have shown that the atoms moving across a single-mode cavity can be prepared in the antisymmetric state via the interaction with the cavity field. In this scheme, the preparation of the antisymmetric state takes place in two steps. In the first step, one atom initially prepared in its excited state $|e_1\rangle$ is sent through a single-mode cavity being in the vacuum state $|0\rangle_c$. During the interaction with the cavity mode, the atomic population undergoes the vacuum Rabi oscillations, and the interaction time was varied by selecting different atomic velocities. If the velocity of the atom is such that the interaction time of the atom with the cavity mode is equal to a quarter of the vacuum Rabi oscillations, then the state of the combined system, the atom plus the cavity mode, is a superposition state:

$$|a_1c\rangle = \frac{1}{\sqrt{2}}(|e_1\rangle|0\rangle_c - |g_1\rangle|1\rangle_c) \quad (74)$$

Hence, the state of the total system, two atoms plus the cavity mode, after the first atom has crossed the cavity is

$$|\Psi_1\rangle = \frac{1}{\sqrt{2}}(|e_1\rangle|0\rangle_c - |g_1\rangle|1\rangle_c)|g_2\rangle \quad (75)$$

If we now send the second atom, which is in its ground state, with the selected velocity such that during the interaction with the cavity mode the atom undergoes half of the vacuum Rabi oscillation, the final state of the system becomes

$$\begin{aligned} |\Psi_{12c}\rangle &= \frac{1}{\sqrt{2}}(|e_1\rangle|0\rangle_c|g_2\rangle - |g_1\rangle|0\rangle_c|e_2\rangle) \\ &= \frac{1}{\sqrt{2}}(|e_1\rangle|g_2\rangle - |g_1\rangle|e_2\rangle)|0\rangle_c = |a\rangle|0\rangle_c \end{aligned} \quad (76)$$

Thus, the final state of the system is a product state of the atomic antisymmetric state $|a\rangle$ and the vacuum state of the cavity mode. In this scheme the cavity mode is left in the vacuum state, which protects the antisymmetric state against any noise of the cavity. The scheme to entangle two atoms in a cavity, proposed by Cirac and Zoller [28], has been realized experimentally by Hagly et al. [36].

Gerry [29] has proposed a similar method based on a dispersive interaction of the atoms with a cavity mode prepared in a coherent state $|\alpha\rangle$. The atoms enter the cavity in superposition states

$$\begin{aligned} |a_1\rangle &= \frac{1}{\sqrt{2}}(|e_1\rangle + i|g_1\rangle) \\ |a_2\rangle &= \frac{1}{\sqrt{2}}(|e_2\rangle - i|g_2\rangle) \end{aligned} \quad (77)$$

After passage of the second atom, the final state of the system is

$$\begin{aligned} |\Psi_{12c}\rangle &= \frac{1}{2}\{(|g_1\rangle|g_2\rangle + |e_1\rangle|e_2\rangle)|-\alpha\rangle \\ &\quad + i(|e_1\rangle|g_2\rangle - |g_1\rangle|e_2\rangle)|\alpha\rangle\} \end{aligned} \quad (78)$$

Thus, if the cavity field is measured and found in the state $|\alpha\rangle$, the atoms are in the antisymmetric state. If the cavity field is found in the state $|\alpha\rangle$, the atoms are in the entangled state:

$$|\Psi_{12}(-\alpha)\rangle = \frac{1}{2}(|g_1\rangle|g_2\rangle + |e_1\rangle|e_2\rangle) \quad (79)$$

The state (79) is called a *two-photon entangled state*. In Section VI, we will discuss another method of preparing the system in the two-photon entangled state based on the interaction of two atoms with a squeezed vacuum field.

C. Preparation of a Superposition of Antisymmetric and Ground States

In the section IV.B.2, we have shown that two nonidentical two-level atoms can be prepared in an arbitrary superposition of the maximally entangled antisymmetric state $|a\rangle$ and the ground state $|g\rangle$

$$|\Phi\rangle = \eta|a\rangle + \sqrt{1 - |\eta|^2}|g\rangle \quad (80)$$

However, the preparation of the superposition state requires that the atoms have different transition frequencies. Beige et al. [32] have proposed a scheme in which the superposition state $|\Phi\rangle$ can be prepared in a system of two identical atoms placed at fixed positions inside an optical cavity.

Here, we discuss an alternative scheme where the superposition state $|\Phi\rangle$ can be generated in two identical atoms driven in free space by a coherent laser field. This can happen when the atoms are in nonequivalent positions in the driving field, where the atoms experience different intensities and phases of the driving field. The populations of the collective states of the system can be found from the master equation (31). We use the set of the collective states (35) as an appropriate representation for the density operator

$$\rho = \sum_{ij} \rho_{ij} |i\rangle \langle j|, \quad i, j = g, s, a, e \quad (81)$$

where ρ_{ij} are the density matrix elements in the basis of the collective states.

After transforming to the collective state basis, the master equation (31) leads to a closed system of 15 equations of motion for the density matrix elements [46]. However, for a specifically chosen geometry for the driving field, namely, that the field is propagated perpendicularly to the atomic axis ($\mathbf{k}_L \cdot \mathbf{r}_{12} = 0$), the system of equations decouples into 9 equations for symmetric and 6 equations for antisymmetric combinations of the density matrix elements [45–50]. In this case, we can solve the system analytically, and find that the steady-state values of the populations are [45,46]

$$\begin{aligned} \rho_{ee} &= \frac{1}{4} \frac{\Omega^4}{D} \\ \rho_{ss} &= \frac{1}{4} \frac{2\Omega^2(\Gamma^2 + \Delta_L^2) + \Omega^4}{D} \\ \rho_{aa} &= \frac{1}{4} \frac{\Omega^4}{D} \end{aligned} \quad (82)$$

where

$$D = \Omega^4 + (\Gamma^2 + \Delta_L^2) \left\{ \Omega^2 + \frac{1}{4} [(\Gamma + \Gamma_{12})^2 + (\Delta_L - \Omega_{12})^2] \right\} \quad (83)$$

and $\Delta_L = \omega_0 - \omega_L$.

In this case all of the collective states are populated with the population distribution $\rho_{ee} = \rho_{aa} < \rho_{ss}$. Moreover, for a very strong driving field ($\Omega \gg \Gamma, \Delta_L$), the excited states are equally populated with $\rho_{ee} = \rho_{ss} = \rho_{aa} = \frac{1}{4}$. The population distribution changes dramatically when the driving field propagates in directions different from perpendicular to the interatomic axis [49–50]. In this situation the populations strongly depend on the interatomic separation and the detuning Δ_L . This can produce the interesting modification that the collective states can be selectively populated. We show this by solving numerically the system of 15 equations for the density matrix elements. The populations are plotted against the detuning Δ_L in Fig. 7 for the laser field propagating in the direction of the interatomic axis. We see from Fig. 7 that the collective excited states are populated for most values of Δ_L , except $\Delta_L = -\Omega_{12}$. At this detuning the antisymmetric state is significantly populated, whereas the population of the symmetric and upper states is close to zero. Since $\rho_{aa} < 1$, the population is distributed between the antisymmetric and the ground

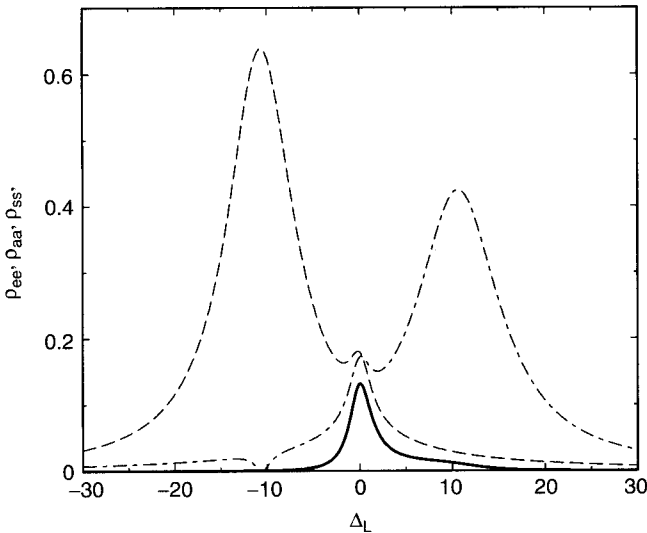


Figure 7. The steady-state populations of the collective atomic states of two identical atoms as a function of Δ_L for the driving field propagating in the direction of the interatomic axis, $\Omega = 2.5\Gamma$, $r_{12}/\lambda_0 = 0.08$ and $\vec{\mu} \perp \vec{r}_{12}$: ρ_{ee} (solid line), ρ_{aa} (dashed line), ρ_{ss} (dashed-dotted line).

states, and therefore at $\Delta_L = -\Omega_{12}$ the system is in a superposition of the maximally entangled state $|a\rangle$ and the ground state $|g\rangle$.

