Ultranarrow Spectral Lines via Quantum Interference

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(Received 18 April 1996)

We show that ultrasharp spectral lines, with widths 2 orders of magnitude below the natural width, may be produced in the resonance fluorescence of a V-type three-level atom excited by a single-mode laser field when the dipole moments are nearly parallel. The smaller the splitting of the excited doublet, the narrower the line. This effect is due to quantum interference between the two transition pathways. [S0031-9007(96)01561-X]

PACS numbers: 42.50.Gy, 32.80.Bx, 42.50.Lc

Quantum interference, one of the basic features of quantum mechanics, lies at the heart of many new effects and applications of quantum optics which have been reported recently. In this Letter we show that ultrasharp lines (less than 1% of the natural width) can be produced in the resonance fluorescence spectra of a three-level atom through the mechanism of quantum interference.

The basic system consists of a singlet state connected to a closely spaced doublet by a single-mode laser. The resurgence of interest in quantum interference stems from the work of Harris and co-workers [1], who considered V-type systems where the excited doublets decay to an additional continuum or to a single auxiliary level, in addition to the ground state. They found that at a certain frequency the absorption rate goes to zero due to destructive interference, whereas the emission rate remains finite. It is possible to amplify a laser field at this frequency without population inversion being present [2]. The first observations of lasing without inversion have recently been reported [3]. In the case of a single auxiliary level, quantum interference can lead to the elimination of the spectral line at the driving laser frequency in the spontaneous emission spectrum [4]. Fluorescence quenching has been recently observed in sodium dimers involving transitions with parallel dipole moments by Xia *et al.* [5].

Various related effects such as electromagnetically induced transparency [6], enhancement of the index of refraction without absorption [7], and modification of the spontaneous emission spectrum [8] in Λ and Ξ atomic systems featuring dark lines and very narrow structures, have been extensively studied.

Here we consider a V-type atom consisting of two excited sublevels coupled to a singlet ground level by a single-mode laser field as shown in Fig. 1. This was studied by Cardimona, Raymer, and Stroud [9], who found that the system can be driven into a dark state in which quantum interference prevents any fluorescence from the excited sublevels, regardless of the intensity of the excited laser. Similar predictions were reported by Scully *et al.* [10], who showed that if the two upper levels of a V-type quantum beat laser are coupled by a microwave field, an atom in the ground state may absorb pump photons and at the same time be free of radiative decay, even if population inversion occurs. The macroscopic dark periods of fluorescence emissions due to quantum interference in such a system have been demonstrated by Hegerfeldt and Plenio [11] using quantum jump theory. Striking experimental observations of destructive interference effects in three-level atoms have been reported in thin systems [12]: The first observation in thick systems was by Boller *et al.* in [6]. (See also [13].)

In this paper we concentrate on the effect of quantum interference on the spectral features of the resonance fluorescence emissions. The fluorescent emission is completely quenched at all frequencies under conditions of maximum quantum interference, if the detuning satisfies condition (6) of [9] ("the quenching condition"). If the quantum interference is not quite maximal, we find that a very sharp line occurs at line center, over a wide range of Rabi frequencies and detunings. The Rabi frequency does, however, need to exceed the doublet splitting. A physical understanding of these results may be obtained by invoking the dressed atom approach. According to this theory, the width of the central sharp line goes to zero with the square of the ratio of the excited doublet splitting to the Rabi frequency. Thus extremely sharp lines, less than 1% of the natural linewidth, are possible.

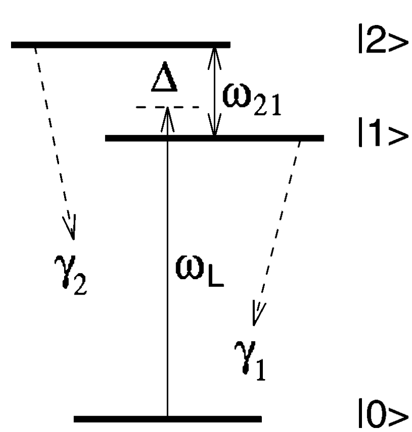

FIG. 1. A V-atom driven by a single-mode laser.

