Cooperative Quantum Jumps with Two Atoms

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Quantum jumps in the fluorescence from two three-level atoms may display cooperative effects, such as unusual intensity levels and transitions from a state with both atoms radiating directly to darkness. However, to see these experimentally, the atoms have to be well within a wavelength of light.

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Atomic spectroscopy has reached the ultimate sensitivity with the progress in ion trapping and laser cooling techniques, in that one ion can be isolated and subjected to continuous observation for a period of hours. This state of affairs naturally promotes studies of the basic light-matter interactions, such as recent "quantum jump"[1] and photon antibunching[2] observations. By the same token it is possible to experiment on samples with a known small number of ions close enough to one another that each one senses the field radiated by the others. Cooperative effects may have to be considered, such as few-atom precursors of superradiance[3,4] as well as modified photon statistics[5] and resonance fluorescence spectrum.[6] It is also conceivable that collective interactions of the ions with light affect the quantum jumps. In fact, an anomalously high probability for quantum jumps involving simultaneously more than one ion in two- and three-ion clouds has been reported in one experiment,[7] whereas in another such correlations were not found.[8]

In this Letter we outline[9] a theory for quantum jumps in the fluorescence from two atoms. Cooperative effects turn out to be dramatic if the phase relations between the two atoms are preserved over the time scale of the quantum jumps. However, in current practice this is a prohibitive requirement, as it presupposes that the distance between the atoms is orders of magnitude smaller than the wavelength.

Let us first recall the quantum jumps in a single three-level atom, say, one with the level scheme of Fig. 1. A resonant laser drives the transition $0 \leftrightarrow 2$ at the Rabi frequency Ω, and the spontaneous transitions $2 \rightarrow 0$ take place at the rate γ_a. The transitions to and from the metastable state 1 at the very low rates γ_b and γ_c are exclusively spontaneous. According to Dehmelt,[10] the atom scatters a continuous stream of photons while it makes rapid transitions between the states 0 and 2, whereas the fluorescence suddenly ceases when the atom jumps to the "shelving" state 1 and equally suddenly reappears upon a new quantum jump from 1 to 0. Cook and Kimble[11] interpreted the rate equations of the atom as giving also the probabilities per unit time for the jumps between the light and dark states of the fluorescence. Subsequent photon-statistics analyses[12] have deduced the quantum jumps in the fluorescence from *ab initio* arguments instead of assuming them at the outset, and confirmed the results of the rate equations. Experiments[1] have finally materialized in substantial agreement with theory.

Here we again resort to a rate-equation treatment, confident that a more rigorous analysis can be given as in Ref. 12 if desired.

Now take two atoms like that in Fig. 1, labeled "left" and "right." Because we focus on the limit of very high Rabi frequency, we employ for both atoms instead of the bare states $|0\rangle$ and $|2\rangle$ their laser-dressed[13] counterparts $|\pm\rangle^{l,r} = 2^{-1/2}(|0\rangle^{l,r} \pm |2\rangle^{l,r})$. The state space of the combined two-atom system is spanned by the dyadic products $|i\rangle^l |j\rangle^r$, with $i,j = 1$, $+$, or $-$. However, with another change of basis we go over to the atom-symmetrized and -antisymmetrized linear combinations[3]

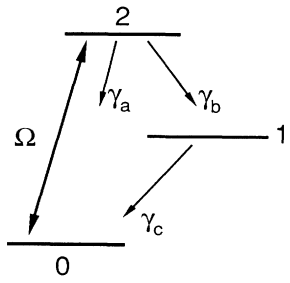

FIG. 1. Notation of one of the three-level atoms. A resonant external field drives the transition $|0\rangle \leftrightarrow |2\rangle$ at the large Rabi frequency Ω. The spontaneous decay rates are $|2\rangle \rightarrow |0\rangle$, γ_a; $|2\rangle \rightarrow |1\rangle$, γ_b; $|1\rangle \rightarrow |0\rangle$, γ_c.

