Possibility of Direct Observation of Quantum Jumps

Richard J. Cook

Department of Physics, Air Force Institute of Technology, Wright-Patterson Air Force Base, Dayton, Ohio 45433

and

H. J. Kimble

Department of Physics, University of Texas at Austin, Austin, Texas 78712

In a single-atom double-resonance experiment involving a strong and a weak transition, quantum jumps on the weak transition cause the fluorescence of the strong transition to turn on and off abruptly. The fluorescence is off when the weak transition is excited and on when it is not. Thus quantum jumps on the weak transition can be directly monitored by observation of the random telegraph signal radiated by the strong transition. We present here a simple theory of this effect for the case of incoherent excitation.

For various reasons, spectroscopists have usually worked with samples containing a large number of atoms. Only very recently has it become possible to confine a single atomic ion in a radio-frequency trap and to subject the ion to spectroscopic study.[1,2] This is important because a number of effects that can be observed in the fluorescence of a single atom (or ion) are totally masked when many atoms contribute to the fluorescence. The effect considered here is of this type.

The idea we shall develop was first suggested by Dehmelt as a way to detect a weak transition in single-atom spectroscopy.[3] Although single-atom fluorescence from a strong optical transition ($\sim 10^8$ photons/sec) is readily detected either visually (using a microscope) or photoelectrically, the direct detection of a fluorescent or absorptive line profile of a very weak transition (say 1 photon/sec) is problematic. To circumvent this difficulty, Dehmelt proposed the double-resonance scheme illustrated in Fig. 1, in which the weak transition of interest, $0 \leftrightarrow 2$, and a strong transition, $0 \leftrightarrow 1$, have a common lower level. Suppose the atomic electron starts in state 0. With the strong transition saturated and no excitation of the

weak transition, the electron undergoes transitions rapidly between states 0 and 1, and the rate of fluorescence (photons/sec) on this transition is $R_1 = A_1 P_1 = A_1/2$, since the probability to be in state 1 is $\frac{1}{2}$ (here A_1 is the Einstein spontaneous emission coefficient for the strong transition). Under these conditions, a detector at distance r from the atom registers a mean fluorescent intensity (irradiance) I_0 which is proportional to the rate $A_1/2$ of photon emission and inversely proportional to r^2. Of course, the true fluorescent signal consists of a sequence of pulses as photons arrive at the detector. But, for the very high photon emission rate of the strong transition, we may ignore the discreteness and treat the intensity I_0 as constant in time. Now let radiation be applied to the weak transition and let the saturating field continue to act on the strong one. Then the detector will continue to register the intensity I_0 until the electron makes a transition to level 2. This turns off the strong fluorescence because the electron is no longer available for transitions between levels 0 and 1. The strong fluorescence turns on again when the electron returns to level 0 either by spontaneous or stimulated emission. Since the weak transitions occur randomly in time, the atomic fluorescence $I(t)$ has the form of a random telegraph signal, as illustrated in Fig. 2. The important point is that the
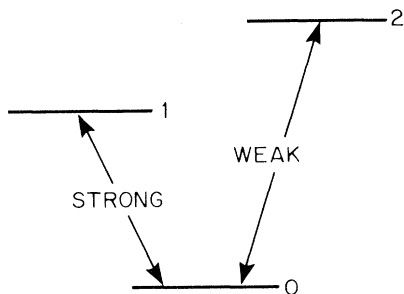

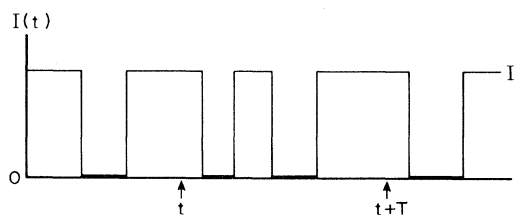FIG. 1. Energy-level scheme for single-atom double-resonance experiment.

FIG. 2. Single-atom fluorescent intensity vs time. Interruptions of fluorescence are due to excitation of the weak transition $0 \rightarrow 2$.

fluorescence emanating from the strong transition is a direct indicator of the state of excitation of the weak transition; the fluorescence is off when level 2 is occupied and on when it is not. So by observation of the atomic fluorescence $I(t)$, say with the eye and a microscope, one can directly monitor the quantum jumps on the weak transition. At the time of writing, this effect has not been observed, but probably will be in the near future.

