Nonclassical interference effects in the radiation from coherently driven uncorrelated atoms

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We demonstrate the existence of nonclassical correlations in the radiation of two atoms, that are coherently driven by a continuous laser source. The photon-photon correlations of the fluorescence light show a spatial interference pattern not present in a classical treatment. A feature of this phenomenon is that bunched and antibunched light is emitted in different spatial directions. The calculations are performed analytically. It is pointed out that the correlations are induced by state reduction due to the measurement process when the detection of the photons does not distinguish between the atoms. It is interesting to note that the phenomena show up even without any interatomic interaction.

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Resonance fluorescence from a single atom driven by a coherent field was the first example allowing observation of nonclassical effects such as antibunching and sub-Poissonian statistics [1–3]. These experiments, first done with atoms in a beam, were later performed using single trapped ions [4,5]. Squeezing in resonance fluorescence has also been investigated [6]. Since the early work of Mollow [7], and Carmichael and Walls [8], the quantum statistical characteristics of radiation produced by a cooperative system of two and more atoms have been studied in much detail [9,10]. There are also some recent proposals to investigate cooperative effects, including the interactions between the atoms [11–13].

In the present paper we study two-atom fluorescence leading to nonclassical effects. We consider a situation where the two atoms are driven coherently by a continuous laser source (Fig. 1). The nonclassical effects are observed in the photon-photon correlations when state reduction occurs in the measurement process. We note that in an earlier study by Mandel [14] correlation effects in a similar system were discussed. However, in that case, in contrast to our paper, no continuously pumped atoms were considered there. We calculate nonclassical two-photon correlations for all times including dissipation of the atoms. The dynamics resulting from excitation by cw fields and by spontaneous emission is very important for the present results. We should mention that on the experimental side first order interference in the radiation produced by a system of two atoms has been reported [15]. We also point out that the creation of entangled states of distant atoms has been discussed in a different context [16].

It may be noted that trapped ion technology is well enough advanced to envisage resonance fluorescence measurements with chains of trapped ions driven by coherent fields. One can also use single molecules in crystalline hosts, where recently nonclassical photon statistics have been measured [17].

We consider two identical atoms with the levels $|e\rangle_i$ and $|g\rangle_i$ $(i=1,2)$ at fixed positions $\mathbf{x}_1$ and $\mathbf{x}_2$ with dipole moment $\mathbf{d}$ and transition frequency ω. They are driven by a resonant external laser field with wave vector $\mathbf{k}_L$. We assume that the only dissipative terms are due to the spontaneous decays of the levels $|e\rangle_{1,2}$. In the rotating-wave, Markov, and Born approximations, the time evolution of the system is given by the master equation [18]

$$\dot{\rho}=-i\Omega\sum_{\mu=1}^{2}[e^{[i(\mathbf{k}_L\cdot\mathbf{x}_\mu-\omega_L t)]}\sigma_\mu^{+}+\text{H.c.},\rho]-\sum_{\mu=1}^{2}\gamma(\sigma_\mu^{+}\sigma_\mu^{-}\rho+\rho\sigma_\mu^{+}\sigma_\mu^{-}-2\sigma_\mu^{-}\rho\sigma_\mu^{+}), \qquad (1)$$

where 2Ω is the Rabi frequency of the atom laser system, $\sigma_\mu^{\pm}$ are the atomic raising and lowering operators for atom number μ, and $2\gamma=\frac{4}{3}|\mathbf{d}|^2\omega^3/\hbar c^3$ is the Einstein A coefficient for the single atom. As we assume that the electromagnetic field outside the laser beam is in the vacuum state, the first and second order correlation functions can be written as

$$G^{(1)}(\mathbf{r},t)=\sum_{\mu,\nu=1}^{2}e^{[ik\mathbf{x}_{\mu,\nu}\cdot\hat{\mathbf{r}}]}\langle\sigma_\mu^{+}(t)\sigma_\nu^{-}(t)\rangle, \qquad (2)$$