Turchette et al. [35] have realized experimentally a superposition state of the ground state and a nonmaximally entangled antisymmetric state in two trapped ions. In the experiment two trapped barium ions were sideband-cooled to their motional ground states. Transitions between the states of the ions were induced by Raman pulses using copropagating lasers. The ions were at positions that experience different Rabi frequencies Ω_1 and Ω_2 of the laser fields. By preparing the initial motional ground state with one ion excited $|e_1\rangle|g_2\rangle|0\rangle$, and applying the laser fields for a time t , the following entangled state $|\Psi(t)\rangle$ was created

$$|\Psi(t)\rangle = -\frac{i\Omega_2}{\Omega} \sin(\Omega t) |g\rangle|1\rangle + \left\{ \left[\frac{\Omega_2^2}{\Omega^2} (\cos \Omega t - 1) + 1 \right] |e_1\rangle|g_2\rangle + \left[\frac{\Omega_1 \Omega_2}{\Omega^2} (\cos \Omega t - 1) \right] |g_1\rangle|e_2\rangle \right\} |0\rangle \quad (84)$$

where $\Omega^2 = \Omega_1^2 + \Omega_2^2$.

For $\Omega t = \pi$ the entangled state (84) reduces to a nonmaximally entangled antisymmetric state

$$|\Psi_a\rangle = \left[\frac{\Omega_1^2 - \Omega_2^2}{\Omega^2} |e_1\rangle|g_2\rangle - \frac{2\Omega_1 \Omega_2}{\Omega^2} |g_1\rangle|e_2\rangle \right] |0\rangle \quad (85)$$

Franke et al. [51] proposed using the nonmaximally entangled state (85) to demonstrate the intrinsic difference between quantum and classical information transfers. The difference arises from the different ways in which the probabilities occur and is particularly clear in terms of entangled states.

V. DETECTION OF THE ENTANGLED STATES

In this section we discuss problems that could be involved in any attempt to detect an internal entangled state of two coupled atoms in free space. Beige et al. [34] have proposed a scheme, based on the quantum Zeno effect, to observe a decoherence-free state in a system of two 3-level atoms located inside an optical cavity. Here, we discuss possible schemes to detect entangled states of two 2-level atoms in free space.

A. Fluorescence Intensity

One of the possible ways to detect an internal state of two coupled atoms is to observe the fluorescence field emitted from the system. It is well known that the fluorescence from the two-atom system exhibits strong directional properties [7,10,48,52].

To show this, we consider the fluorescence intensity detected at a point \mathbf{R} in the far-field zone of the radiation emitted by the atomic system. The intensity is proportional to the first-order correlation functions of the atomic dipole operators as [7,8]

$$I(\mathbf{R}, t) = U(\mathbf{R}) \sum_{i,j=1}^2 \left\langle S_i^+ \left(t - \frac{R}{c} \right) S_j^- \left(t - \frac{R}{c} \right) \right\rangle e^{ik\mathbf{R} \cdot \mathbf{r}_{ij}}, \quad (86)$$

where

$$U(\mathbf{R}) = \left(\frac{\omega_0^4 \mu^2}{2R^2 \pi \epsilon_0} \right) \sin^2 \varphi \quad (87)$$

is the geometric factor with φ the angle between the observation direction $\mathbf{R} = R\hat{\mathbf{R}}$ and the atomic dipole moment $\boldsymbol{\mu}$.

From Eqs. (86) and (81) the fluorescence intensity can be written in terms of the density matrix elements in the collective states representation as

$$\begin{aligned} I(\mathbf{R}, t) = U(\mathbf{R}) \{ & (\rho_{ee} + \rho_{ss})[1 + \cos(kr_{12} \cos \theta)] \\ & + (\rho_{ee} + \rho_{aa})[1 - \cos(kr_{12} \cos \theta)] \\ & + i(\rho_{sa} - \rho_{as}) \sin(kr_{12} \cos \theta) \} \end{aligned} \quad (88)$$

where θ is the angle between the observation direction $\hat{\mathbf{R}}$ and the vector \mathbf{r}_{12} .

The first term in Eq. (88) arises from the fluorescence emitted on the $|e\rangle \rightarrow |s\rangle \rightarrow |g\rangle$ transitions, which involve the symmetric state. The second term arises from the $|e\rangle \rightarrow |a\rangle \rightarrow |g\rangle$ transitions through the antisymmetric state. These two terms describe two different channels of transitions for which the angular distribution is proportional to $[1 \pm \cos(kr_{12} \cos \theta)]$. The last term in Eq. (88) originates from interference between these two radiation channels. It is seen from Eq. (1.88) that the angular distribution of the fluorescence field depends on the population of the entangled states $|s\rangle$ and $|a\rangle$. Moreover, independent of the interatomic separation r_{12} , the antisymmetric state does not radiate in the direction perpendicular to the atomic axis, as for $\theta = \pi/2$ the factor $[1 - \cos(kr_{12} \cos \theta)]$ vanishes. In contrast, the symmetric state radiates in all directions.

It is evident from Eq. (88) that the radiation pattern is nonspherical unless $\rho_{ss} = \rho_{aa}$ and then the pattern is spherically symmetric independent of the interatomic separation. Therefore, an asymmetry in the radiation pattern would be compelling evidence that the entangled states $|s\rangle$ and $|a\rangle$ are not equally populated. If the fluorescence were detected in the direction perpendicular to the atomic axis, the observed intensity (if any) would correspond to the fluorescence field emitted from the symmetric state $|s\rangle$ and/or the upper state $|e\rangle$. On the other hand, if there is no fluorescence detected in the direction perpendicular

to the atomic axis, the population is entirely in a superposition of the anti-symmetric state $|a\rangle$ and the ground state $|g\rangle$.

Guo and Yang [53] have analyzed spontaneous decay from two atoms initially prepared in an entangled state. They have shown that the time evolution of the population inversion, which is proportional to the intensity (87), depends on the degree of entanglement of the initial state of the system. Ficek et al. [10] have shown that in the case of two nonidentical atoms, the time evolution of the intensity $I(\mathbf{R}, t)$ can exhibit quantum beats that result from the presence of correlations between the symmetric and antisymmetric states. In fact, quantum beats are present only if initially the system is in a nonmaximally entangled state, and no quantum beats are predicted for maximally entangled as well as unentangled states.

B. Interference Pattern

An alternative way to detect an internal state of the two atom system is to observe an interference pattern of the fluorescence field emitted in the direction \mathbf{R} , not necessary perpendicular to the interatomic axis. The usual measure of the depth of modulation of the interference fringes is a visibility defined as

$$\mathcal{V} = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}} \quad (89)$$

where I_{\max} corresponds to $\cos(k\bar{\mathbf{R}} \cdot \mathbf{r}_{12}) = 1$, whereas I_{\min} corresponds to $\cos(k\bar{\mathbf{R}} \cdot \mathbf{r}_{12}) = -1$. This scheme is particularly useful when the antisymmetric state is a decoherence-free (dark) state.

Using Eq. (88), we can write the visibility in the basis of the collective states as

$$\mathcal{V} = \frac{\rho_{ss} - \rho_{aa}}{\rho_{ss} + \rho_{aa} + 2\rho_{ee}} \quad (90)$$

This equation shows that the sign of \mathcal{V} depends on the population difference between the symmetric and antisymmetric states. For $\rho_{ss} > \rho_{aa}$ the visibility \mathcal{V} is positive, and then the interference pattern exhibits a maximum (bright center), whereas for $\rho_{ss} < \rho_{aa}$ the visibility \mathcal{V} is negative and then there is a minimum (dark center). The optimum positive (negative) value is $\mathcal{V} = 1$ ($\mathcal{V} = -1$), and there is no interference pattern when $\mathcal{V} = 0$. The later happens when $\rho_{ss} = \rho_{aa}$.

Similar to the fluorescence intensity distribution, the visibility can provide us an information about the internal state of the system. When the system is prepared in the antisymmetric state or in a superposition of the antisymmetric and the ground states, $\rho_{ss} = \rho_{ee} = 0$, and then the visibility has the optimum negative value $\mathcal{V} = -1$. On the other hand, when the system is prepared in the symmetric state or in a linear superposition of the symmetric and ground states, the visibility has the maximum positive value $\mathcal{V} = 1$.

There have been several theoretical studies of the fringe visibility in the fluorescence field emitted by two coupled atoms, and the Young's interference-type pattern has been observed experimentally in the resonance fluorescence of two trapped ions [11]. The experimental results have been explained theoretically by Wong et al. [54], and can be understood by treating the ions as independent radiators that are synchronized by the constant phase of the driving field. It has been shown that for a weak driving field, the fluorescence field is predominantly composed of an elastic component and therefore the ions behave as point sources of coherent light producing an interference pattern. Under strong excitation the fluorescence field is mostly composed of the incoherent part and consequently there is no interference pattern. Dung and Ujihara [52] have shown that an interference pattern can be observed in spontaneous emission from two interacting atoms even if it is known for certain which atom is excited initially. Michelson type (temporary) interference pattern has been predicted in spontaneous emission from two non-identical atoms [43]. Kochan et al. [55] have shown that the interference pattern of the strongly driven atoms can be partially recovered by placing the atoms inside an optical cavity. The coupling of the atoms to the cavity mode induces atomic correlations, which improves the fringe visibility. Meyer and Yeoman [56] have reported an even stronger cavity induced modification of the interference pattern that occurs when the coherent driving field is replaced by an incoherent field. They have shown that in contrast to the coherent excitation, the incoherent field produces an interference pattern with a dark center. Interference pattern with a dark center has also been predicted when the atoms experience different intensities of the driving field [57] or in the case where the driving field is replaced by a squeezed vacuum field [58].

VI. TWO-PHOTON ENTANGLED STATES

We have already discussed different methods of generating two-atom entangled states of the form

$$|\Psi\rangle = c_1|e_1\rangle|g_2\rangle \pm c_2|g_1\rangle|e_2\rangle \quad (91)$$

These states are generated by the dipole-dipole interaction between the atoms and the preparation of these states is sensitive to the difference Δ between the atomic transition frequencies and to the atomic decay rates.