The purpose of this Letter is to present a first theoretical treatment of the above single-atom double-resonance effect. For simplicity we limit the analysis to the case of incoherent excitation. To begin, we look at the rate equations for the probabilities P_0, P_1, and P_2 that the levels in Fig. 1 are occupied. Let A_1, B_1 and A_2, B_2 be the Einstein coefficients for the strong and weak transitions, respectively ($A_1 \gg A_2$). Then, if U_1 and U_2 are the spectral energy densities of radiation acting on the two transitions, the Einstein rate equations for P_1 and P_2 read

$$\dot{P}_1 = -A_1 P_1 + B_1 U_1 (P_0 - P_1), \tag{1a}$$

$$\dot{P}_2 = -A_2 P_2 + B_2 U_2 (P_0 - P_2). \tag{1b}$$

A rate equation for P_0 would be superfluous, since

$$P_0 + P_1 + P_2 = 0. \tag{2}$$

The *mean* rates of fluorescence (photons/sec) on the two transitions are $R_1 = A_1 P_1$ and $R_2 = A_2 P_2$.

We assume that the strong transition is strongly saturated ($U_1 \to \infty$). In this case $P_1 = P_0$, and it is convenient to work with the variables

$$\mathscr{P}_+ = P_2, \quad \mathscr{P}_- = P_0 + P_1, \tag{3}$$

which are the probabilities that the weak transition is excited or not excited, respectively. From (1) and (2), which now reads

$$\mathscr{P}_+ + \mathscr{P}_- = 1, \tag{4}$$

we obtain

$$\dot{\mathscr{P}}_+ = -R_- \mathscr{P}_+ + R_+ \mathscr{P}_-, \tag{5a}$$

$$\dot{\mathscr{P}}_- = R_- \mathscr{P}_+ - R_+ \mathscr{P}_-, \tag{5b}$$

where $R_+ = \frac{1}{2} B_2 U_2$ and $R_- = A_2 + B_2 U_2$. We may think of the system as an effective two-level system with upward transition rate R_+ and downward rate R_-. In steady state ($\dot{\mathscr{P}}_+ = \dot{\mathscr{P}}_- = 0$), the solution of Eqs. (4) and (5) is

$$\mathscr{P}_+ = \frac{R_+}{(R_+ + R_-)}, \quad \mathscr{P}_- = \frac{R_-}{(R_+ + R_-)}. \tag{6}$$

The rate equations for $\mathscr{P}_+$ and $\mathscr{P}_-$ do not describe the stochastic process of Fig. 2. In fact, for steady state, the probabilities (6) determine only the mean value of the random telegraph signal ($\bar{I} = I_0 \mathscr{P}_-$) and

say nothing about the fluctuations of this signal. A more complete description of the stochastic process is provided by the following probabilities.[4] Let $P_{n,+}(t,T)$ denote the probability that n transitions occur between levels $+$ and $-$ in the time interval $[t, t+T]$ and that the atom is left in level $+$ at time $t+T$. Similarly, let $P_{n,-}(t,T)$ be the probability that n transitions occur on $[t, t+T]$ with the atom in level $-$ at time $t+T$. Then $P_n(t,T) = P_{n,+}(t,T) + P_{n,-}(t,T)$ is clearly the probability that n transitions occur on $[t, t+T]$, regardless of the final state; and

$$\mathscr{P}_+(t+T) = \sum_{n=0}^{\infty} P_{n,+}(t,T), \tag{7a}$$

$$\mathscr{P}_-(t+T) = \sum_{n=0}^{\infty} P_{n,-}(t,T), \tag{7b}$$

are the probabilities for final states $+$ and $-$, regardless of the number of transitions that have occurred. If the stochastic process $I(t)$ is stationary, then $P_{n,\pm}(T)$ are functions of T only and $\mathscr{P}_\pm$ are constant.

In Fig. 3 the probabilities $P_{n,\pm}$ are represented by solid dots. Since a transition to level $+$ ($-$) always originates in level $-$ ($+$), and since transitions occur one at a time ($n \to n+1$), the flow of probability between the dots as T increases is that indicated by the arrows in the figure. Finally, because the rates of upward and downward transitions are R_+ and R_-, respectively, regardless of the number of transitions that have occurred, we must associate the rate R_+ with each of the up arrows and the rate R_- with each of the down arrows. It follows then from Fig. 3 that the rate equations for the probabilities $P_{n,\pm}$ are

$$dP_{n,+}/dT = R_+ P_{n-1,-} - R_- P_{n,+}, \tag{8a}$$

$$dP_{n,-}/dT = R_- P_{n-1,+} - R_+ P_{n,-}. \tag{8b}$$

It is also clear from the figure that, for these equations to be valid for $n = 0$, we must set $P_{-1,\pm} = 0$, since no probabilities feed $P_{0,\pm}$, i.e., the first term on the right is absent in each of Eqs. (8) for $n = 0$. As a check on Eqs. (8) one may sum each of them over all n and use (7). The result is the Einstein rate equations (5).