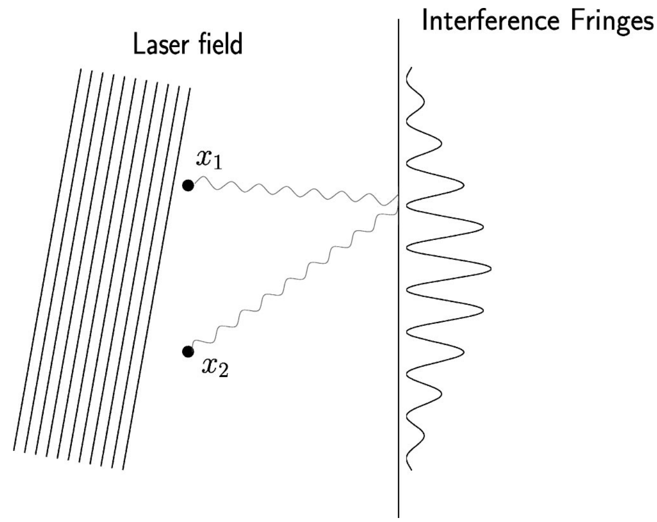

FIG. 1. Two atoms in a coherent laser field show interference fringes in the first order correlation function.

$$G^{(2)}(\mathbf{r}_1,t;\mathbf{r}_2,t+\tau)=\sum_{\mu,\nu,\lambda,\rho=1}^{2} e^{[ik(\mathbf{x}_{\lambda,\nu}\cdot\hat{\mathbf{r}}_1+\mathbf{x}_{\rho,\mu}\cdot\hat{\mathbf{r}}_2)]} \times\langle\sigma_\lambda^+(t)\sigma_\rho^+(t+\tau)\sigma_\mu^-(t+\tau)\sigma_\nu^-(t)\rangle \tag{3}$$

with $\mathbf{x}_{\mu,\nu}:=\mathbf{x}_\mu-\mathbf{x}_\nu$ and $\hat{\mathbf{r}}=\mathbf{r}/|\mathbf{r}|$.

The two-time correlations (3) can be calculated using the quantum regression theorem and the time evolution of the density operator. Therefore the normalized second order correlation function can be written as

$$g^{(2)}(\mathbf{r}_1,t;\mathbf{r}_2,t+\tau)=\frac{P(\mathbf{r}_2,t+\tau|\mathbf{r}_1,t)}{P(\mathbf{r}_2,t)} \tag{4}$$

where $P(\mathbf{r},t)$ is the probability of finding a photon at position $\mathbf{r}$ at time t, and $P(\mathbf{r}_2,t+\tau|\mathbf{r}_1,t)$ the conditional probability of finding a photon at position $\mathbf{r}_2$ at time $t+\tau$ assuming the detection of a photon at point $\mathbf{r}_1$ at time t. Using the inequality

$$\mathrm{Tr}\{\rho[\alpha I(\mathbf{r}_1)+\beta I(\mathbf{r}_2)]^2\}\geqslant 0 \quad \forall \alpha,\beta \tag{5}$$

leads us to

$$\prod_{i=1}^{2}[g^{(2)}(\mathbf{r}_i;\mathbf{r}_i)-1]\geqslant[g^{(2)}(\mathbf{r}_1;\mathbf{r}_2)-1]^2 \tag{6}$$

for classical systems. This gives us a good possibility of estimating the nonclassical behavior of our system.

As one state can always be found that does not interact with the laser field it seems appropriate to use this state to build up the basis for further calculations. We call this state $|a\rangle$ and define

$$|e\rangle:=|e,e\rangle,$$

$$|s\rangle:=\frac{1}{\sqrt{2}}(e^{-i\phi}|e,g\rangle+e^{i\phi}|g,e\rangle),$$

$$|a\rangle:=\frac{1}{\sqrt{2}}(e^{-i\phi}|e,g\rangle-e^{i\phi}|g,e\rangle),$$

$$|g\rangle:=|g,g\rangle, \tag{7}$$

with $|i,j\rangle:=|i\rangle_1\otimes|j\rangle_2$ and $\phi:=\frac{1}{2}\mathbf{k}_L\cdot\mathbf{x}_{12}$.