There are two other collective states of the two-atom system: the double atomic ground state $|g\rangle = |g_1\rangle|g_2\rangle$ and the double atomic excited state $|e\rangle = |e_1\rangle|e_2\rangle$, which are also product states of the individual atomic states. These states are not affected by the dipole-dipole interaction Ω_{12} , the detuning Δ and the spontaneous emission rates.

Here, we discuss a method of preparing a two atom system in entangled states involving only the double atomic ground $|g\rangle$ and excited $|e\rangle$ states

$$|\Upsilon\rangle = c_g|g\rangle \pm c_e|e\rangle \quad (92)$$

where c_g and c_e are constant parameters such that $|c_g|^2 + |c_e|^2 = 1$. The entangled states of the form (92) are known in the literature as *pairwise atomic states* [22] or *multiatom squeezed states* [23]. According to Eq. (36), the collective ground and excited states are separated in energy by $2\hbar\omega_0$, and therefore we can call the states $|\Upsilon\rangle$ as two-photon entangled (TPE) states.

The two-photon entangled states cannot be generated by a simple coherent excitation. A coherent field applied to the two-atom system couples to one-photon transitions. The problem is that coherent excitation populates the upper state $|e\rangle$ but also populated the intermediate states $|s\rangle$ and $|a\rangle$. The two-photon entangled states (92) are superpositions of the collective ground and excited states with no contribution from the intermediate collective states $|s\rangle$ and $|a\rangle$.

The two-photon behavior of the entangled states (92) suggests that the simplest technique for generating the TPE states would be by applying a two-photon excitation process. An obvious candidate is a squeezed vacuum field, which is characterized by strong two-photon correlations that would enable the transition $|g\rangle \rightarrow |e\rangle$ to occur effectively in a single step without populating the intermediate states. We will illustrate this effect by analyzing the populations of the collective atomic states.

A. Two Atoms in a Squeezed Vacuum

The dynamics of the collective two-atom system in a squeezed vacuum can be determined from the master equation of the density operator of the system or from the equations of motion for the transition probability amplitudes [22]. In Section II.B, we derived the master equation for the density operator of a two atom system interacting with the ordinary vacuum field. It is our purpose to extend the master equation to the case of a squeezed vacuum field. The method of derivation of the master equation is a straightforward extension of that presented in Section II.B.

The correlation functions for the field operators $\hat{a}_{\mathbf{k}s}$ and $\hat{a}_{\mathbf{k}s}^\dagger$, which describe a three-dimensional field in a squeezed vacuum state, are given by [24,25]

$$\begin{aligned} \langle \hat{a}_{\mathbf{k}s} \rangle &= \langle \hat{a}_{\mathbf{k}s}^\dagger \rangle = 0 \\ \langle \hat{a}_{\mathbf{k}s} \hat{a}_{\mathbf{k}'s'}^\dagger \rangle &= (N(\omega_k) + 1) \delta^3(\mathbf{k} - \mathbf{k}') \delta_{ss'} \\ \langle \hat{a}_{\mathbf{k}s}^\dagger \hat{a}_{\mathbf{k}'s'} \rangle &= N(\omega_k) \delta^3(\mathbf{k} - \mathbf{k}') \delta_{ss'} \\ \langle \hat{a}_{\mathbf{k}s}^\dagger \hat{a}_{\mathbf{k}'s'}^\dagger \rangle &= M(\omega_k) \delta^3(2\mathbf{k}_s - \mathbf{k} - \mathbf{k}') \delta_{ss'} \\ \langle \hat{a}_{\mathbf{k}s} \hat{a}_{\mathbf{k}'s'} \rangle &= M^*(\omega_k) \delta^3(2\mathbf{k}_s - \mathbf{k} - \mathbf{k}') \delta_{ss'} \end{aligned} \quad (93)$$

where $N(\omega_k)$ is the number of photons in the mode ω_k and $M(\omega_k) = M(2\omega_s - \omega_k)$ is the two-photon correlation function, which is symmetric about the squeezing carrier frequency $2\omega_s$. The parameters $N(\omega_k)$ and $M(\omega_k) = |M(\omega_k)|\exp(i\phi)$ are not independent of each other but are related by the inequality

$$|M(\omega_k)|^2 \leq N(\omega_k)(N(2\omega_s - \omega_k) + 1) \quad (94)$$

where the term $+1$ on the r.h.s arises from the quantum nature of the squeezed field [25], and ϕ is the phase of the squeezed vacuum field.

Substituting the interaction Hamiltonian (7), we find that the evolution of the density operator depends on the second order correlation functions of the reservoir operators. We assume that a part of the reservoir modes is in a squeezed vacuum state for which the correlation functions are given by Eq. (93).

In order to optimize the squeezing effects on the atom, the mode function $U_s(\omega_k)$ of the squeezed vacuum field should be perfectly matched to the mode function $\mathbf{g}_{\mathbf{k}s}(\mathbf{r}_i)$ of the three-dimensional vacuum field coupled to the atoms. Such a requirement of the perfect matching is practically impossible to achieve in present experiments [59]. Therefore, we consider mode functions that correspond to an imperfect matching of the squeezing modes to the vacuum modes surrounding the atoms. In this case, we can write the mode function $U_s(\omega_k)$ as

$$U_s(\omega_k) = \begin{cases} [\mathcal{N}(\omega_k)]^{-1/2} \boldsymbol{\mu}_i^* \cdot \mathbf{g}_{\mathbf{k}s}^*(\mathbf{r}_i) D(\omega_k) & \text{for } \theta_k \leq \theta_m \\ 0 & \text{for } \theta_k > \theta_m \end{cases} \quad (95)$$

where $\mathcal{N}(\omega_k)$ is the normalization constant such that $|U_s(\omega_k)|^2 = 1$, the parameter $D(\omega_k)$ determines the coupling efficiency of the squeezed field mode function $U_s(\omega_k)$ to the vacuum field mode function $\mathbf{g}_{\mathbf{k}s}(\mathbf{r}_i)$, and θ_m is the maximum angle over which the squeezed modes are propagated. For perfect coupling efficiency $|D(\omega_k)| = 1$, whereas $|D(\omega_k)| < 1$ for an imperfect coupling. The parameter $D(\omega_k)$ contains both the amplitude and phase coupling, and its explicit form depends on the method of propagation and focusing the squeezed field. For example, in the case of a Gaussian profile of a focused squeezed field, the parameter $D(\omega_k)$ is given by [60,61]

$$D(\omega_k) = \exp[-W_0 \sin^2 \theta_k - ikz_f \cos \theta_k] \quad (96)$$

where W_0 is the beam spot size at the focal point z_f . In a cavity situation, for example, the parameter $D(\omega_k)$ is identified as the cavity transfer function, the absolute value square of which is the Airy function of the cavity [61,62].

Before returning to the derivation of the master equation, we should remark that in the squeezing propagation case in which the squeezed modes lie inside the cone of angle $\theta_m < \pi$, we assume that the modes outside the cone are in their ordinary vacuum state. In practice, the modes will be in a finite-temperature

blackbody state, which means that inside the cone the modes are in mixed squeezed vacuum and blackbody states. However, this is not a serious practical problem as experiments are usually performed at low temperatures where the blackbody radiation is negligible. In principle, we can include the blackbody radiation effect (thermal noise) to the problem replacing $N(\omega_k)$ in (93) by $N(\omega_k) + \bar{N}$, where \bar{N} is proportional to the photon number in the blackbody radiation.

We now return to the derivation of the master equation of the atom in a squeezed vacuum field. Substituting the interaction Hamiltonian (7) into Eq. (18) and using the correlation functions (93), we obtain

$$\begin{aligned} \frac{\partial}{\partial t} \tilde{\rho}(t) = & \sum_{i,j=1}^2 \{ [S_i^- Y_{ij}(t, \tau), S_j^+] + [S_i^-, Y_{ij}(t, \tau) S_j^+] \\ & + [S_i^+ N_{ij}(t, \tau), S_j^-] + [S_i^+, N_{ij}(t, \tau) S_j^-] \\ & + [S_i^+ M_{ij}(t, \tau), S_j^+] + [S_i^+, M_{ij}(t, \tau) S_j^+] \\ & + [S_i^- M_{ij}^*(t, \tau), S_j^-] + [S_i^-, M_{ij}^*(t, \tau) S_j^-] \} \\ & - i \sum_{i \neq j} \Omega_{ij} [S_i^+ S_j^-, \tilde{\rho}(t)] \end{aligned} \quad (97)$$

where Ω_{ij} is given in Eq. (28),

$$\begin{aligned} Y_{ij}(t, \tau) &= \frac{1}{c^3} \int d\omega_k \omega_k^2 [\chi_{ij}^{(N)}(t) + \chi_{ij}(t)] \int_0^t d\tau \tilde{\rho}(t - \tau) e^{i(\omega_i - \omega_k)\tau} \\ N_{ij}(t, \tau) &= \frac{1}{c^3} \int d\omega_k \omega_k^2 \chi_{ij}^{(N)}(t) \int_0^t d\tau \tilde{\rho}(t - \tau) e^{-i(\omega_i - \omega_k)\tau} \\ M_{ij}(t, \tau) &= \frac{1}{c^3} \int d\omega_k \omega_k (2\omega_s - \omega_k) \chi_{ij}^{(M)}(t) \int_0^t d\tau \tilde{\rho}(t - \tau) e^{i(\omega_i - \omega_k)\tau} \end{aligned} \quad (98)$$