The Hamiltonian in the frame rotating with the laser frequency ω_L is of the form

$$H = (\Delta - \omega_{21})A_{11} + \Delta A_{22} + [(\Omega_1 A_{10} + \Omega_2 A_{20}) + \text{H.c.}], \tag{1}$$

where we use units such that $\hbar = 1$. Here $\Delta = (E_2 - E_0) - \omega_L$ is the detuning between the $|0\rangle \leftrightarrow |2\rangle$ transition and the driving laser $\Omega_k = E_L \mathbf{e}_L \cdot \mathbf{d}_{k0}$ ($k = 1, 2$) denotes the Rabi frequencies, E_L is the laser field amplitude, $\mathbf{d}_{k0}$ is the dipole moment of the atomic transition from $|0\rangle$ to $|k\rangle$, which is assumed to be real, $\omega_{21} = E_2 - E_1$ is the level splitting between the excited sublevels $|1\rangle$ and $|2\rangle$, and $A_{lk} \equiv |l\rangle\langle k|$ represents a population operator when $l = k$ and a dipole transition operator for $l \neq k$. (We note that $\langle A_{kl}\rangle = \rho_{lk}$, where ρ denotes the density matrix of the system.) Direct transitions between the excited sublevels $|1\rangle$ and $|2\rangle$ are dipole forbidden.

The equations of motion of the reduced density matrix elements for the atomic variables take the form [9]

$$\begin{aligned}
\dot\rho_{11} &= -\gamma_1\rho_{11} - \frac{1}{2}\gamma_{12}(\rho_{12} + \rho_{21}) - i\Omega_1(\rho_{01} - \rho_{10}), \quad \dot\rho_{22} = -\gamma_2\rho_{22} - \frac{1}{2}\gamma_{12}(\rho_{12} + \rho_{21}) - i\Omega_2(\rho_{02} - \rho_{20}),\\
\dot\rho_{10} &= -\left[\frac{1}{2}\gamma_1 + i(\Delta - \omega_{21})\right]\rho_{10} - \frac{1}{2}\gamma_{12}\rho_{20} + i\Omega_2\rho_{12} + i\Omega_1(\rho_{11} - \rho_{00}),\\
\dot\rho_{20} &= -\left(\frac{1}{2}\gamma_2 + i\Delta\right)\rho_{20} - \frac{1}{2}\gamma_{12}\rho_{10} + i\Omega_1\rho_{21} + i\Omega_2(\rho_{22} - \rho_{00}),\\
\dot\rho_{21} &= -\left[\frac{1}{2}(\gamma_1 + \gamma_2) + i\omega_{21}\right]\rho_{21} - \frac{1}{2}\gamma_{12}(\rho_{22} + \rho_{11}) + i\Omega_1\rho_{20} - i\Omega_2\rho_{01},
\end{aligned} \tag{2}$$

where γ_k is the spontaneous decay constant of the excited sublevel k ($k = 1, 2$) to the ground level $|0\rangle$. However, γ_{12} represents the effect of quantum interference resulting from the cross coupling between the transitions $|1\rangle \leftrightarrow |0\rangle$ and $|2\rangle \leftrightarrow |0\rangle$. The effects of quantum interference are very sensitive to the orientations of the atomic dipole polarizations. For example, if $\mathbf{d}_{10}$ is parallel to $\mathbf{d}_{20}$, then $\gamma_{12} = \sqrt{\gamma_1\gamma_2}$ and the interference effect is maximal, while if $\mathbf{d}_{10}$ is perpendicular to $\mathbf{d}_{20}$, then $\gamma_{12} = 0$ and the quantum interference disappears. Quantum interference plays a crucially important role in the spectral narrowing and fluorescence quenching in the system considered here.