In this representation (Fig. 2) the master equation reduces to the following set of $6+3$ equations:

$$\dot{\rho}_{ee}=4(\alpha\rho_{es}^i-\rho_{ee}), \tag{8}$$

$$\dot{\rho}_{ss}=2[\rho_{ee}-\rho_{ss}+2\alpha(\rho_{sg}^i-\rho_{es}^i)],$$

$$\dot{\rho}_{aa}=2(\rho_{ee}-\rho_{aa}),$$

$$\dot{\rho}_{es}^i=-3\rho_{es}^i-2\alpha(\rho_{ee}-\rho_{ss}+\rho_{eg}^r),$$

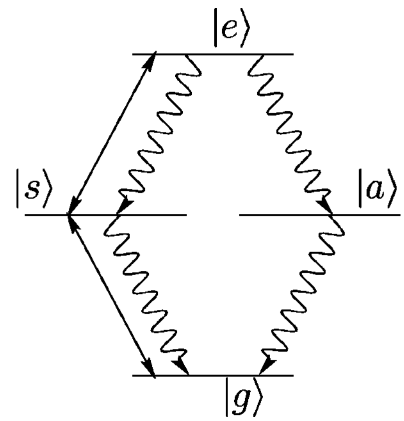

FIG. 2. Level scheme of the atomic system for the symmetrized set of states.

$$\dot{\rho}_{sg}^i=2\rho_{es}^i-\rho_{sg}^i+2\alpha(1-\rho_{ee}-\rho_{aa}-2\rho_{ss}+\rho_{eg}^r),$$

$$\dot{\rho}_{eg}^r=-2[\rho_{eg}^r+\alpha(\rho_{sg}^i-\rho_{es}^i)],$$

$$\dot{\rho}_{ea}^r=-3\rho_{ea}^r-2\alpha\rho_{sa}^i,$$

$$\dot{\rho}_{sa}^i=2[\alpha(\rho_{ea}^r+\rho_{ag}^r)-\rho_{sa}^i],$$

$$\dot{\rho}_{ag}^r=-2\rho_{ea}^r-2\alpha\rho_{sa}^i-\rho_{ag}^r, \tag{9}$$

with $\alpha:=\Omega/\sqrt{2}\gamma$, $\rho_{kl}^r:=\mathrm{Re}(\rho_{kl})$, and $\rho_{kl}^i:=\mathrm{Im}(\rho_{kl})$. By solving the first set of equations (8) we find the diagonal elements in the steady state to be

$$\rho_{gg}^{SS}=\frac{(\gamma^2+\Omega^2)^2}{(\gamma^2+2\Omega^2)^2}, \tag{10}$$

$$\rho_{ss}^{SS}=\frac{\Omega^2[2\gamma^2+\Omega^2]}{(\gamma^2+2\Omega^2)^2}, \tag{11}$$

$$\rho_{aa}^{SS}=\rho_{ee}^{SS}=\frac{\Omega^4}{(\gamma^2+2\Omega^2)^2}. \tag{12}$$

This calculation does not depend on the direction of the driving laser, although it is included by the proper definition of our symmetric and antisymmetric states.

If the laser direction is perpendicular to the atom separation ($\phi=0$) this solution corresponds to the solutions of Richter found earlier [10], where we neglect the dipole-dipole interaction. This is justified when the atom distance is in the range of several wavelengths of the atomic transition or larger. In this case the solutions are completely independent of the separation of the atoms. For strong laser fields ($\Omega\gg\gamma$) the populations are equal for each of the atomic states ($\rho_{gg}^{SS}=\rho_{ss}^{SS}=\rho_{ee}^{SS}=\rho_{aa}^{SS}=1/4$) (Fig. 3).