To solve Eqs. (8) we need initial conditions at $T = 0$. These are easily found as follows. In the first place,
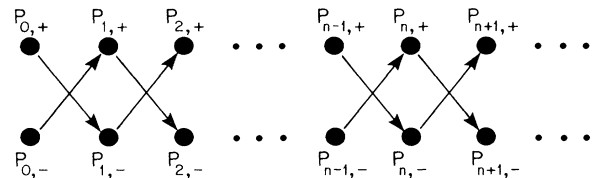

FIG. 3. Solid dots represent probabilities $P_{n,\pm}(T)$ that n transitions occur in time T and that atom is left in state $\pm$. Arrows indicate the flow of probability as T increases.

since no transitions can occur in a time interval of zero length, we must have $P_{n,\pm}(t,0) = 0$ for $n > 0$. Then from Eqs. (7) we find that $P_{0,\pm}(t,0) = \mathscr{P}_\pm(t)$, which completes the specification of initial conditions.

The rate equations (8) enable us to make a number of predictions which may be tested by observing the atomic fluorescence $I(t)$. Consider the distribution of the lengths of time intervals during which the fluorescence is off. Suppose $I(t)$ turns off at time t. Then we know that the atomic electron has just entered the upper level $[\mathscr{P}_+(t) = 1$ and $\mathscr{P}_-(t) = 0]$, and we have the initial condition $P_{0,+}(t,0) = \mathscr{P}_+(t) = 1$. The solution of Eq. (8a) for $n = 0$ [if we remember that $P_{-1,-}(t,T) = 0$] is

$$P_{0,+}(t,T) = \exp(-R_-T). \tag{9}$$

This is the probability that no transitions have occurred in time T, i.e., the probability that the fluorescence $I(t)$ is still off after time T. Accordingly, $F(T) = 1 - \exp(-R_-T)$ is the probability that the fluorescent intensity has turned on in time T, and the derivative of this distribution, $W_{\text{off}} = dF/dT$, is the probability density for the durations T of interruptions in the fluorescent signal ("off times"):

$$W_{\text{off}}(T) = R_-\exp(-R_-T). \tag{10}$$

An exactly analogous argument shows that the probability density for "on times" is

$$W_{\text{on}}(T) = R_+\exp(-R_+T). \tag{11}$$

Thus both the off times and the on times are distributed exponentially, but with different time constants, namely, R_- and R_+, respectively. Clearly both R_+ and R_- can be obtained from a statistical analysis of the random singal $I(t)$, and hence the A coefficient of the weak transition $(A_2 = R_- - 2R_+)$ can also be obtained.

Next consider the two-time intensity correlation function

$$C(T) = \langle I(t)I(t+T)\rangle, \tag{12}$$

which we write as a function of T on the assumption that $I(t)$ is stationary. A glance at Fig. 2 reveals that the intensity product $I(t)I(t+T)$ can assume only two values; it is I_0^2 when the fluorescence is on at time t and at time $t + T$, and it is zero otherwise. Therefore the mean value of the intensity product, namely, $C(T)$ is I_0^2 times the probability $P_{\text{on,on}}(T)$ that $I(t)$ is on both at time t and $t + T$:

$$C(T) = I_0^2 P_{\text{on,on}}(T). \tag{13}$$

For a particular realization of the fluorescent signal that is on at time $t + T$, the signal will also be on at time t provided that there is an even number of transitions between these two times. Thus the total proba-

bility for the signal to be on at t and $t + T$ is

$$P_{\text{on,on}}(T) = \sum_{n \text{ even}} P_{n,-}(T), \tag{14}$$

since $P_{n,-}(T)$ is the probability for signal on (atom in level $-$) at time $t + T$ with n transitions between t and $t + T$. A similar argument shows that

$$P_{\text{on,off}}(T) = \sum_{n \text{ odd}} P_{n,+}(T) \tag{15}$$

is the total probability that the signal is on at time t and off at $t + T$. We must consider this probability also because $P_{\text{on,on}}$ and $P_{\text{on,off}}$ are coupled through the equations

$$dP_{\text{on,on}}/dT = R_- P_{\text{on,off}} - R_+ P_{\text{on,on}}, \tag{16a}$$

$$dP_{\text{on,off}}/dT = R_+ P_{\text{on,on}} - R_- P_{\text{on,off}}. \tag{16b}$$