instead, defined respectively as

$$|ij\rangle^s = 2^{-1/2}(|i\rangle^l |j\rangle^r + |j\rangle^l |i\rangle^r)$$

and

$$|ij\rangle^a = 2^{-1/2}(|i\rangle^l |j\rangle^r - |j\rangle^l |i\rangle^r).$$

$|ij\rangle^{s,a}$ are stationary (quasi)energy eigenstates of the system consisting of the atoms and the laser field; $|++\rangle^s$ and $|--\rangle^s$ have the energies $\pm\hbar\Omega$, $|+1\rangle^s$ and $|+1\rangle^a$ can be assigned the energy $\frac{1}{2}\hbar\Omega$, and so forth. In this representation, spontaneous emission is the sole cause of transitions. For instance, the piece in the master equation of the atoms which governs the radiative transitions $2 \rightarrow 1$ reads[9,14]

$$\dot{\rho} = \cdots + \tfrac{1}{8}[\gamma_b + \gamma_b(R)][(\sigma^s_{1+} - \sigma^s_{1-})\rho(\sigma^s_{+1} - \sigma^s_{-1}) - (\sigma^s_{+1} - \sigma^s_{-1})(\sigma^s_{1+} - \sigma^s_{1-})\rho + \text{H.c.}]$$

$$+ \tfrac{1}{8}[\gamma_b - \gamma_b(R)][(\sigma^a_{1+} - \sigma^a_{1-})\rho(\sigma^a_{+1} - \sigma^a_{-1}) - (\sigma^a_{+1} - \sigma^a_{-1})(\sigma^a_{1+} - \sigma^a_{1-})\rho + \text{H.c.}] + \cdots . \quad (1)$$

Here $\sigma^{s,a}_{ij} = |i\rangle^{l\,l}\langle j| \pm |i\rangle^{r\,r}\langle j|$ are the symmetrized and antisymmetrized transition operators between $|i\rangle$ and $|j\rangle$, and $\gamma_b(R)$ entails the modifications of spontaneous emission because of cooperative effects of the atoms at the distance R. For our discussion it suffices to recognize that $\gamma_b(R) \rightarrow \gamma_b$ quadratically as $R \rightarrow 0$, that $\gamma_b(R) \rightarrow 0$ as $R \rightarrow \infty$, and that the wavelength of the transition provides the scale of variation of $\gamma_b(R)$ with R.[4,14] Similar collective emission rates can be defined for the other two spontaneous emission transitions called a and c.

The density operator ρ in principle has 81 independent real components. Fortunately, in the particular atomic basis we have chosen, all off-diagonal elements can be shown to tend to zero in the limit $\Omega \rightarrow \infty$. For high intensities of the driving field, the nine occupation probabilities of the states and a nine-component rate equation suffice to describe the physics of the two atoms. Moreover, Eq. (1) demonstrates that the six rates $\gamma_a \pm \gamma_a(R)$, $\gamma_b \pm \gamma_b(R)$, and $\gamma_c \pm \gamma_c(R)$ determine the evolution time scales.

Below we assume that the time scale corresponding to $\gamma_a + \gamma_a(R)$, essentially the photon emission rate in the strong transitions $2 \rightarrow 0$, is faster than the resolution of the experiment. The same may or may not apply to $\gamma_a - \gamma_a(R)$, but the rest of the time scales are always taken to be much slower than $\gamma_a + \gamma_a(R)$. The fast decay rates divide the nine states into groups such that *within* each group an equilibrium is reached in a time too short to be resolved, whereas the transitions *between* the groups are slow. We thus adopt the following approach to quantum jumps in the scattered light. (i) At every instant of time the atom system is assumed to be in one of the state blocks, sending out a light intensity as appropriate for the adiabatic equilibrium within that group. (ii) The atoms jump between the groups, and hence between states of different fluorescent intensity, with probabilities per unit time equal to the calculated transition rates between the groups.