We next discuss how a detection event leads to state reduction and entanglement. After a detection at the point $\mathbf{r}$ we have to find a new density operator in the following way:

$$\rho(\mathbf{r})=\frac{\sigma^-(\mathbf{r})\rho^{SS}\sigma^+(\mathbf{r})}{\langle\sigma^+(\mathbf{r})\sigma^-(\mathbf{r})\rangle}, \tag{13}$$

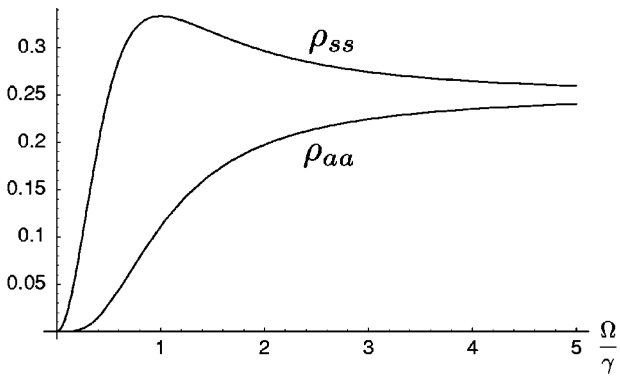

FIG. 3. Steady state populations of the symmetric and antisymmetric states as functions of the laser intensity Ω.

where

$$\sigma^{\pm}(\mathbf{r}) := \sigma_1^{\mp} + e^{ik_L \hat{\mathbf{r}}\cdot\mathbf{x}_{12}}\sigma_2^{\pm}. \tag{14}$$

If, for instance, the system is in state $|e\rangle$ before a detection at $\mathbf{r}$ with $\hat{\mathbf{r}}\cdot\mathbf{x}_{12}=0$, we find it in $|s\rangle$ after the detection. This explains how entanglement can come into the system, as the system is transformed from a nonentangled state $|e\rangle$ to an entangled state $|s\rangle$ by a detection event.

Let us now check if the atoms remain uncorrelated after a detection event, i.e., if $\rho(\mathbf{r})$ factorizes into a product of density matrices ρ_i with ρ_i referring to the density matrix of the ith atom. We find that this is not the case; for example,

$$\mathrm{Im}[\rho(\mathbf{r})_{sa}] = \frac{\Omega^2 \sin\phi}{2\{\Omega^2 + \gamma^2[1+\cos\delta(\mathbf{r})\}}, \quad \rho_{ee}(\mathbf{r}) = 0, \tag{15}$$

with $\delta(\mathbf{r}) = (\mathbf{k}_L - k_L\hat{\mathbf{r}})\cdot\mathbf{x}_{12}$. On the other hand a factorized density matrix would imply that $\mathrm{Im}[\rho(\mathbf{r})_{sa}]=0$ after any detection event. Furthermore, the dependence of $\mathrm{Im}[\rho(\mathbf{r})_{sa}]$ on the coordinates of the two atoms through $\delta(\mathbf{r})$ also points out the nonexistence of a factorized density matrix after detection. Thus there are values of $\mathbf{r}$ where the state reduction by the measurement leaves the system in an entangled state. With this mechanism the entanglement that is necessary for nonclassical effects is simply generated by the detection of a single photon.

To calculate the intensity at position $\mathbf{r}$ we use the relations

$$\langle\sigma_1^+\sigma_1^-\rangle = \langle\sigma_2^+\sigma_2^-\rangle = \rho_{ee} + \frac{1}{2}(\rho_{ss}+\rho_{aa}), \tag{16}$$

$$\langle\sigma_1^+\sigma_2^-\rangle = \langle\sigma_2^+\sigma_1^-\rangle^* = \frac{1}{2}e^{2i\phi}(\rho_{ss}-\rho_{aa}), \tag{17}$$

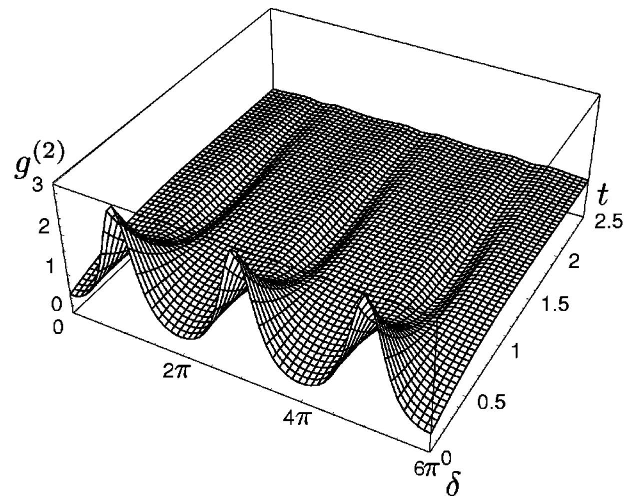

FIG. 4. Second order correlation function for one detector with $\delta_1=\delta_2=\delta$, $\Omega=0.8\gamma$.

