Possibility of quantum jumps

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A quantum-statistical theory is developed for a single-atom, three-state quantum system where a strong (1-3) and a weak (2-3) transition are driven resonantly by an incoherent field. From a second-order correlation function of the fluorescence intensity, it is concluded that a quantum jump can occur, in the sense that the single atom can undergo large fluctuations between a state of full emission in the strong transition to a state of no emission when the electron is shelved in the metastable state (2).

Quantum mechanics is an inherently statistical theory that describes the behavior of microscopic systems in a probabilistic way. For example, one cannot predict the result of a single event or a single measurement such as the instant in time that a single excited atom decays to a lower state or when a detector will observe the emitted photon. However, it is possible to predict the average time an atom spends in its excited state or the average time it takes to observe the photon after excitation. In the case of a single atom, these averages are realized experimentally in a series of repeated measurements, where each time an atom is prepared in an identical way. For a large ensemble of independent atoms, a single measurement also yields an average of the individual atomic emission signals which follows the well-known exponential decay law.

Now that a single atom or ion can be observed spectroscopically, either in a faint atomic beam or in an electromagnetic trap,[1,2] a fundamental test of an atom's quantum statistical behavior is at hand. An important example is the observation of antibunching[3] in resonance fluorescence—an effect that is based on the properties[4] of a single atom and that is erased by the presence of other excited atoms.

Another one-atom effect, "shelving," has been proposed by Dehmelt,[5] and is the subject of this and the preceding[6] article. It involves, for example, the three-level quantum system of Fig. 1(a), where a strong ($1 \leftrightarrow 3$) and a weak ($2 \leftrightarrow 3$) transition are coupled, each being driven by a resonant field. Many photons will be scattered in resonance fluorescence by the strong transition, but occasionally this scattering will be suppressed, or perhaps even extinguished, when the weak transition shelves the atom in the metastable state (2). Thus, the strong transition registers the presence of the weak transition, and because the weak transition linewidth may be exceptionally narrow, this scheme has been proposed for an ultimate laser frequency standard.[5]

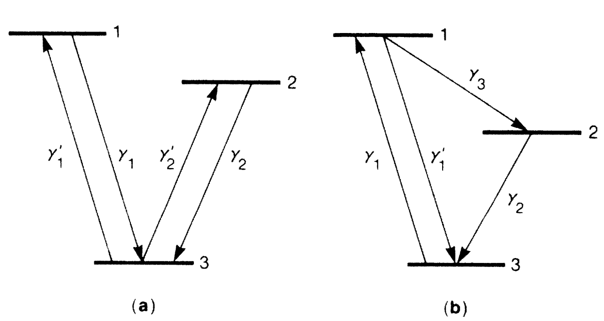

FIG. 1. Energy-level diagrams for the case where (a) the transitions 1-3 and 2-3 are driven by two resonant fields and (b) only the 1-3 transition is excited.

Consider now the case of a large ensemble of atoms where one subgroup undergoes spontaneous emission in the strongly driven $1 \leftrightarrow 3$ transition, the events occurring randomly in time, while a second subgroup does not contribute to the $1 \leftrightarrow 3$ scattering because they are shelved. An individual atom may fluoresce randomly in time and be shelved for other periods, but the time average of the number in each subgroup is a constant. Therefore, for an ensemble, the fluorescence intensity of the strong transition would merely be slightly reduced due to the shelving effect.[6]

We now ask the following questions: How is the reduction of the fluorescence intensity realized for the case of a single atom? Does the single atom emit light continuously, but at a reduced rate, or will the fluorescence intensity undergo large fluctuations between a state of full emission, while the $1 \leftrightarrow 3$ transition is driven, to a state of no emission when the electron is shelved? If the second possibility is correct, then the single atom would display an individual quantum jump, not by the emission of a single photon, but by the turning on and off of the strong fluorescence of the $1 \leftrightarrow 3$ transition. While a forbidden transition may have a feeble rate of emission (say 1 photon/sec), which would be difficult to detect, the presence or absence of fluorescence from the strong transition could be of macroscopic order, say 10^8 photons/sec, and easily detected.

The potential of the shelving principle to indicate individual quantum events through a macroscopic signal has been discussed recently by Cook and Kimble.[7] Indeed, their discussion adopts the intuitive view that quantum jumps occur, whereas *in this paper we predict their existence.* The answer to whether quantum jumps occur or not can only come from a detailed quantum statistical calculation. We acknowledge, however, that for a single atom such a theory will never describe any single event including a discontinuous quantum jump, or an individual time trajectory of the fluorescence intensity. On the other hand, a calculation of the average fluorescence intensity would clearly be insufficient in deciding whether the intensity is continuous in time or widely fluctuating between periods of emission and darkness. Detailed statistical information about the dynamics of the emission process is required, and that follows from the hierarchy of correlation functions of the emitted field. As a cautionary note, we mention that the more intuitive classical statistics can be misleading, since it can differ substantially from a quantum statistical approach.