Equations (16) are derived by our summing Eq. (8a) over even n, summing Eq. (8b) over odd n, and using (14) and (15). The initial conditions for these equations follows from (14), (15), and the previously derived values of $P_{n,\pm}(0)$. In steady state these are $P_{\text{on,on}}(0) = \mathscr{P}_- = R_-/(R_+ + R_-)$ and $P_{\text{on,off}}(0) = 0$. The solution of Eqs. (16) with these initial conditions is, in part,

$$P_{\text{on,on}}(T) = \frac{R_-^2}{(R_+ + R_-)^2}$$
$$+ \frac{R_+ R_-}{(R_+ + R_-)^2}e^{-(R_+ + R_-)T}, \tag{17}$$

and use of this in (13) gives

$$C(T) = M_I^2 + \sigma_I^2 e^{-(R_+ + R_-)T}, \tag{18}$$

where $m_I = \langle I\rangle = I_0R_-/(R_+ + R_-)$ is the mean intensity and $\sigma_I^2 = \langle I^2\rangle - \langle I\rangle^2 = I_0^2 R_+R_-/(R_+ + R_-)^2$ is the variance of intensity. Thus the two-time intensity correlation function falls exponentially from the mean square value $\langle I^2\rangle$ to the squared mean value $\langle I\rangle^2$ as T goes from zero to infinity. The frequency spectrum of intensity fluctuations,

$$S(\omega) = \int_0^\infty C(T)\cos\omega T\,dT$$
$$= \pi m_I^2\delta(\omega) + \frac{\sigma_I^2(R_+ + R_-)}{\omega^2 + (R_+ + R_-)^2}, \tag{19}$$

is also of interest. Except for a zero-frequency component, it is Lorentzian with half-width $\Delta\omega = R_+ + R_-$.

In summary, we emphasized that, in a single-atom double-resonance experiment with a strong and a weak transition, the atomic fluorescence flashes on and off as the atom undergoes weak transitions. This effect is noteworthy because it offers a means by which quantum jumps in an atomic system can be directly monitored and by which the Einstein A coefficient of a very weak transition can be measured. More specifically,

we derived equations which govern the statistics of the fluorescent random telegraph signal, and used these equations to calculate the distributions of on times and off times and the two-time intensity correlation function. These results apply when the atomic excitation is incoherent, i.e., when the spectral energy density of the exciting radiation is a slowly varying function of frequency across each atomic line. This restriction greatly simplifies the analysis. In the case of coherent monochromatic excitation, one must consider a variety of effects, such as the Autler-Townes splitting and broadening of the weak transition, which complicate the calculations considerably. We plan to present elsewhere a detailed theory for the case of coherent excitation.

[1]W. Neuhauser, M. Hohenstatt, P. Toschek, and H. Dehmelt, Phys. Rev. Lett. **41**, 233 (1978), and Phys. Rev. A **22**, 1137 (1980).

[2]W. Nagourney, G. Janik, and H. Dehmelt, Proc. Natl. Acad. Sci. U.S.A. **80**, 643 (1983).

[3]H. Dehmelt, Bull. Am. Phys. Soc. **20**, 60 (1975), and in *Advances in Laser Spectroscopy*, edited by F. T. Arecchi, F. Strumia, and H. Walther, NATO Advanced Study Institute Series, Vol. 95 (Plenum, New York, 1983).

[4]The mathematical method we shall employ is similar to that used by A. Burshtein {Zh. Eksp Teor. Fiz. **49**, 1362 (1965) [Sov. Phys. JETP **22**, 939 (1966)]} and by P. Zoller and F. Ehlotzky [J. Phys. B **10**, 3023 (1977)] in the analysis of random jump processes. It is a generalization of the method used by S. O. Rice [in *Selected Topics in Noise and Stochastic Processes*, edited by N. Wax (Dover, New York, 1954), pp. 176–181] to study the bivalued random telegraph signal. An extensive list of references to the literature on jump processes is given by B. W. Shore, J. Opt. Soc. Am. B **1**, 176 (1984).