where we assumed the system to be in the steady state (i.e., $\rho_{sa}^{SS}=0$). We then find

$$G^{(1)}(\mathbf{r},t) = [1+\cos\delta(\mathbf{r})][\rho_{ee}(t)+\rho_{ss}(t)] + [1-\cos\delta(\mathbf{r})][\rho_{ee}(t)+\rho_{aa}(t)]. \tag{18}$$

This leads to the well known interference fringes in the first order correlation function as studied in the experiment of Eichmann *et al.* [15].

In this case the system behaves as in the well known double slit experiment where the atoms act as slits and the two optical paths interfere. As in a system with prepared initial states, the photons emitted by symmetric transitions $|e\rangle\to|s\rangle$ and $|s\rangle\to|g\rangle$ show inverse fringes compared to those emitted by antisymmetric transitions $|e\rangle\to|a\rangle$ or $|a\rangle\to|g\rangle$. This follows from the $1\pm\cos\delta(\mathbf{r})$ terms in Eq. (18). One major difference is that the contrast depends on the intensity of the laser light so that the fringes disappear for higher laser intensities when $\Omega \gg \gamma$. The populations ρ_{ss}^{SS} and ρ_{aa}^{SS} then equalize.

To get the second order correlation function we solve the master equation by calculating the Liouville operator and use for the initial state the density matrix given by Eq. (13). This procedure is also equivalent to using the quantum regression theorem.

Remarkably enough we are able to give an analytical expression for the intensity-intensity correlation:

$$\begin{aligned} g^{(2)}(\mathbf{r}_1,0;\mathbf{r}_2,\tau) = 1 + \frac{e^{-3t}}{4\nu^2(s+\cos\delta_1)(s+\cos\delta_2)}\Big[& 4e^{2t}\nu^2 s\sin\delta_1\sin\delta_2 + s[e^t\nu^2 s + (s-1)^2]\cos\delta_1\cos\delta_2 \\ & - e^{3t/2}\nu s^2[2\nu\cos(\nu t) + 3\sin(\nu t)] + 2e^{3t/2}\nu s(\cos\delta_1+\cos\delta_2)[(2s-3)\sin(\nu t) \\ & - 2\nu\cos(\nu t)] + e^{t/2}\nu(2e^t\cos\delta_1\cos\delta_2 + s\sin\delta_1\sin\delta_2)[2\nu(s-2)\cos(\nu t) + (5s-6)\sin(\nu t)] \\ & + \frac{1}{4}\cos\delta_1\cos\delta_2(\{s[s(4s-33)+64]-36\}\cos(2\nu t) + 2\nu(s-2)(5s-6)\sin(2\nu t))\Big] \end{aligned} \tag{19}$$

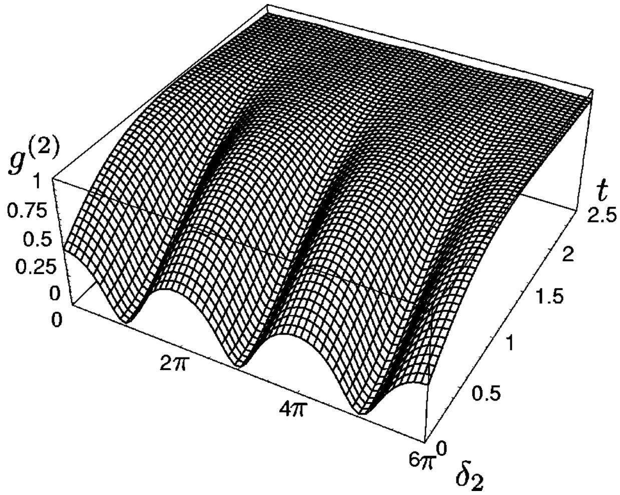

FIG. 5. Second order correlation function for two detectors with $\delta_1 = 2n\pi$, $\Omega = 0.8\gamma$.