We describe the dynamics of the atom, corresponding to either Figs. 1(a) or 1(b), by a set of fermion operators,

$$a_i^\dagger \text{ and } a_i, \quad i=1,2,3$$

for the levels 1, 2, and 3. The amplitude of the two fields emitted, $E_i^{\pm}$ $(i=1,2)$, is proportional to the polarization[8] of the medium

$$E_i^- = \sqrt{\Gamma_i} a_i^\dagger a_3, \quad i=1,2 \ ,$$
$$E_i^+ = \sqrt{\Gamma_i} a_3^\dagger a_i \ , \tag{1}$$

where all unimportant factors have been absorbed in the renormalization and $\Gamma_{1,2}$ are the spontaneous decay rates. The retardation, due to the propagation from the source to the detector, is not displayed explicitly either. We assume that detectors can spectrally resolve either $E_1^{\pm}$ or $E_2^{\pm}$ and that quantum beats,[9] characteristic of spontaneous emission from multilevel systems, need not be considered.

Some insight into the characteristics of the emitted field is provided by the first-order correlation function[10]

$$G_i(t) = \langle E_i^-(t) E_i^+(t) \rangle = \Gamma_i \rho_{ii}(t) \ , \tag{2}$$

which is proportional to the average fluorescence intensity since $\rho_{ii}(t)$ is the density matrix of the three-level system. For long times, the stationary value

$$\lim_{t\to\infty} G_i(t) = G_i^{SS} = \Gamma_i \rho_{ii}^{SS}, \quad i=1,2 \tag{3}$$

is approached. The existence of the metastable state $|2\rangle$ merely reduces the average fluorescence intensity[6] and hence G_1^{SS}, but there is no indication whether the reduction occurs in a continuous or discontinuous fashion. Insight into the dynamics and statistical nature of the three-level atomic emission is revealed in the stationary second-order correlation function[10]

$$G_{ij}^{SS}(t) = \langle E_j^-(0) E_i^-(t) E_i^+(t) E_j^+(0) \rangle, \quad t>0 \ . \tag{4}$$

With the insertion of (1) into (4), we obtain

$$G_{ij}^{SS}(t) = \Gamma_i \Gamma_j \langle a_j^\dagger(0) a_3(0) a_i^\dagger(t) a_i(t) a_3^\dagger(0) a_j(0) \rangle \ , \tag{5}$$

where we have made use of the fermion commutation relation $a_{33}^\dagger a_{33} + a_{33} a_{33}^\dagger = 1$. Under the assumption that spontaneous emission establishes a Markov process, the above correlation function can be evaluated using the quantum regression theorem.[11] For this purpose, we use the time evolution operator

$$a_j^\dagger(t) a_i(t) = \sum_{l,n} K_{ln}^{ij}(t) a_n^\dagger(0) a_l(0) + \text{noise} \ . \tag{6}$$

It obeys the same equations of motion as the density matrix

$$\rho_{ij}(t) = \sum_{l,n} K_{ln}^{ij}(t) \rho_{ln}(0) \ , \tag{7}$$

except for the noise terms. Inserting this result into the photon correlation function (5), we find

$$G_{ij}^{SS}(t) = \Gamma_i \Gamma_j K_{33}^{ii}(t) \rho_{jj}^{SS} \ . \tag{8}$$

From Eq. (7), the following properties are evident:

$$K_{ln}^{ij}(t=0) = \delta_{il}\delta_{jn} \text{ and } K_{33}^{ij}(t=\infty) = \rho_{ij}^{SS} \ , \tag{9}$$

where $\rho_{33}(0)=1$. Thus, the corresponding correlation functions are

$$G_{ij}(t=0) = 0 \ , \tag{10a}$$

$$G_{ij}(t=\infty) = \Gamma_i \Gamma_j \rho_{ii}^{SS} \rho_{jj}^{SS} \ , \tag{10b}$$

where (10a) reflects the well-known property of antibunching[3,4] in resonance fluorescence. For intermediate times, the two-photon correlation function $G_{ij}(t)$ can be interpreted as the probability of observing a photon at time $t=0$, corresponding to the $j \to 3$ transition, and then subsequently at time t, a second photon for the $i \to 3$ transition. Since the time origin of G_{ij} has been chosen arbitrarily, the probability of observing the first photon at $t=0$ is just $G_j^{SS} \equiv \Gamma_j \rho_{jj}^{SS}$. Therefore, the conditional probability of observing a second photon at time t in a time window Γ_1^{-1} is proportional to