The fluorescence emission spectrum is proportional to the Fourier transform of the steady-state correlation function $\lim_{t\to\infty}\langle \mathbf{E}^{(-)}(\mathbf{r}, \tau + t) \cdot \mathbf{E}^{(+)}(\mathbf{r}, t)\rangle$, where $\mathbf{E}^{(\pm)}(\mathbf{r}, t)$ are the positive and negative frequency parts of the radiation field in the far zone, which consists of a free-field operator, and a source-field operator that is proportional to the atomic polarization operator. The fluorescence emission spectrum $G(\omega)$ is composed of coherent and incoherent components. The coherent Rayleigh part, due to the elastic scattering of the driving field, gives rise to only δ-function contributions, while the incoherent one, which stems from the fluctuations of the dipole polarizations, makes the main contribution. Hereafter we pay attention only to the incoherent resonance fluorescence spectrum, defined as

$$G_{\text{inc}}(\omega) = \Re \int_0^\infty \lim_{t\to\infty} \langle \Delta\mathbf{D}^\dagger(\tau + t) \cdot \Delta\mathbf{D}(t)\rangle e^{-i\omega\tau} d\tau, \tag{3}$$

where $\Delta\mathbf{D}(t) = \mathbf{D}(t) - \langle\mathbf{D}(\infty)\rangle$ represents the deviation of the dipole polarization operator $\mathbf{D}(t)$ from its mean steady-state value, and

$$\mathbf{D}^\dagger(t) = \mathbf{d}_{10}A_{10}(t) + \mathbf{d}_{20}A_{20}(t). \tag{4}$$

The two-time correlation function $\lim_{t\to\infty}\langle \Delta\mathbf{D}^\dagger(\tau + t) \cdot \Delta\mathbf{D}(t)\rangle$ can be obtained by invoking the quantum regression theorem and making use of the equations of motion (2).

Defining $\alpha = d_{10}/d_{20}$ as the ratio of the transition amplitudes of the two allowed transition pathways, we have the relationships $\gamma_1 = \alpha^2\gamma_2$ and $\Omega_1 = \alpha\Omega_2$. Figs. 2–4 present the results of numerical calculations of the fluorescence spectrum. We consider a few special values of the detuning. First of all, we consider the single-photon resonance $\Delta = 0$ in Fig. 2. In the absence of quantum interference $\gamma_{12} = 0$, as shown in frames (a) and (c), the spectra demonstrate three-peaked and seven-peaked features when the level splitting of the excited doublet is $\omega_{21} = \gamma_2$ and $\omega_{21} = 5\gamma_2$, respectively. However, a remarkably narrow spectral line occurs at line center in the case of maximum quantum interference, when $\gamma_{12} = \sqrt{\gamma_1\gamma_2}$, as shown in the frames (b) and (d).

We next consider the detuning to satisfy the quenching condition $\Delta\Omega_1^2 + (\Delta - \omega_{21})\Omega_2^2 = 0$ [9], as we can obtain analytic solutions for this case. Figure 3 (where $\omega_{21} = \gamma_2$) shows at once that when the atom has nearly parallel or parallel transition dipole moments, the incoherent fluorescence spectrum is dramatically modified by quantum interference. We have $\gamma_{12} = 0.999\sqrt{\gamma_1\gamma_2}$ in Fig. 3(c) and $\gamma_{12} = \sqrt{\gamma_1\gamma_2}$ in Fig. 3(d). In the former situation a significant sharp peak occurs at the line center: For the latter situation, the fluorescent emission is completely quenched. The quantum interference in Figs. 2 and 3 is clearly destructive.