with

$$\begin{aligned} \chi_{ij}(t) &= \int d\Omega_k \sum_s [\boldsymbol{\mu}_i \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_i)] [\boldsymbol{\mu}_j^* \cdot \mathbf{g}_{\mathbf{k}s}^*(\mathbf{r}_j)] e^{i(\omega_i - \omega_j)t} \\ \chi_{ij}^{(N)}(t) &= N(\omega_k) |D(\omega_k)|^2 e^{-i(\omega_i - \omega_j)t} \\ &\quad \times \sum_s \int_{\Omega_s} d\Omega_k [\boldsymbol{\mu}_i^* \cdot \mathbf{g}_{\mathbf{k}s}^*(\mathbf{r}_i)] [\boldsymbol{\mu}_j \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_j)] \\ \chi_{ij}^{(M)}(t) &= M(\omega_k) |D(\omega_k)|^2 e^{i(2\omega_0 - \omega_i - \omega_j)t} \\ &\quad \times \sum_s \int_{\Omega_s} d\Omega_k [\boldsymbol{\mu}_i \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_i)] [\boldsymbol{\mu}_j \cdot \mathbf{g}_{\mathbf{k}s}(\mathbf{r}_j)] \end{aligned} \quad (99)$$

and Ω_s is the solid angle over which the squeezed modes are propagated.

The master equation (97) with parameters (98) is quite general in terms of the matching of the squeezed modes to the vacuum modes and the bandwidth of the squeezed field relative to the atomic linewidths. The master equation is in the

form of a differential integral equation, and we can simplify the form employing the Markov approximation. In this approximation the integral over the time delay τ contains functions that decay to zero over a short correlation time τ_c . This correlation time is of the order of the inverse bandwidth of the squeezed field, and the short correlation time approximation is formally equivalent to assume that squeezing bandwidths are much larger than the atomic linewidths. Over this short timescale, the density operator would hardly have changed from $\rho(t)$, thus we can replace $\tilde{\rho}(t - \tau)$ by $\tilde{\rho}(t)$ in (98) and extend the integral to infinity. Next, we can perform the integration and find

$$\lim_{t \rightarrow \infty} \int_0^t d\tau \tilde{\rho}(t - \tau) e^{i\tau} \approx \tilde{\rho}(t) \left[\pi \delta(x) + i \frac{P}{x} \right] \quad (100)$$

where P indicates the principal value of the integral. Finally, to carry out the polarization sums and integrals over $d\Omega_k$ in (99), we use the plane-wave description of the vacuum mode function $\mathbf{g}_{\mathbf{k}s}(\mathbf{r})$, and assuming that the dipole moments are parallel ($\boldsymbol{\mu}_1 \parallel \boldsymbol{\mu}_2$), the sums over s and the integrals over $d\Omega_k$ in (99) lead to

$$\begin{aligned} Y_{ij}(t, \tau) &= \Gamma_{ij} [1 + N(\omega_i) |D(\omega_i)|^2 v(\theta_m)] \tilde{\rho}(t) e^{i(\omega_i - \omega_j)t} \\ N_{ij}(t, \tau) &= \Gamma_{ij} N(\omega_i) |D(\omega_i)|^2 v(\theta_m) \tilde{\rho}(t) e^{-i(\omega_i - \omega_j)t} \\ M_{ij}(t, \tau) &= \Gamma_{ij} M(\omega_i) |D(\omega_i)|^2 v(\theta_m) \tilde{\rho}(t) e^{i(2\omega_s - \omega_i - \omega_j)t} \end{aligned} \quad (101)$$

where

$$v(\theta_m) = \frac{1}{2} \left[1 - \frac{1}{4} (3 + \cos^2 \theta_m) \cos \theta_m \right] \quad (102)$$

In the derivation of (101), we have ignored the principal value parts that contribute to the energy shifts of the atomic levels. In fact, the shifts are very small for a broadband squeezed field, and their contribution to the atomic dynamics are negligible [63].

With the parameters (101), the master equation of two atoms in a broadband squeezed vacuum, written in the Schrödinger picture, reads as

$$\begin{aligned} \frac{\partial \rho}{\partial t} &= -\frac{1}{2} \sum_{i,j=1}^2 \Gamma_{ij} [1 + \tilde{N}(\omega_i)] (\rho S_i^+ S_j^- + S_i^+ S_j^- \rho - 2 S_j^- \rho S_i^+) \\ &\quad - \frac{1}{2} \sum_{ij} \Gamma_{ij} \tilde{N}(\omega_i) (\rho S_i^- S_j^+ + S_i^- S_j^+ \rho - 2 S_j^+ \rho S_i^-) \\ &\quad - \frac{1}{2} \sum_{ij} \Gamma_{ij} \tilde{M}(k_i) ([S_i^+ \rho, S_j^+] + [S_j^+, \rho S_i^+]) \\ &\quad - \frac{1}{2} \sum_{ij} \Gamma_{ij} \tilde{M}^*(\omega_i) ([S_i^- \rho, S_j^-] + [S_j^-, \rho S_i^-]) - \frac{i}{\hbar} [H', \rho] \end{aligned} \quad (103)$$

where

$$\begin{aligned}\tilde{N}(\omega_i) &= \eta N(\omega_i) \\ \tilde{M}(\omega_i) &= \eta M(\omega_i)\end{aligned}\quad (104)$$

with $\eta = |D(\omega_i)|^2 v(\theta_m)$, and the Hamiltonian H' as given in Eq. (32).

The parameter η determines the matching of the squeezed modes to the modes surrounding the atom. This includes the coupling efficiency of the mode functions, given by the parameter $D(\omega_k)$, and the angular dimensions, given by the angle θ_m , of the squeezed modes. For an imperfect matching the master equation (103) is formally identical to that for perfect matching: the only difference is the replacement of the squeezing parameters N and M by the matching modified parameters \tilde{N} and \tilde{M} . The master equation (103) is a starting equation to calculate the stationary state of a two-atom system interacting with a squeezed vacuum field.

B. Steady-State Populations

In order to calculate the stationary state of the two-atom system, we have to know the steady-state populations ρ_{ii} of the collective atomic states and the coherencies ρ_{ij} ($i \neq j$). First, we consider a system of two identical atoms ($\Delta = 0, \Gamma_1 = \Gamma_2$) separated by an arbitrary distance r_{12} and interacting with a squeezed vacuum field. Moreover, we assume that the carrier frequency ω_s of the squeezed vacuum field is resonant to the atomic transition frequencies ($\omega_s = \omega_0$).

From the master equation (103), we find equations of motion for the populations of the collective atomic states, which in the absence of the coherent driving field ($\Omega_1 = \Omega_2 = 0$) can be written as

$$\begin{aligned}\frac{\partial}{\partial t} \rho_{ee} &= -\Gamma(n+1)\rho_{ee} + \frac{1}{2}(n-1)[(\Gamma + \Gamma_{12})\rho_{ss} + (\Gamma - \Gamma_{12})\rho_{aa}] \\ &\quad + \Gamma_{12}(\tilde{M}^* \rho_{eg} + \tilde{M} \rho_{ge}) \\ \frac{\partial}{\partial t} \rho_{ss} &= \frac{1}{2}(\Gamma + \Gamma_{12})\{(n-1) - (3n-1)\rho_{ss} - (n-1)\rho_{aa} + 2\rho_{ee} \\ &\quad - 2(\tilde{M}^* \rho_{eg} + \tilde{M} \rho_{ge})\} \\ \frac{\partial}{\partial t} \rho_{aa} &= \frac{1}{2}(\Gamma - \Gamma_{12})\{(n-1) - (3n-1)\rho_{aa} - (n-1)\rho_{ss} + 2\rho_{ee} \\ &\quad + 2(\tilde{M}^* \rho_{eg} + \tilde{M} \rho_{ge})\} \\ \frac{\partial}{\partial t} \rho_{eg} &= \left(\frac{\partial}{\partial t} \rho_{ge}\right)^* = \Gamma_{12}\tilde{M} - n\Gamma\rho_{eg} \\ &\quad - \tilde{M}[(\Gamma + 2\Gamma_{12})\rho_{ss} - (\Gamma - 2\Gamma_{12})\rho_{aa}]\end{aligned}\quad (105)$$

where $n = 2\tilde{N}(\omega_0) + 1$ and $\tilde{M} = \tilde{M}(\omega_0)$.

It is seen from Eq. (105) that the evolution of the populations depends on the two-photon coherencies ρ_{eg} and ρ_{ge} , which can transfer the population from the ground state $|g\rangle$ directly to the upper state $|e\rangle$ leaving the states $|s\rangle$ and $|a\rangle$ unpopulated. The evolution of the populations depends on Γ_{12} , but is completely independent of the dipole-dipole interaction Ω_{12} .

There are two different steady-state solutions of Eqs. (105) depending on whether $\Gamma_{12} = \Gamma$ or $\Gamma_{12} \neq \Gamma$. This fact is connected with the existence of a combination of the density matrix elements involving the antisymmetric state

$$S^2(t) = 2 - 2\rho_{aa}(t) \quad (106)$$

which, for $\Gamma_{12} = \Gamma$ is a constant of motion. In this case the population in the antisymmetric state does not change in time. Thus an initially unpopulated antisymmetric state remains unpopulated for all times, and then the population is distributed only between three collective states $|e\rangle$, $|s\rangle$, and $|g\rangle$.