$$P_{ij}(t) = G_{ij}^{SS}(t) / (\Gamma_i \Gamma_j \rho_{jj}^{SS}) \ . \tag{11}$$

The only quantity remaining to be evaluated is the coefficient $K_{ln}^{ij}(t)$, which follows from the solution of the three-level density matrix. The level structure of Figs. 1(a) and 1(b) requires the solution of either eight or four coupled linear differential equations, and these are amenable only to numerical integration. To obtain analytic results and thus insight into the properties of spontaneous emission from a three-level system, we simplify the dynamics by assuming incoherent driving fields as suggested in Fig. 1. The resulting rate equations are solved easily, but since the solutions are still involved, an additional simplification is introduced in Fig. 1(a) by driving both transitions strongly, but not at an infinite rate, so that $\gamma_1' \cong \gamma_1$ and $\gamma_2 \cong \gamma_2'$ and in Fig. 1(b) by saturating the $1 \leftrightarrow 3$ transition so that $\gamma_1' \cong \gamma_1$ and assuming that the relaxation rates in and out of the metastable state are equal $(\gamma_2 = \gamma_3)$. These restrictions are by no means essential for obtaining a solution, but they reduce the complexity of the result and the number of independent parameters. Here, $\gamma_1' = 4\alpha^2/\Gamma_1$, $\gamma_1 = \Gamma_1 + \gamma_1'$, $\gamma_2' = 4\beta^2/\Gamma_2$, and $\gamma_2 = \Gamma_2 + \gamma_2'$. Observing the inequality

$$\gamma_1 >> \gamma_2 \ ,$$

which is physically relevant for this problem, we obtain for either Fig. 1(a) or 1(b) the simple expressions

$$K_{33}^{11}(t) = \tfrac{1}{3}[1 + \tfrac{1}{2}(e^{-(3/2)\gamma_2 t} - 3e^{-2\gamma_1 t})] + O\left(\frac{\gamma_2}{\gamma_1}\right) \ , \tag{12a}$$

$$K_{33}^{22}(t) = \tfrac{1}{3}(1 - e^{-(3/2)\gamma_2 t}) + O\left(\frac{\gamma_2}{\gamma_1}\right) \ . \tag{12b}$$

The conditional probability of observing a photon E_1 at time t, after having seen a photon E_1 at $t=0$, follows from (8), (11), and (12a) as

$$P_{11} = \tfrac{1}{3}[1 + \tfrac{1}{2}(e^{-(3/2)\gamma_2 t} - 3e^{-2\gamma_1 t})] \ , \tag{13}$$

and is plotted in Fig. 2 as curve 2. Also, the average fluorescence intensity of the strong transition is $G_1^{SS} = \frac{1}{3}\Gamma_1$, since $\rho_{11} = \frac{1}{3}$ in a fully saturated three-level system.

To rationalize the features of Fig. 2, we first note that for short times, $t << \gamma_2^{-1}$, Eq. (13) is essentially indistinguishable from a saturated two-level system (curve 1), which with the same parameters obeys the relationships

$$P_{11}^{(a)} = \tfrac{1}{2}(1 - e^{-2\gamma_1 t}), \quad G_1^{SS} = \tfrac{1}{2}\Gamma_1 \ . \tag{14}$$

Over this period, the three-level system behaves as if the metastable state (2) did not exist, and the conditional pro-

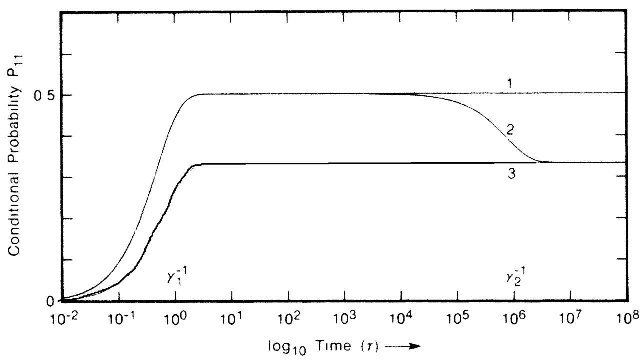

FIG. 2. The conditional probability $P_{11}(\tau)=K_{33}^{11}(\tau)$, Eq. (12a), of a three-level system for a photon E_1, being emitted at time τ, following the emission of a photon E_1 at an earlier time $\tau=0$ (curve 2). Similar conditional probabilities for a fully (curve 1) and a partially (curve 3) saturated two-level system, where the former overlaps the three-level case for short times $\tau \leq \gamma_1^{-1}$ and the latter for long times $\tau \geq \gamma_2^{-1}$. The condition $\gamma_1/\gamma_2=10^6$ applies.