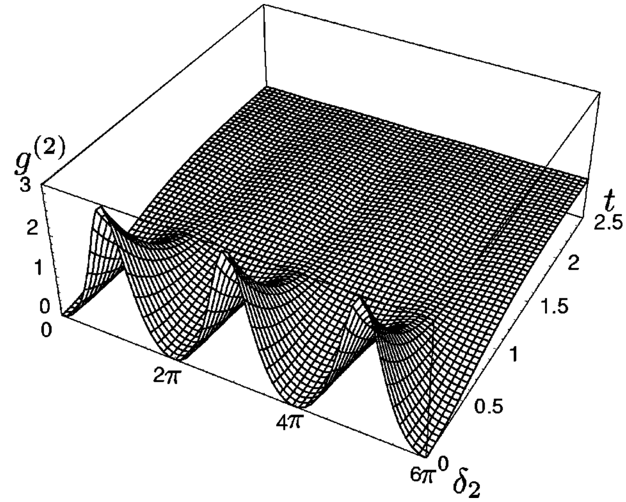

FIG. 6. Second order correlation function for two detectors with $\delta_1 = (2n+1)\pi$, $\Omega = 0.8\gamma$.

with $s := 2(\Omega/\gamma)^2 + 1$, $\delta_i = \delta(\mathbf{r}_i)$, and $\nu := \frac{1}{2}\sqrt{8s-9}$ being parameters for the intensity of the driving field and $t := \gamma\tau$ the time scaled by the atom spontaneous emission rate. For $\tau = 0$ Eq. (19) reduces to

$$g^{(2)}(\mathbf{r}_1,0;\mathbf{r}_2,0) = \frac{s^2 \cos^2[(\delta_1 - \delta_2)/2]}{(s+\cos\delta_1)(s+\cos\delta_2)}. \tag{20}$$

Assuming measurements with only one detector at position $\mathbf{r}$, $g^{(2)}(\mathbf{r},0;\mathbf{r},0)$ shows interferences fringes with maxima larger and minima smaller than 1 (Fig. 4). This means that the system is emitting super-Poissonian light in one and sub-Poissonian light in the other direction. This behavior has no analogy in the first order correlation, i.e., in the two-slit experiment.

For two detectors the situation is different. For any detector position with $\delta_2 = (2n+1)\pi + \delta_1$, $g^{(2)}(\mathbf{r}_1,0;\mathbf{r}_2,0)$ vanishes completely, while at other positions we find maximum detection probability (Fig. 5). After a detection at position $\mathbf{r}_1$ with $\delta_1 = 2n\pi$ we find $\rho_{ee} = \rho_{aa} = 0$. Therefore, there is no probability of detecting any photon at a position with $\delta_2 = (2n+1)\pi$ at the same time, as no emission into the antisymmetric channel can take place (18). So we again find fringes as a function of δ_2. For $\delta_1 = (2n+1)\pi$ we detect an emission in the antisymmetric channel first, so we find $g^{(2)}(\mathbf{r}_1,0;\mathbf{r}_2,0)$ vanishing for positions $\mathbf{r}_2$ with $\delta_2 = 2n\pi$ for the same reasons as above (Fig. 6).

After any detection in a specific channel the system needs time for reexcitation to emit to the orthogonal channel; on the other hand, cascade emissions can take place only in the same channel. Note that the behavior of $g^{(2)}(\mathbf{r}_1,0;\mathbf{r}_2,\tau)$ is well understood in terms of our symmetrized basis (7). These results show also that the inequality (6) could be violated by choosing the two detector positions in such a way that $g^{(2)}(\mathbf{r}_1,0;\mathbf{r}_1,0) > 1$ and $g^{(2)}(\mathbf{r}_2,0;\mathbf{r}_2,0) < 1$. In this manner we find nonclassical behavior in our system as a consequence of the detection induced entanglement of the two atoms. We note that dependence of the type $\cos^2$ in two-photon correlations was also derived by Mandel in his work on radiation from atoms prepared in atomic coherent states [14]. In contrast to that, we are dealing here with the more general case of continuous excitation of the atoms including dissipation.