To explore the origin of the unusual spectral features produced by quantum interference, we employ the dressed atomic state representation [14]. Analytic expressions may be obtained when the quenching condition, equivalent to $\Delta = \omega_{21}/(1 + \alpha^2)$, is satisfied, so we concentrate on this case here. For simplicity, we also take the magnitudes of both dipole moments to be identical so that $\gamma_1 = \gamma_2 = \gamma$ and $\Omega_1 = \Omega_2 = \Omega$. In this situation the eigenvalues and

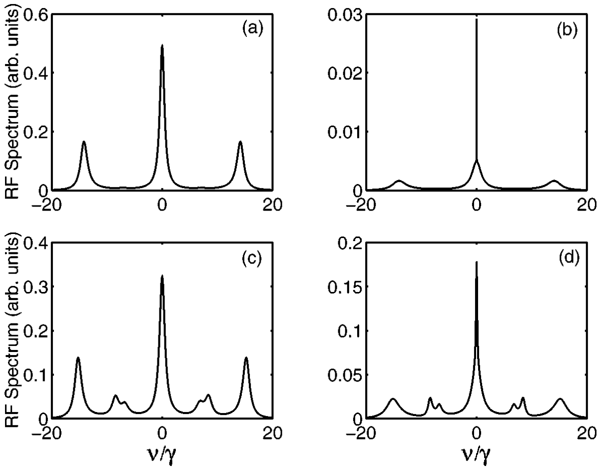

FIG. 2. The incoherent resonance fluorescence spectrum as a function of $\nu = (\omega - \omega_L)/\gamma$ for $\alpha = 1$, $\Omega_1 = \Omega_2 = 5\gamma$, $\Delta = 0$, and different splittings: (a) $\omega_{21} = \gamma$, $\gamma_{12} = 0$, (b) $\omega_{21} = \gamma$, $\gamma_{12} = \gamma$, (c) $\omega_{21} = 5\gamma$, $\gamma_{12} = 0$, (d) $\omega_{21} = 5\gamma$, $\gamma_{12} = \gamma$. Here $\gamma_1 = \gamma_2 = \gamma$.

eigenstates of the interaction Hamiltonian (1) are given by

$$\lambda_a = -\frac{1}{2}\Omega_R, \quad \lambda_b = 0, \quad \lambda_c = \frac{1}{2}\Omega_R, \tag{5}$$

and

$$\begin{aligned} |a\rangle &= \frac{1}{2}[-(1-\varepsilon)|2\rangle - (1+\varepsilon)|1\rangle + 4\eta|0\rangle], \\ |b\rangle &= -2\eta|2\rangle + 2\eta|1\rangle + \varepsilon|0\rangle, \\ |c\rangle &= \frac{1}{2}[(1+\varepsilon)|2\rangle + (1-\varepsilon)|1\rangle + 4\eta|0\rangle], \end{aligned} \tag{6}$$

where $\Omega_R = \sqrt{\omega_{21}^2 + 8\Omega^2}$, $\eta = \Omega/\Omega_R$, and $\varepsilon = \omega_{21}/\Omega_R$.

In the high field limit, where the effective Rabi frequency is much greater than all relaxation rates, i.e., $\Omega_R \gg \gamma$, the equations for the diagonal and off-diagonal elements uncouple. We are interested here in the features at line center, which are governed by the diagonal matrix elements. We find

$$\begin{aligned} \dot\rho_{bb} &\simeq -\Gamma_1\rho_{bb} + \Gamma_0, \\ \dot\rho_{cc} - \dot\rho_{aa} &\simeq -\Gamma_2(\rho_{cc} - \rho_{aa}), \end{aligned} \tag{7}$$

with

$$\begin{aligned} \Gamma_0 &= \frac{1}{2}[(\gamma + \gamma_{12})\varepsilon^2 + (\gamma - \gamma_{12})\varepsilon^4], \\ \Gamma_1 &= \frac{1}{2}[(\gamma + \gamma_{12})\varepsilon^2 + (\gamma - \gamma_{12})(3\varepsilon^4 - 4\varepsilon^2 + 2)], \\ \Gamma_2 &= \frac{1}{2}[(\gamma + \gamma_{12}) + (\gamma - \gamma_{12})\varepsilon^2]. \end{aligned} \tag{8}$$

In this case, the incoherent resonance fluorescence spectrum consists of five spectral components: the central resonance, the inner sidebands placed at frequencies $\pm\Omega_R/2$, and the outer sidebands located at frequencies $\pm\Omega_R$.