As the first illustration we consider the case when $\gamma_a + \gamma_a(R)$ and $\gamma_a - \gamma_a(R)$ are both regarded as fast decay rates. There are three distinct groups of atomic states: $\{|++\rangle^s, |--\rangle^s, |+-\rangle^{s,a}\}$, $\{|1+\rangle^{s,a}, |1-\rangle^{s,a}\}$, and $\{|11\rangle^s\}$. In the fast equilibrium the populations of the states within each group are equal. The total fluorescence intensities from the groups are, respectively, 2, 1, and 0 times the fluorescence intensity of one atom in the "on" state of its quantum jump. The equations for the probabilities of the fluorescence levels read

$$\dot{P}_2 = -\gamma_b P_2 + \gamma_c P_1, \quad (2a)$$

$$\dot{P}_1 = -(\tfrac{1}{2}\gamma_b + \gamma_c)P_1 + \gamma_b P_2 + 2\gamma_c P_0, \quad (2b)$$

$$\dot{P}_0 = -2\gamma_c P_0 + \tfrac{1}{2}\gamma_b P_1. \quad (2c)$$

Equations (2) are valid in particular when the atoms are infinitely far apart, $R \rightarrow \infty$, and $\gamma_a(R) = 0$. Then both atoms jump independently between their "on" and "off" states at the rates γ_c and $\frac{1}{2}\gamma_b$; the fluorescence levels 2, 1, and 0 correspond to the possible number of the "on" atoms; and equal populations within each group result as the one-atom states $|\pm\rangle^{l,r}$ have the same population and the exchange symmetry must be inconsequential. The collective effects do not show at all; especially the dependence on $\gamma_a(R)$, $\gamma_b(R)$, and $\gamma_c(R)$ is absent. We emphasize that this is the outcome of a quantum-jump experiment *whenever* $\gamma_a - \gamma_a(R)$ is a rate too fast to be resolved, even if the atoms are close to one another. Other cooperative effects[3-6] may or may not be observed, fairly independently of the quantum jumps.

We next take up the more general case when $\gamma_a - \gamma_a(R)$ is also treated as a slow rate. There are five different state groups,

$$\{|++\rangle^s, |--\rangle^s, |+-\rangle^s\}, \ \{|+1\rangle^s, |-1\rangle^s\},$$

$$\{|+1\rangle^a, |-1\rangle^a\}, \ \{|+-\rangle^a\}, \ \{|11\rangle^s\}.$$

The adiabatic equilibrium populations within each group are again equal, and the respective group intensities are $\frac{8}{3}$:1:1:0:0. Although some of the fluorescence states share the same intensity, they have to be considered separately since the transition rates to and from them are different.

We abbreviate $\gamma_a \pm \gamma_a(R)$ by $\gamma_a^{\pm}$, etc., and use an obvious notation for the fluorescence states. All nonzero transition rates are specified by

$$0^s \to 1^s,\ \gamma_c^+;\quad 0^s \to 1^a,\ \gamma_c^-; \tag{3a}$$

$$0^a \to 1^s,\ \tfrac{1}{2}\gamma_b^-;\quad 0^a \to 1^a,\ \tfrac{1}{2}\gamma_b^+;\quad 0^a \to (\tfrac{8}{3})^s,\ \gamma_a^-; \tag{3b}$$

$$1^s \to 0^s,\ \tfrac{1}{2}\gamma_b^+;\quad 1^s \to 0^a,\ \tfrac{1}{4}\gamma_c^-;\quad 1^s \to 1^a,\ \tfrac{1}{4}\gamma_a^-;\quad 1^s \to (\tfrac{8}{3})^s,\ \tfrac{3}{4}\gamma_c^+; \tag{3c}$$

$$1^a \to 0^s,\ \tfrac{1}{2}\gamma_b^-;\quad 1^a \to 0^a,\ \tfrac{1}{4}\gamma_c^+;\quad 1^a \to 1^s,\ \tfrac{1}{4}\gamma_a^-;\quad 1^a \to (\tfrac{8}{3})^s,\ \tfrac{3}{4}\gamma_c^-; \tag{3d}$$

$$(\tfrac{8}{3})^s \to 0^a,\ \tfrac{1}{3}\gamma_a^-;\quad (\tfrac{8}{3})^s \to 1^s,\ \tfrac{1}{2}\gamma_b^+;\quad (\tfrac{8}{3})^s \to 1^a,\ \tfrac{1}{2}\gamma_b^-. \tag{3e}$$