In conclusion, it has been shown by analytic calculations that a system of two atoms coherently excited by a cw laser source shows nonclassical features that are dependent on the distance between the atoms, although there is no assumed interaction between the particles. This can be understood by measurement induced entanglement, which comes into the system only if the detection does not distinguish between the two atoms, i.e., the detected photon does not carry which-way information. At first glance the system studied seems to be similar to the two-slit experiment; however, the nonclassical effects discussed here do not show up in the first order correlation and can be seen only in the second order correlation, as should be the case for a quantum phenomenon.

To see the reported effects single ions stored in a linear rf trap can be used; the remaining micromotion could be overcome by phase-sensitive detection [19]. In this paper the case was discussed where the detector or detectors do not discriminate the light emitted by individual atoms. There are, however, further interesting phenomena observable when the which-way information is available. One example is that the excitation of one atom and the selective observation of the fluorescence from the other one opens, among other effects, the possibility of investigating the dipole-dipole interaction between the atoms. These results will be described elsewhere.

[1] H. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. Lett. **39**, 691 (1977).
[2] R. Short and L. Mandel, Phys. Rev. Lett. **51**, 384 (1983).
[3] J. D. Cresser, J. Häger, G. Leuchs, F.-M. Rateike, and H. Walther, *Dissipative Systems in Quantum Optics*, Vol. 27 of *Topics in Current Physics* (Springer, Berlin, 1982).
[4] F. Diedrich and H. Walther, Phys. Rev. Lett. **58**, 203 (1987).
[5] H. Schubert, I. Siemers, R. Blatt, W. Neuhauser, and P.

Toschek, Phys. Rev. Lett. **68**, 3016 (1992).
[6] Z. Lu, S. Bali, and J. Thomas, Phys. Rev. Lett. **81**, 3635 (1998).
[7] B. Mollow, Phys. Rev. **188**, 1969 (1969).
[8] H. Carmichael and D. Walls, J. Phys. B **9**, 1199 (1976).
[9] G. S. Agarwal, A. Brown, L. Narducci, and G. Vetri, Phys. Rev. A **15**, 1613 (1977).
[10] For a good discussion on interference effects in the fluorescence of two atoms, see T. Richter, Opt. Commun. **80**, 285 (1991).
[11] R. DeVoe and R. Brewer, Phys. Rev. Lett. **76**, 2049 (1996).
[12] T. Rudolph, Z. Ficek, and B. Dalton, Phys. Rev. A **52**, 636 (1995).
[13] A. Beige and G. Hegerfeldt, Phys. Rev. A **58**, 4133 (1998); **59**, 2385 (1999).
[14] L. Mandel, Phys. Rev. A **28**, 929 (1983).
[15] U. Eichmann, J. Bergquist, J. Bollinger, J. Gilligan, W. Itano, D. Wineland, and M. Raizen, Phys. Rev. Lett. **70**, 2359 (1993). For theoretical discussions related to this experiment, see, for example, T. Wong *et al.*, Phys. Rev. A **55**, 1288 (1997); W. M. Itano *et al.*, *ibid.* **57**, 4176 (1998); C. Schön and A. Beige, *ibid.* **64**, 023806 (2001). The paper by Schön and Beige also calculates $g^{(2)}(r_{1,0};\, r_{1,0})$ for measurements with a single detector.
[16] C. Cabrillo, J. Cirac, P. Garcia-Fernandez, and P. Zoller, Phys. Rev. A **59**, 1025 (1999).
[17] W. Moerner and M. Orrit, Science **283**, 1670 (1999); L. Fleury, J. Segura, G. Zumofen, B. Hecht, and U. Wild, Phys. Rev. Lett. **84**, 1148 (2000).
[18] G. S. Agarwal, *Quantum Optics*, Vol. 70 of *Springer Tracts in Modern Physics* (Springer, Berlin, 1974).
[19] J. Höffges, H. Baldauf, T. Eichler, S. Helmfrid, and H. Walther, Opt. Commun. **133**, 170 (1997).