In the dressed state representation, the underlying physical processes are evident. The various components in the resonance fluorescence spectrum are associated with different transitions between neighboring dressed state manifolds. The central peak comes from transitions between identical dressed levels of adjacent manifolds and consists of a superposition of two Lorentzians with linewidths $2\Gamma_1$ and $2\Gamma_2$, the decay rates of ρ_{bb} and $\rho_{cc} - \rho_{aa}$, respectively. However, one of the inner sidebands (located at $-\Omega_R/2$) is the result of the transitions from $|a\rangle$ to $|b\rangle$ and from $|b\rangle$ to $|c\rangle$. The other pair of inner sidebands arises from the transitions $|c\rangle \to |b\rangle$ and $|b\rangle \to |a\rangle$. Transitions between the dressed states $|a\rangle$ and $|c\rangle$ contribute the outer sidebands. As the stationary dressed-state populations ρ_{aa} and ρ_{cc} are identical in the secular approximation, the spectrum is symmetric.

The physical mechanisms of fluorescence quenching and spectral narrowing are as follows.

Fluorescence quenching.—If $\gamma_{12} = \sqrt{\gamma_1\gamma_2} = \gamma$, i.e., the dipole moments of the two transitions are exactly parallel, it can be shown from our analytical results that $\rho_{aa} = \rho_{cc} = 0$ and $\rho_{bb} = 1$: The population is trapped in the dressed state $|b\rangle$. As a consequence, there is no photon emission at all, $G_{\rm inc}(\omega) = 0$, as one sees in Fig. 3(d). However, if γ_{12} deviates from the maximum value γ, the destructive interference is incomplete, and the corresponding fluorescence emission can take place.

Spectral narrowing.—It has been shown numerically that an extremely narrow peak may arise at the line center. In order to explore the physical origin of the spectral narrowing, we assume that γ_{12} has a value slightly less than its maximum $\sqrt{\gamma_1\gamma_2}$, so that the dressed states $|a\rangle$ and $|c\rangle$ have a very small, but nonzero, population. Consequently, some fluorescent emission takes place, $G_{\rm inc}(\omega) \neq 0$. The linewidths of the two Lorentzians comprising the central component are $\Gamma_1 \simeq \gamma\varepsilon^2$ and $\Gamma_2 \simeq \gamma$, respectively. If $\varepsilon \ll 1$, which requires that the effective Rabi frequency Ω_R greatly exceed the level splitting ω_{21}, then $\Gamma_1 \ll \gamma$, giving rise to a pronounced and very sharp feature at line

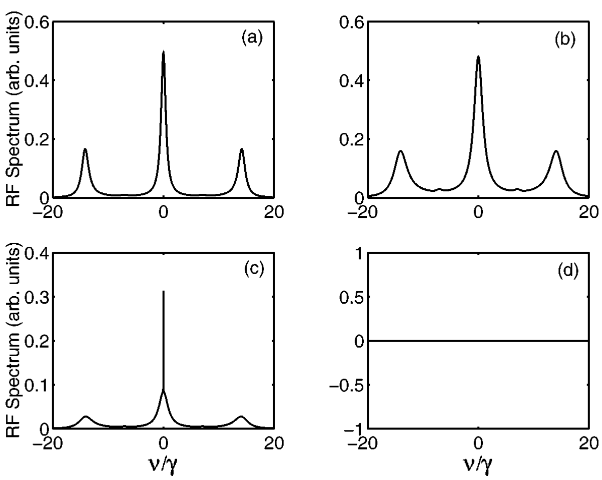

FIG. 3. Same as Fig. 2, but for $\alpha = 1$, $\Omega_1 = \Omega_2 = 5\gamma$, $\omega_{21} = \gamma$, $\Delta = 0.5\gamma$, with different values of γ_{12}: (a) $\gamma_{12} = 0$, (b) $\gamma_{12} = 0.9\gamma$, (c) $\gamma_{12} = 0.999\gamma$, (d) $\gamma_{12} = \gamma$. Here $\gamma_1 = \gamma_2 = \gamma$.