Assuming that $\Gamma_{12} = \Gamma$ and setting the left-hand side of Eqs. (105) equal to zero, we obtain the steady-state solutions for the populations of the states $|e\rangle$ and $|s\rangle$, and the two-photon coherence $|\rho_{eg}|$. A straightforward algebraic manipulation of Eqs. (105) leads to the following steady-state solutions

$$\begin{aligned} \rho_{ee} &= \frac{n(n-1)^2 - 4(n-2)|\tilde{M}|^2}{n(3n^2 + 1 - 12|\tilde{M}|^2)} \\ \rho_{ss} &= \frac{(n^2 - 1) - 4|\tilde{M}|^2}{3n^2 + 1 - 12|\tilde{M}|^2} \\ \rho_u &= \frac{8|\tilde{M}|}{n(3n^2 + 1 - 12|\tilde{M}|^2)} \end{aligned} \quad (107)$$

where $\rho_u = \rho_{eg}e^{-i\phi} + \rho_{ge}e^{i\phi}$.

The steady-state populations depend on the squeezing correlations M and the coupling efficiency η . For a classical squeezed field with the maximal correlations $M = N$, the steady-state populations reduces to

$$\begin{aligned} \rho_{ss} &= \frac{\eta N}{3\eta N + 1} \\ \rho_{ee} &= \frac{\eta^2 N^2}{(\eta N + 1)(3\eta N + 1)} \end{aligned} \quad (108)$$

In this case both the excited states are populated and the populations obey a Boltzmann distribution with $\rho_{gg} > \rho_{ss} > \rho_{ee}$.

The population distribution is qualitatively different for a quantum squeezed field with $|M|^2 = N(N+1)$. In this case the populations are given by

$$\begin{aligned}\rho_{ss} &= \frac{\eta N(1-\eta)}{3\eta N(1-\eta) + 1} \\ \rho_{ee} &= \frac{\eta N[1 + 2(1-\eta)N]}{(2\eta N + 1)[3\eta(1-\eta)N + 1]}\end{aligned}\quad (109)$$

Clearly, the population in the symmetric state can be reduced to zero. This happens for $\eta = 1$, that is, when the squeezed field is perfectly matched to the atoms. In this case the population is distributed only between the ground state $|g\rangle$ and the upper state $|e\rangle$.

The issue we are interested in concerns the final state of the system and its purity. To answer this question, we apply the steady-state solutions (107) and find the stationary density matrix of the system

$$\rho = \begin{pmatrix} \rho_{gg} & 0 & \rho_{ge} \\ 0 & \rho_{ss} & 0 \\ \rho_{eg} & 0 & \rho_{ee} \end{pmatrix} \quad (110)$$

where ρ_{ij} are the nonzero steady-state density matrix elements.

It is evident from Eq. (110) that in the squeezed vacuum the density matrix of the system is not diagonal, due to the presence of the two-photon coherencies ρ_{ge} and ρ_{eg} . In this case the collective states $|g\rangle$, $|s\rangle$ and $|e\rangle$ are no longer eigenstates of the system. The density matrix can be re-diagonalized by including ρ_{eg} and ρ_{ge} to give the new (entangled) states

$$\begin{aligned}|\Upsilon_1\rangle &= [(P_1 - \rho_{ee})|g\rangle + \rho_{eg}|e\rangle] / [(P_1 - \rho_{ee})^2 + |\rho_{eg}|^2]^{1/2} \\ |\Upsilon_2\rangle &= [\rho_{ge}|g\rangle + (P_2 - \rho_{gg})|e\rangle] / [(P_2 - \rho_{gg})^2 + |\rho_{eg}|^2]^{1/2} \\ |\Upsilon_3\rangle &= |s\rangle\end{aligned}\quad (111)$$

where the diagonal probabilities are

$$\begin{aligned}P_1 &= \frac{1}{2}(\rho_{gg} + \rho_{ee}) + \frac{1}{2}[(\rho_{gg} - \rho_{ee})^2 + 4|\rho_{eg}|^2]^{1/2} \\ P_2 &= \frac{1}{2}(\rho_{gg} + \rho_{ee}) - \frac{1}{2}[(\rho_{gg} - \rho_{ee})^2 + 4|\rho_{eg}|^2]^{1/2} \\ P_3 &= \rho_{ss}\end{aligned}\quad (112)$$

In view of Eq. (111), it is easy to see that the squeezed vacuum causes the system to decay into entangled states $|\Upsilon_i\rangle$, which are linear superpositions of the collective ground state $|g\rangle$ and the upper state $|e\rangle$. The intermediate symmetric state remains unchanged under the squeezed vacuum excitation. In general, the states (111) are mixed states. However, for perfect coupling of the squeezed vacuum to the atoms ($\eta = 1$) and $|M|^2 = N(N+1)$ the populations P_2 and P_3 are zero, leaving the population only in the state $|\Upsilon_1\rangle$. Hence, in the limit of perfect coupling $\eta = 1$ the state $|\Upsilon_1\rangle$ is a pure state of the system of two atoms driven by a squeezed vacuum field. From Eqs. (111), we find that the pure entangled state $|\Upsilon_1\rangle$ is given by

$$|\Upsilon_1\rangle = \frac{1}{\sqrt{2n}} [\sqrt{n+1}|g\rangle + \sqrt{n-1}|e\rangle] \quad (113)$$

The pure state (113) is nonmaximally entangled state; it reduces to a maximally entangled state for $N \gg 1$. The entangled state is analogous to the pairwise atomic state [22] or the multiatom squeezed state [23], (see also Ref. 24), predicted in the small sample model of two coupled atoms.

C. Effect of the Antisymmetric State on the Purity of the System

The preparation of a two-atom system in the pure entangled state $|\Upsilon_1\rangle$ requires perfect matching of the squeezed modes to the atoms and interatomic separations much smaller than the optical wavelength. To achieve perfect matching ($\eta = 1$), it is necessary to squeeze of all the modes to which the atoms are coupled: that is, the squeezed modes must occupy the whole 4π solid angle of the space surrounding the atoms. This is not possible to achieve with the present experiments in free space, and in order to avoid the difficulty cavity environments have been suggested [59]. Inside a cavity the atoms interact strongly only with the privileged cavity modes. By the squeezing of these cavity modes, which occupy only a small solid angle about the cavity axis, it would be possible to achieve perfect matching of the squeezed field to the atoms.

However, it is difficult experimentally to fulfil the second requirement that interatomic separations should be much smaller than the resonant wavelength. In fact, present atom trapping and cooling techniques can trap two atoms only within distances of the order of a resonant wavelength [11–13]. It is therefore of interest to examine the effect of increasing the interatomic separation so that the simple three-state representation of two atoms, presented in the preceding section, eventually ceases to be valid. With a finite interatomic separation, the two-atom system is represented by the full four-level system of (35).

With the interatomic separation included, the antisymmetric state $|a\rangle$ fully participates in the dynamics of the two-atom system. In this case $\Gamma_{12} \neq \Gamma$ and

the steady-state solutions of Eqs. (105) are

$$\begin{aligned}
 \rho_{ee} &= \frac{(n-1)^2}{4n^2} + \frac{a^2|\tilde{M}|^2(2n-1)}{Q} \\
 \rho_{ss} &= \frac{(n^2-1)}{4n^2} - \frac{a|\tilde{M}|^2(2n^2-a)}{Q} \\
 \rho_{aa} &= \frac{(n^2-1)}{4n^2} + \frac{a|\tilde{M}|^2(2n^2+a)}{Q} \\
 \rho_u &= \frac{2an^3|\tilde{M}|}{Q}
 \end{aligned} \tag{114}$$

where

$$Q = n^2[n^4 + 4|\tilde{M}|^2(a^2 - n^2)] \tag{115}$$

This result shows that the antisymmetric state is populated in the steady-state even for small interatomic separations ($a \approx 1$). For large interatomic separations $a \approx 0$, and then the symmetric and antisymmetric states are equally populated. When the interatomic separation decreases, the population of the state $|a\rangle$ increases, whereas the population of the state $|s\rangle$ decreases and $\rho_{ss} = 0$ for very small interatomic separations. In Fig. 8, we plot the steady-state populations as a function of the interatomic separation. We see that the collective states are unequally populated and for small r_{12} the state $|a\rangle$ is the most populated state of the system, whereas the state $|s\rangle$ is not populated.

However, the vanishing of the population in the state $|s\rangle$ does not mean that the system is in a pure TPE state. This is due to the presence of the antisymmetric state $|a\rangle$ which is significantly populated for small interatomic separations. To show this, we calculate the quantity

$$\text{Tr}(\rho^2) = \rho_{gg}^2 + \rho_{ss}^2 + \rho_{aa}^2 + \rho_{ee}^2 + |\rho_u|^2 \tag{116}$$

which determines the purity of the system. $\text{Tr}(\rho^2) = 1$ corresponds to a pure state of the system, while $\text{Tr}(\rho^2) < 1$ corresponds to a mixed state. $\text{Tr}(\rho^2) = \frac{1}{4}$ describes a completely mixed state of the system. In Fig. 9, we display $\text{Tr}(\rho^2)$ as a function of the interatomic separation r_{12} for perfect matching $\eta = 1$, $|\tilde{M}|^2 = N(N+1)$, and various N . Clearly, the system is in a mixed state independent of the interatomic separation. Moreover, the purity decreases as N increases.