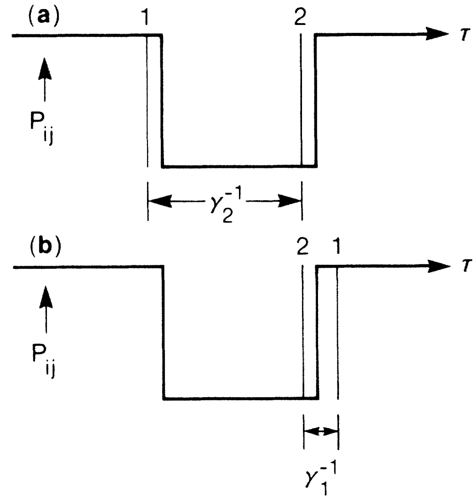

FIG. 3. Cross-correlation conditional probabilities are shown for P_{12} and P_{21}, Eq. (16), where the asymmetry in the delay times of photons 1 and 2 is evident.

bability for the two- and three-level systems is the same. For the shortest times, $t \ll \gamma_1^{-1}$, the conditional probability drops to zero, indicating that there is no second photon available immediately after absorption of the first photon. This is the antibunching phenomenon.[3,4]

For times $t \sim \gamma_2^{-1}$ and longer, the three-level conditional probability coincides with that of a partially saturated two-level system (curve 3), which obeys

$$P_{11}^{(b)}=\tfrac{1}{3}(1-e^{-(3/2)\gamma_1 t}),\quad G_1^{SS}=\tfrac{1}{3}\Gamma_1 \ . \tag{15}$$

We see that while the two-level solutions (a) and (b) represent continuous fluorescence signals with average intensities $\frac{1}{2}\Gamma_1$ and $\frac{1}{3}\Gamma_1$, respectively, the three-level result (13) bridges the two cases. Furthermore, at intermediate times $\gamma_1^{-1} < t < \gamma_2^{-1}$, the instantaneous intensity lies significantly above the limiting average value at long times $t \gg \gamma_2^{-1}$. Thus, we conclude that there must exist significant periods of darkness or extremely weak emission so that the time-average intensity reduces to the correct asymptotic value $\frac{1}{3}\Gamma_1$.

Additional confirmation of these ideas appears in the cross-correlation conditional probabilities

$$G_{12}^{SS}(t)/(\Gamma_1\Gamma_2\rho_{22}^{SS})=K_{33}^{11}(t)=P_{12}(t)=P_{11}(t) \ , \tag{16a}$$

$$G_{21}^{SS}(t)/(\Gamma_1\Gamma_2\rho_{11}^{SS})=K_{33}^{22}(t)=P_{21}(t)=P_{22}(t) \ . \tag{16b}$$

These relations correspond to the emission of photon 2 ($2\rightarrow 3$ transition), followed by photon 1 ($1\rightarrow 3$ transition) or in the inverse order. Since K_{33}^{11} and K_{33}^{22} given by (12) are different, the conditional probability depends on the order of emission. This feature is summarized in Fig. 3, and shows for P_{21} that, on the average, when photon 1 is emitted, a long time γ_2^{-1} will elapse before photon 2 is emitted. Looking backward in time, the emission of photon 2 signifies that a dark interval γ_2^{-1} preceded it, since on the average a photon 1 was not emitted. Conversely, P_{12} implies that after photon 2 is emitted, only a brief period γ_1^{-1} is required on the average before photon 1 appears. The slow 2-3 transition allows the fast 3-1 transition to follow almost immediately. Then, according to P_{11}, a sequence of rapid bursts of photon 1 light follows, to be interrupted again by a period of darkness. A period of photon 1 light lasting $2\gamma_2^{-1}$, with an intensity of $\frac{1}{2}\gamma_1$ and a period of darkness lasting γ_2^{-1}, would then yield the correct time-averaged value $G_1^{SS}=\frac{1}{3}\Gamma_2$. It should be noted that the time asymmetry in P_{12} and P_{21} is a pure quantum effect, whereas in classical statistics the behavior is independent of the order of emission.

One might question whether the higher-order correlation functions, which have been neglected, are required. Since we are dealing with a Markov process, an assumption that is vital to the application of the regression theorem, nothing new is learned by considering higher-order correlation functions. This is because the higher-order function decomposes into a product of second-order correlations.

Thus, it appears that the intuitive picture of jumps in a three-level quantum system, as initially envisioned by Dehmelt, agrees with the quantum statistical theory developed here. Clearly, an experimental test would now be timely, but the assumption of an incoherent driving field should be replaced by a coherent source, and this case will be treated in a forthcoming article.

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