center. Thus the spectral feature at line center consists of a sharp peak superimposed on a broad Lorentzian profile. We emphasize that the sharp peak can be very narrow. For example, the linewidth in Fig. 3(c) is predicted to be $\Gamma_1 \simeq \gamma/200$ by the dressed atom theory, and found to be so numerically. The linewidth of the sharp feature in Fig. 2(b) is also close to this value.

The spectral narrowing is due to the slow decay of the dressed state population ρ_{bb}, and originates from the quantum interference between the two transitions. In fact, when γ_{12} is far from the maximum allowed value γ, the decay rate of the population in the dressed state $|b\rangle$ increases and becomes comparable with Γ_2. In the case of $\gamma_{12} = 0$ (no quantum interference) and $\varepsilon \ll 1$ we have $\Gamma_1 = \gamma, \Gamma_2 = \gamma/2$, and no significant spectral narrowing occurs.

The line narrowing effect occurs over a wide range of detunings and Rabi frequencies, not just the ones chosen in Figs. 2 and 3. We consider a quite different set of parameter values in Fig. 4, by way of illustration. We show the central region of the spectrum as a function of Ω/γ for a larger value of the detuning, $\Delta = 30\gamma$, and for $\omega_{21} = 5\gamma$, in frames (a) and (b), and $\omega_{21} = 20\gamma$ in frames (c) and (d), with $\gamma_{12} = 0.99\gamma$ and 0.999γ. The line narrowing is most pronounced in frame (d), whereas frames (c) and (d) show clearly how the broad natural linewidth apparent for very small Ω evolves rapidly into a much narrower peak as Ω increases.

For the experimental observation of these phenomena, we need a closely spaced excited doublet with almost parallel dipole moments. The realization of such systems has been discussed in some detail in [9]. It is necessary to employ frequency stabilized lasers and to minimize the effects of Doppler broadening. Recently, the $|2s\rangle$ and $|2p\rangle$ states of the hydrogen atom, mixed by an electric field to produce a separation of 40γ, as employed in quantum interference experiments [6], has been suggested [4] as a suitable system for investigating this class of phenomena.

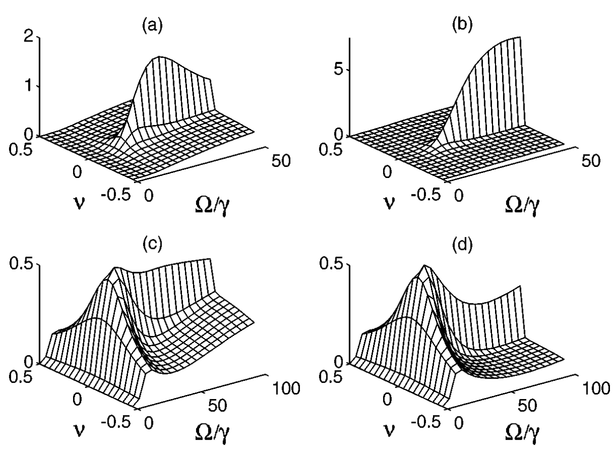

FIG. 4. The incoherent spectrum as a function of ν and Ω/γ, with $\alpha = 1$, $\Delta = 30\gamma$, and $\omega_{21} = 5\gamma$ in (a) and (b), and $\omega_{21} = 20\gamma$ in (c) and (d). We take $\gamma_{12} = 0.99\gamma$ in (a) and (c), and $\gamma_{12} = 0.999\gamma$ in (b) and (d).