For small interatomic separation, the mixed state of the system is composed of two states: the TPE state $|\Upsilon_1\rangle$ and the antisymmetric state $|a\rangle$. We can

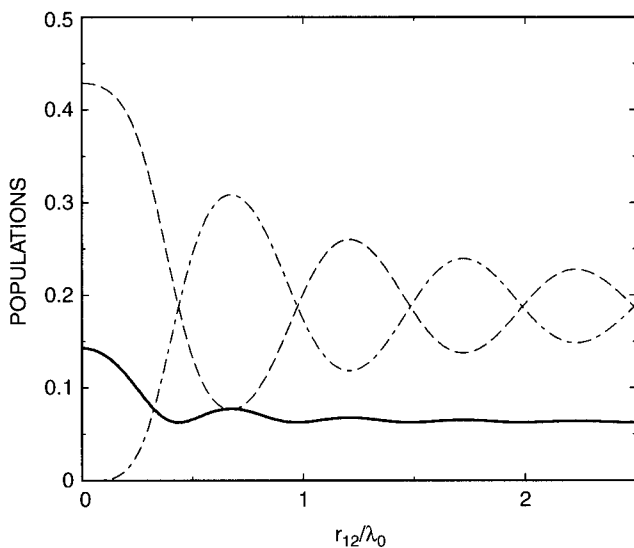


Figure 8. The steady-state populations of the collective atomic states as a function of the interatomic separation for $\eta = 1$, $|M|^2 = N(N+1)$, $N = 0.5$, $\bar{\mu} \perp \bar{r}_{12}$ and ρ_{ee} (solid line), ρ_{aa} (dashed line), ρ_{vv} (dashed-dotted line).

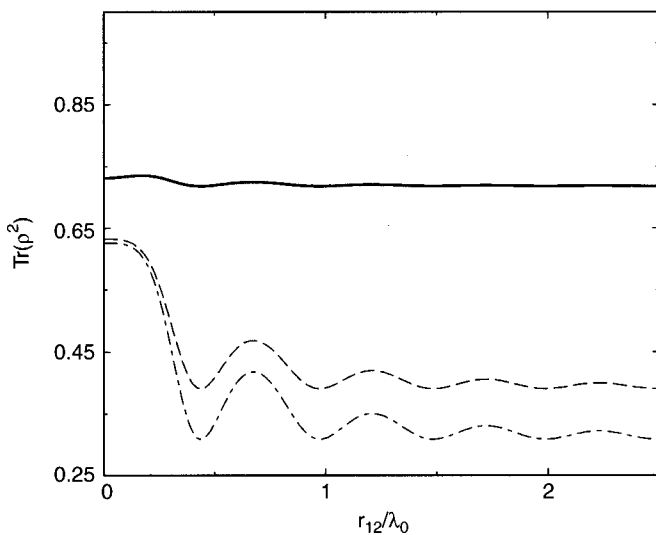


Figure 9. $\text{Tr}(\rho^2)$ as a function of the interatomic separation for $\eta = 1$, $|M|^2 = N(N+1)$, $\bar{\mu} \perp \bar{r}_{12}$ and different N : $N = 0.1$ (solid line), $N = 0.5$ (dashed line), $N = 1$ (dashed-dotted line).

illustrate this by diagonalizing the steady-state density matrix of the system

$$\rho = \begin{pmatrix} \rho_{gg} & 0 & 0 & \rho_{ge} \\ 0 & \rho_{aa} & 0 & 0 \\ 0 & 0 & \rho_{ss} & 0 \\ \rho_{eg} & 0 & 0 & \rho_{ee} \end{pmatrix} \quad (117)$$

When we diagonalize the matrix (117), we find the new (entangled) states

$$\begin{aligned} |\Upsilon_1\rangle &= \frac{(P_1 - \rho_{ee})|g\rangle + \rho_{eg}|e\rangle}{[(P_1 - \rho_{ee})^2 + |\rho_{eg}|^2]^{1/2}} \\ |\Upsilon_2\rangle &= \frac{\rho_{ge}|g\rangle + (P_2 - \rho_{gg})|e\rangle}{[(P_2 - \rho_{gg})^2 + |\rho_{eg}|^2]^{1/2}} \\ |\Upsilon_3\rangle &= |s\rangle \\ |\Upsilon_4\rangle &= |a\rangle \end{aligned} \quad (118)$$

where the diagonal probabilities (populations of the entangled states) are

$$\begin{aligned} P_1 &= \frac{1}{2}(\rho_{gg} + \rho_{ee}) + \frac{1}{2}[(\rho_{gg} - \rho_{ee})^2 + 4|\rho_{eg}|^2]^{1/2} \\ P_2 &= \frac{1}{2}(\rho_{gg} + \rho_{ee}) - \frac{1}{2}[(\rho_{gg} - \rho_{ee})^2 + 4|\rho_{eg}|^2]^{1/2} \\ P_3 &= \rho_{ss} \\ P_4 &= \rho_{aa} \end{aligned} \quad (119)$$

Note that the states $|\Upsilon_1\rangle$, $|\Upsilon_2\rangle$ and $|\Upsilon_3\rangle$ are the same as for the small sample model, discussed in the preceding section. This means that the presence of the antisymmetric state does not affect the two-photon entangled states, but it can affect the population distribution between the states and the purity of the system. In Fig. 10, we plot the populations P_i of the states $|\Upsilon_i\rangle$ as a function of the interatomic separation. The figure demonstrates that the atoms are driven into a mixed state composed of two states $|\Upsilon_1\rangle$ and $|a\rangle$, and there is a vanishing probability that the system is in the states $|\Upsilon_2\rangle$ and $|s\rangle$.

However, the system can decay to the pure TPE state $|\Upsilon_1\rangle$ with the interatomic separation included, provided the observation time is shorter than Γ^{-1} . The antisymmetric state $|a\rangle$ decays on a time scale $\sim(\Gamma - \Gamma_{12})^{-1}$, and for $\Gamma_{12} \approx \Gamma$ the decay rate of the antisymmetric state is much longer than Γ^{-1} . By contrast, the state $|s\rangle$ decays on a time scale $\sim(\Gamma + \Gamma_{12})^{-1}$, which for $\Gamma_{12} \approx \Gamma$ is shorter than Γ^{-1} . Clearly, for observation times shorter than Γ^{-1} , the antisymmetric state does not participate in the interaction and the system reaches the steady-state only between the triplet states. Thus, for perfect

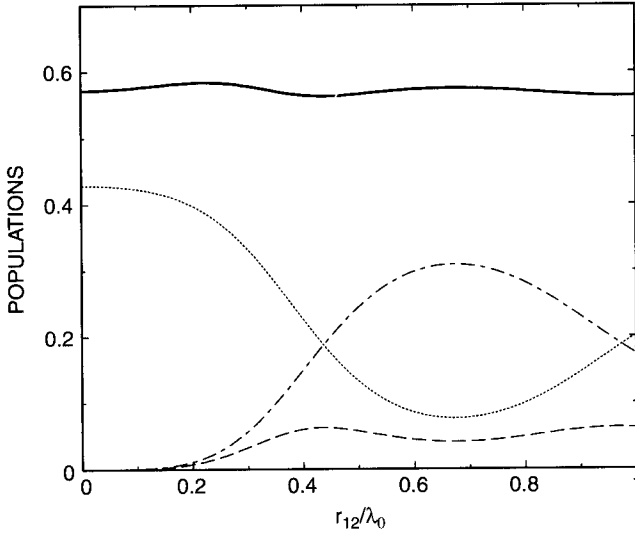


Figure 10. The populations P_i of the entangled states (115) as a function of the interatomic separation for $\eta = 1$, $|M|^2 = N(N+1)$, $\vec{\mu} \perp \vec{r}_{12}$ and $N = 0.5$; P_1 (solid line), P_2 (dashed line), P_3 (dashed-dotted line), P_4 (dotted line).

matching of the squeezed modes to the atoms the symmetric state is not populated and then the system is in the pure TPE state $|\Upsilon_1\rangle$.

D. Two-Photon Entangled States for Two Nonidentical Atoms

By employing two nonidentical atoms of significantly different transition frequencies ($\Delta \gg \Gamma$), it is possible to achieve the pure TPE state with the interatomic separation comparable to the resonant wavelength, and the anti-symmetric state fully participating in the interaction.