Difference rates with minus sign as the superscript govern the crossing between symmetric and antisymmetric two-atom configurations. Direct transitions between

$$(\tfrac{8}{3})^s = \{|++\rangle^s, |--\rangle^s, |+-\rangle^s\}$$

and $0^a = \{|+-\rangle^a\}$, the superradiant and nonradiating states already discussed by Dicke,[3] are the qualitatively new feature of this case. In the experiments they would emerge as double quantum jumps where both atoms turn on and off simultaneously.

Equations (2) can be derived from the rates (3) by our letting $\gamma_a - \gamma_a(R) \to \infty$, whence the symmetric and antisymmetric states are mixed faster than can be resolved. The fluorescence states we have denoted by $(\frac{8}{3})^s$ and 0^a then shrink into one, as do the states 1^s and 1^a. Conversely, this limit also hints at an explanation of the surprising intensity of the state $(\frac{8}{3})^s$. For two independent atoms the intensity of the "both atoms on" state is 2 and the states $|++\rangle^s$, $|--\rangle^s$, $|+-\rangle^s$, and $|+-\rangle^a$ are equally populated. Now, the same must apply to the averages over the time scale set by $\gamma_a - \gamma_a(R)$, even if the states $(\frac{8}{3})^s$ and 0^a are resolved. The atom spends one-fourth of the time in the nonradiating state $|+-\rangle^a$, and to produce the average fluorescence 2 it makes up for the dark time by emitting at $\frac{8}{3}$ while it is in the active atomic state triplet $(\frac{8}{3})^s$.

In the limit when the atoms are very close to one another, ideally $\gamma_a - \gamma_a(R) = 0$, crossing between symmetric and antisymmetric configurations does not occur. Instead, the atoms either dwell in the fluorescence state triplet $\{(\frac{8}{3})^s, 1^s, 0^s\}$ displaying quantum jumps at the intensities $\frac{8}{3}$:1:0 (with no double jumps), or execute two-state quantum jumps in $\{1^a, 0^a\}$. A very small nonzero value of $\gamma_a - \gamma_a(R)$ manifests itself in random jumps between these two quantum-jump schemes.

Unfortunately, our nontrivial predictions may be hard to verify experimentally. In the ion-trap experiments the collection efficiency of light is typically $\eta \simeq 10^{-4}$. Several photons must be detected in order to identify a state with nonzero fluorescence, and so its lifetime must be much larger than η^{-1} times the interval between the emitted photons; i.e., $\gamma_a - \gamma_a(R) \ll \eta[\gamma_a + \gamma_a(R)]$. This requires that the distance between the atoms be at most of the order of $\eta^{1/2} \simeq 10^{-2}$ wavelengths.

Our discussion applies directly only to our specific model with all its assumptions, such as the very high light intensity. Nonetheless, we conjecture more generally that, to observe collective quantum jumps, the crossing of the two-atom system between antisymmetric and symmetric configurations must be slower than the experimental time resolution for quantum jumps. More work is needed to clarify the nature of the double and triple jumps reported in Ref. 7. In fact, ion traps may be inherently unsuitable for studies of collective quantum jumps because the Coulomb forces push the ions apart. Neutral-atom traps[15] do not suffer from the Coulomb repulsion; so in the long run they may be better suited for this purpose.

In summary, cooperative interactions of two nearby atoms with light may drastically shape the quantum jumps. Our predictions include intensity ratios different from the 2:1:0 scheme of two independent atoms, double jumps where both atoms appear to turn on or off simultaneously, and long-term alternations between different quantum-jump patterns. It appears, though, that it is exceedingly difficult to see collective quantum jumps experimentally.

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