With $\Omega = 20\gamma$, for example, we find numerically that only modest line narrowing is possible (by a factor of $\frac{1}{2}$), but with $\Omega = 100\gamma$ and $\Delta = 83\gamma$, pronounced narrowing occurs with a width of the order of $\gamma/20$.

In summary, we have shown that the resonance fluorescence spectrum in a V system is strongly modified by quantum interference. For slightly less than maximum quantum interference, extremely narrow lines may be produced at the center of the spectrum for small splittings of the excited sublevels. The effects may be understood in terms of the trapping and slow decay of the population of one of the dressed states.

This work is supported by the United Kingdom EPSRC, by the EC, and by a NATO Collaborative Research Award. P. Z. wishes to thank the Queen's University of Belfast for financial support.

[1] S. E. Harris, Phys. Rev. Lett. **62**, 1033 (1989); S. E. Harris and J. J. Macklin, Phys. Rev. A **40**, 4135 (1989); A. Imamoğlu, *ibid.* **40**, 2835 (1989).
[2] O. Kocharovskaya and Ya. A. Khanin, JETP Lett. **48**, 630 (1988); A. Imamoğlu, J. E. Field, and S. E. Harris, Phys. Rev. Lett. **66**, 1154 (1991).
[3] A. S. Zibrov, M. D. Lukin, D. E. Nikonov, L. Hollberg, V. L. Velichansky, and H. G. Robinson, Phys. Rev. Lett. **75**, 1499 (1995); G. G. Padmabandu, G. R. Welch, I. N. Shubin, E. S. Fry, D. E. Nikonov, M. D. Lukin, and M. O. Scully, Phys. Rev. Lett. **76**, 2053 (1996).
[4] S. Y. Zhu and M. O. Scully, Phys. Rev. Lett. **76**, 388 (1996); S. Y. Zhu, Quantum Opt. **7**, 385 (1995).
[5] H. R. Xia, C. Y. Ye, and S. Y. Zhu, Phys. Rev. Lett. **77**, 1032 (1996).
[6] K. Hakuta, L. Marmet, and B. P. Stoicheff, Phys. Rev. Lett. **66**, 596 (1991); K. J. Boller, A. Imamoğlu, and S. E. Harris, *ibid.* **66**, 2593 (1991); M. Xiao, Y. Li, S. Jin, and J. Gea–Banacloche, *ibid.* **74**, 666 (1995); J. Gea–Banacloche, Y. Li, S. Jin, and M. Xiao, Phys. Rev. A **51**, 576 (1995); D. J. Fulton, S. Shepherd, R. R. Mosely, B. D. Sinclair, and M. H. Dunn, *ibid.* **52**, 2302 (1995).
[7] M. O. Scully, Phys. Rev. Lett. **67**, 1855 (1991); M. O. Scully and S. Y. Zhu, Opt. Commun. **87**, 134 (1992).
[8] S. Y. Zhu, L. M. Narducci, and M. O. Scully, Phys. Rev. A **52**, 4791 (1995); A. H. Toor, S. Y. Zhu, and M. S. Zubairy, *ibid.* **52**, 4803 (1995).
[9] D. A. Cardimona, M. G. Raymer, and C. R. Stroud, Jr., J. Phys. B **15**, 65 (1982).
[10] M. O. Scully, S. Y. Zhu, and A. Gavrielides, Phys. Rev. Lett. **62**, 2813 (1989).
[11] G. C. Hegerfeldt and M. B. Plenio, Phys. Rev. A **46**, 373 (1992).
[12] H. R. Gray, R. M. Whitely, and C. R. Stroud, Jr., Opt. Lett. **3**, 218 (1978); R. P. Hackel and S. Ezekiel, Phys. Rev. Lett. **42**, 1736 (1979); M. Kaivola, P. Thorsen, and O. Poulsen, Phys. Rev. A **32**, 207 (1985).
[13] Y. Li and M. Xiao, Phys. Rev. A **51**, 4959 (1995).
[14] C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 345 (1977).