Assuming that $\Delta \neq 0$, the master equation (103) leads to the following equations of motion for the density matrix elements

$$\begin{aligned} \frac{\partial}{\partial t} \rho_{ee} &= -\Gamma(n+1)\rho_{ee} + \frac{1}{2}(n-1)[\Gamma(\rho_{ss} + \rho_{aa}) + \Gamma_{12}(\rho_{ss} - \rho_{aa})e^{i\Delta t}] \\ &\quad + \Gamma_{12}|\tilde{M}|(\rho_{eg}e^{-i[2(\omega_s - \omega_0)t + \phi]} + \rho_{ge}e^{i[2(\omega_s - \omega_0)t + \phi]}) \\ \frac{\partial}{\partial t} \rho_{ss} &= \frac{1}{2}(\Gamma + \Gamma_{12}e^{i\Delta t})[(n-1) - (3n-1)\rho_{ss} - (n-1)\rho_{aa} + 2\rho_{ee}] \\ &\quad - \Gamma|\tilde{M}|(\rho_{eg}e^{-i[2(\omega_s - \omega_1)t + \phi]} + \rho_{ge}e^{i[2(\omega_s - \omega_1)t + \phi]}) \\ &\quad - \Gamma_{12}|\tilde{M}|(\rho_{eg}e^{-i[2(\omega_s - \omega_0)t + \phi]} + \rho_{ge}e^{i[2(\omega_s - \omega_0)t + \phi]}) \end{aligned}$$

$$\begin{aligned}
\frac{\partial}{\partial t} \rho_{aa} &= \frac{1}{2} (\Gamma - \Gamma_{12} e^{i\Delta t}) [(n-1) - (3n-1)\rho_{aa} - (n-1)\rho_{ss} + 2\rho_{ee}] \\
&\quad + \Gamma |\tilde{M}| (\rho_{eg} e^{-i[2(\omega_s - \omega_1)t + \phi]} + \rho_{ge} e^{i[2(\omega_s - \omega_1)t + \phi]}) \\
&\quad - \Gamma_{12} |\tilde{M}| (\rho_{eg} e^{-i[2(\omega_s - \omega_0)t + \phi]} + \rho_{ge} e^{i[2(\omega_s - \omega_0)t + \phi]}) \\
\frac{\partial}{\partial t} \rho_{eg} &= \left(\frac{\partial}{\partial t} \rho_{ge} \right)^* = \Gamma_{12} \tilde{M} e^{i[2(\omega_s - \omega_1)t + \phi]} - n\Gamma \rho_{eg} \\
&\quad - \Gamma \tilde{M} e^{i[2(\omega_s - \omega_1)t + \phi]} (\rho_{ss} - \rho_{aa}) \\
&\quad + 2\Gamma_{12} \tilde{M} e^{i[2(\omega_s - \omega_0)t + \phi]} (\rho_{ss} + \rho_{aa})
\end{aligned} \tag{120}$$

where $\omega_0 = \frac{1}{2}(\omega_1 + \omega_2)$.

Equations (120) contain time-dependent terms that oscillate at frequencies $\exp(\pm i\Delta t)$ and $\exp[\pm 2i(\omega_s - \omega_0)t + \phi]$. If we tune the squeezed vacuum field to the middle of the frequency difference between the atomic frequencies, namely, $\omega_s = (\omega_1 + \omega_2)/2$, the terms proportional to $\exp[\pm 2i(\omega_s - \omega_0)t + \phi]$ become stationary in time. None of the other time-dependent components is resonant with the frequency of the squeezed vacuum field. Consequently, for $\Delta \gg \Gamma$, the time-dependent components oscillate rapidly in time and average to zero over long times. Therefore, we can formulate a secular approximation in which we ignore the rapidly oscillating terms, and find that Eqs. (120) give us the following steady-state solutions [64]:

$$\begin{aligned}
\rho_{ee} &= \frac{1}{4} \left[\frac{(n-2)}{n} + \frac{1}{(n^2 - 4a^2 |\tilde{M}|^2)} \right] \\
\rho_{ss} = \rho_{aa} &= \frac{1}{4} \left[1 - \frac{1}{(n^2 - 4a^2 |\tilde{M}|^2)} \right] \\
\rho_u &= \frac{2a |\tilde{M}|}{n(n^2 - 4a^2 |\tilde{M}|^2)}
\end{aligned} \tag{121}$$

These equations are quite different from Eqs. (114) that in the case of non-identical atoms the symmetric and antisymmetric states are equally populated independent of the interatomic separation. These are, however, similar to the steady-state solutions for the small sample model that for small interatomic separations $\rho_{ss} = \rho_{aa} \approx 0$ and then only the collective ground and the upper states are populated.

E. Mapping of the Entanglement of Light on Atoms

The generation of the pure TPE state is an example of mapping of a state of quantum correlated light onto an atomic system. The two-photon correlations

contained in the squeezed vacuum field can be completely transferred to the atomic system. It is seen from Eq. (121) that the collective damping parameter $a(a = \Gamma_{12}/\sqrt{\Gamma_1\Gamma_2})$ plays the role of a degree of the correlation transfer from the squeezed vacuum to the atomic system. For large interatomic separations, $\Gamma_{12} \approx 0$, and there is no transfer of the correlations to the system. In contrast, for very small separations, $\Gamma_{12} \approx \Gamma$, and then the correlations are completely transferred to the atomic system.

However, the complete transfer of the correlations does not necessary mean that the two-photon correlations are stored in the pure TPE state. This happens only for the small sample model and two atoms with significantly different transition frequencies, where the steady state is the pure TPE state. For identical atoms separated by a finite distance r_{12} , a part of the correlations is stored in the antisymmetric state. This fact can lead to an interesting modification of the interference pattern of the fluorescence field. Using the steady-state solutions (114), we find that the visibility in the interference pattern is given by [58]

$$\mathcal{V} = -\frac{4a\eta^2|M|^2}{n^3(n-1) + 4\eta^2|M|^2(a+n-n^2)} \quad (122)$$

This visibility is negative, indicating that the squeezing correlations stored in the antisymmetric state generate an interference pattern with a dark center. In Fig. 11, we plot the visibility \mathcal{V} as a function of the interatomic separation for

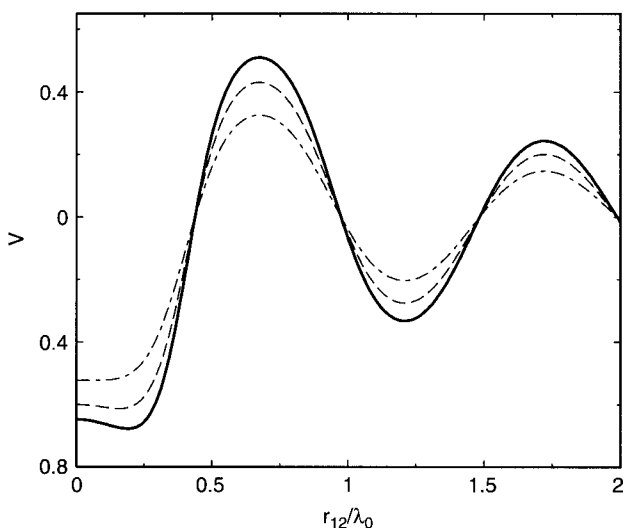


Figure 11. The visibility \mathcal{V} as a function of the interatomic separation for $\eta = 1$, $|M|^2 = N(N+1)$, $\vec{\mu} \perp \vec{r}_{12}$ and different N : $N = 0.1$ (solid line), $N = 0.5$ (dashed line), $N = 5$ (dashed-dotted line).

$\eta = 1$ and $|M| = \sqrt{N(N+1)}$. An interference pattern with a dark center is observed for small interatomic separations ($r_{12} < \lambda_0$) with the maximal negative value $\mathcal{V} \approx -\frac{1}{2}$. The value $\mathcal{V} \approx -\frac{1}{2}$ compared to the possible negative value $\mathcal{V} = -1$ indicates that 50% of the squeezing correlations are stored in the antisymmetric state. Thus, the visibility can be used to measure the degree of correlations stored in the entangled state $|a\rangle$.

The two-photon correlations stored in the pure TPE state can be measured by detecting fluctuations of the fluorescence field emitted by the atomic system. Squeezing in the fluorescence field is proportional to the squeezing in the atomic dipole operators (squeezing in the atomic spins) which, on the other hand, can be found from the steady-state solutions for the density matrix elements.

The fluctuations of the electric field are determined by the normally ordered variance of the field operators as

$$\langle :(\Delta E_\theta)^2: \rangle = \sum_{\mathbf{k}s} E_k \langle 2\langle a_{\mathbf{k}s}^\dagger a_{\mathbf{k}s} \rangle + \langle a_{\mathbf{k}s} a_{\mathbf{k}s} \rangle e^{2i\theta} + \langle a_{\mathbf{k}s}^\dagger a_{\mathbf{k}s}^\dagger \rangle e^{-2i\theta} \rangle \quad (123)$$

Using the correlation functions (93) and choosing $\theta = \pi/2$, the variance of the incident squeezed vacuum field can be written as

$$\langle :(\Delta E_{\pi/2}^v)^2: \rangle = 2E_0(N - |M|) \quad (124)$$

where E_0 is a constant.

Since $|M| = \sqrt{N(N+1)}$, the variance (124) is negative, indicating that we have a squeezed field.

On the other hand, the normally ordered variance of the emitted fluorescence field can be expressed in terms of the density matrix elements of the two-atom system as

$$\langle :(\Delta E_\theta^F)^2: \rangle = E_0(2\rho_{ss} + 2\rho_{ee} + |\rho_u| \cos 2\theta) \quad (125)$$

Using the steady-state solutions (107) and choosing $\theta = \pi/2$, we find

$$\langle :(\Delta E_{\pi/2}^F)^2: \rangle = 2E_0 \frac{(N - |M|)}{2N + 1} \quad (126)$$

Thus, at low intensities of the squeezed vacuum field ($N \ll 1$) the fluctuations in the incident squeezed vacuum field are perfectly mapped onto the atomic system. For large intensities ($N > 1$), the thermal fluctuations of the atomic dipoles dominate over the squeezed fluctuations, resulting in a reduction of squeezing in the fluorescence field.

Kozhokin et al. [38] proposed a method of mapping of quantum states onto an atomic system based on the stimulated Raman absorption of propagating quantum light by a cloud of three-level atoms. Hald et al. [40] have experimentally observed the squeezed spin states of a system of three-level atoms driven by a squeezed field. The observed squeezed spin states have been generated via entanglement exchange with the squeezed field completely absorbed in the process. Fleishhauer et al. [39] have considered a similar system of three-level atoms and have found that quantum states of single-photon fields can be mapped onto collective states of the atomic system. In this case the quantum state of the field is stored in a dark state of the collective states of the system.

Acknowledgments

This work was supported by the Australian Research Council. We gratefully acknowledge helpful conversations with Stuart Swain and Almut Beige.

References

1. R. H. Dicke, *Phys. Rev.* **93**, 99 (1954).
2. G. S. Agarwal, *Phys. Rev. A* **2**, 2038 (1970).
3. R. Bonifacio and L. A. Lugiato, *Phys. Rev. A* **11**, 1507 (1975).
4. M. Gross and S. Haroche, *Phys. Rep.* **93**, 301 (1982).
5. J. H. Eberly and N. E. Rehler, *Phys. Rev. A* **2**, 1607 (1970).
6. C. Greiner, B. Boggs, and T. W. Mossberg, *Phys. Rev. Lett.* **85**, 3793 (2000).
7. R. H. Lehmburg, *Phys. Rev. A* **2**, 883, 889 (1970).
8. G. S. Agarwal, in G. Höhler (Ed.), *Quantum Statistical Theories of Spontaneous Emission and Their Relation to Other Approaches*, Springer Tracts in Modern Physics, Vol. 70, Springer-Verlag, Berlin, 1974.
9. P. W. Milonni and P. L. Knight, *Phys. Rev. A* **10**, 1096 (1974).
10. Z. Ficek, R. Tanaś, and S. Kielich, *Physica* **146A**, 452 (1987).
11. U. Eichmann, J. C. Bergquist, J. J. Bollinger, J. M. Gilligan, W. M. Itano, D. J. Wineland, and M. G. Raizen, *Phys. Rev. Lett.* **70**, 2359 (1993).
12. R. G. DeVoe and R. G. Brewer, *Phys. Rev. Lett.* **76**, 2049 (1996).
13. D. J. Berkeland, in R. Blatt, J. Eschner, D. Leibfried, and F. Schmidt-Kaler (Eds.), *Laser Spectroscopy* World Scientific, Singapore, 1999, p. 352.
14. M. B. Plenio and V. Vedral, *Contemp. Phys.* **39**, 431 (1998).
15. A. Barenco, *Contemp. Phys.* **37**, 375 (1996).
16. L. K. Grover, *Phys. Rev. Lett.* **79**, 325 (1997).
17. A. Barenco and A. Ekert, *J. Mod. Opt.* **42**, 1253 (1995).
18. S. F. Pereira, Z. Y. Ou, and H. J. Kimble, *Phys. Rev. A* **62**, 042311 (2000).
19. A. Ekert, *Phys. Rev. Lett.* **67**, 661 (1991).
20. J. J. Bollinger, W. M. Itano, D. J. Wineland, and D. J. Heinzen, *Phys. Rev. A* **54**, R4649 (1996); S. F. Huelga, C. Macchiavello, T. Pellizzari, A. K. Ekert, M. B. Plenio, and J. I. Cirac, *Phys. Rev. Lett.* **79**, 3865 (1997).

21. M. J. Stephen, *J. Chem. Phys.* **40**, 669 (1964).
22. G. M. Palma and P. L. Knight, *Phys. Rev. A* **39**, 1962 (1989); A. K. Ekert, G. M. Palma, S. M. Barnett, and P. L. Knight, *Phys. Rev. A* **39**, 6026 (1989); G. S. Agarwal and R. R. Puri, *Phys. Rev. A* **41**, 3782 (1990); Z. Ficek, *Phys. Rev. A* **44**, 7759 (1991).
23. S. M. Barnett and M. A. Dupertuis, *J. Opt. Soc. Am. B* **4**, 505 (1987).
24. A. S. Parkins, in M. Evans and S. Kielich (Eds.), *Modern Nonlinear Optics*, Part II, Wiley, New York, 1993, p. 607.
25. Z. Ficek and P. D. Drummond, *Physics Today* **34** (Sept. 1997); B. J. Dalton, Z. Ficek, and S. Swain, *J. Mod. Opt.* **46**, 379 (1999).
26. S. J. D. Phoenix and S. M. Barnett, *J. Mod. Opt.* **40**, 979 (1993).
27. I. K. Kudravytsev and P. L. Knight, *J. Mod. Opt.* **40**, 1673 (1993).
28. J. I. Cirac and P. Zoller, *Phys. Rev. A* **50**, R2799 (1994).
29. C. C. Gerry, *Phys. Rev. A* **53**, 2857 (1996).
30. M. B. Plenio, S. F. Huelga, A. Beige, and P. L. Knight, *Phys. Rev. A* **59**, 2468 (1999).
31. A. Beige, S. F. Huelga, P. L. Knight, M. B. Plenio, and R. C. Thompson, *J. Mod. Opt.* **47**, 401 (2000).
32. A. Beige, W. J. Munro, and P. L. Knight, *Phys. Rev. A* **62**, 052102 (2000).
33. U. Akram, Z. Ficek, and S. Swain, *Phys. Rev. A* **62**, 013413 (2000).
34. A. Beige, D. Braun, B. Tregenna, and P. L. Knight, *Phys. Rev. Lett.* **85**, 1762 (2000).
35. Q. A. Turchette, C. S. Wood, B. E. King, C. J. Myatt, D. Leibfried, W. M. Itano, C. Monroe, and D. J. Wineland, *Phys. Rev. Lett.* **81**, 3631 (1998).
36. E. Hagley, X. Maitre, G. Nogues, C. Wunderlich, M. Brune, J. M. Raimond, and S. Haroche, *Phys. Rev. Lett.* **79**, 1 (1997).
37. A. Kuzmich, L. Mandel, and N. P. Bigelow, *Phys. Rev. Lett.* **85**, 1594 (2000).
38. A. E. Kozhekin, K. Molmer, and E. Polzik, *Phys. Rev. A* **62**, 033809 (2000).
39. M. Fleischhauer, S. F. Yelin, and M. D. Lukin, *Opt. Commun.* **179**, 395 (2000).
40. J. Hald, J. L. Sorensen, L. Leich, and E. S. Polzik, *Opt. Express* **2**, 93 (1998); J. Hald, J. L. Sorensen, C. Schori, and E. S. Polzik, *Phys. Rev. Lett.* **83**, 1319 (1999).
41. W. H. Louisell, *Statistical Properties of Radiation*, Wiley, New York, 1973.
42. L. Allen and J. H. Eberly, *Resonance Fluorescence and Two-Level Atoms*, Wiley, New York, 1975.
43. Z. Ficek, R. Tanaś, and S. Kielich, *Optica Acta* **33**, 1149 (1986).
44. G. J. Yang, O. Zobay, and P. Meystre, *Phys. Rev. A* **59**, 4012 (1999).
45. T. Richter, *Optica Acta* **29**, 265 (1982).
46. Z. Ficek, R. Tanaś, and S. Kielich, *Optica Acta* **30**, 713 (1983).
47. T. Richter, *Optica Acta* **30**, 1769 (1983).
48. T. G. Rudolph, Z. Ficek, and B. J. Dalton, *Phys. Rev. A* **52**, 636 (1995).
49. H. S. Freedhoff, *Phys. Rev. A* **26**, 684 (1982).
50. Z. Ficek and B. C. Sanders, *Phys. Rev. A* **41**, 359 (1990).
51. S. Franke, G. Huyet, and S. M. Barnett, *J. Mod. Opt.* **47**, 145 (2000).
52. H. T. Dung and K. Ujihara, *Phys. Rev. Lett.* **84**, 254 (2000).
53. G. C. Guo and C. P. Yang, *Physica A* **260**, 173 (1998); C. P. Yang and G. C. Guo, *Physica A* **273**, 352 (1999).

54. T. Wong, S. M. Tan, M. J. Collett, and D. F. Walls, *Phys. Rev. A* **55**, 1288 (1997).
55. P. Kochan, H. J. Carmichael, P. R. Morrow, and M. G. Raizen, *Phys. Rev. Lett.* **75**, 45 (1995).
56. G. M. Meyer and G. Yeoman, *Phys. Rev. Lett.* **79**, 2650 (1997).
57. T. Rudolph and Z. Ficek, *Phys. Rev. A* **58**, 748 (1998).
58. Z. Ficek and R. Tanaś, *Opt. Commun.* **153**, 245 (1998).
59. Q. A. Turchette, N.P. Georgiades, C. J. Hood, H. J. Kimble, and A. S. Parkins, *Phys. Rev. A* **58**, 4056 (1998); N.P. Georgiades, E. S. Polzik, and H. J. Kimble, *Phys. Rev. A* **59**, 123 (1999).
60. Z. Ficek and P. D. Drummond, *Europhys. Lett.* **24**, 455 (1993).
61. A. Yariv, *Quantum Electronics*, Wiley, New York, 1989.
62. M. Born and E. Wolf, *Principles of Optics*, Macmillan, New York, 1964, Chapter 7.
63. Z. Ficek and P. D. Drummond, *Phys. Rev. A* **43**, 6247 (1991); **43**, 6258 (1991).
64. Z. Ficek and M. R. B. Wahiddin, *Opt. Commun.* **134**, 387 (1997).