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Single-Atom Laser Spectroscopy. Looking for Dark Periods in Fluorescence Light.

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Abstract. – The random sequence of pulses given by a photodetector recording the fluorescence light emitted by a single atom can exhibit periods of darkness if two transitions, one weak and one strong, are simultaneously driven (Dehmelt's electron shelving scheme). We introduce new statistical functions for characterizing these periods of darkness (average length, repetition rate) and we show how to extract spectroscopic information from this type of signals.

Recent developments in methods of laser spectroscopy make it possible now to observe the fluorescence light emitted by a single atom or a single ion. Several experiments of this type have been performed on very dilute atomic beams [1] or laser cooled trapped ions [2].

The signal given by a broadband photodetector recording the fluorescence light looks like a random sequence of pulses. One interesting property of this sequence of pulses, in the case of a single atomic emitter, is the so-called photon anti-bunching. The probability per unit time $g_2(t, t+\tau) = g_2(\tau)$ (¹), if one has detected one photon at time t, to detect another one at time $t+\tau$, tends to zero when τ tends to zero [1]. The interpretation of this effect is that the detection of one photon projects the atom into the ground state, so that we have to wait that the laser re-excites the atom, before we can detect a second photon [3-5].

Another interesting example of single-atom effect is the phenomenon of «electron shelving» proposed by DEHMELT as a very sensitive double-resonance scheme for detecting very weak transitions on a single trapped ion [6]. Consider for example the 3-level atom of fig. 1*a*), with two transitions starting from the ground state g, one very weak $g \leftrightarrow e_R$, one

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(¹) We only consider in this letter stationary random processes, so that all correlation functions such as g_2 only depend on τ.

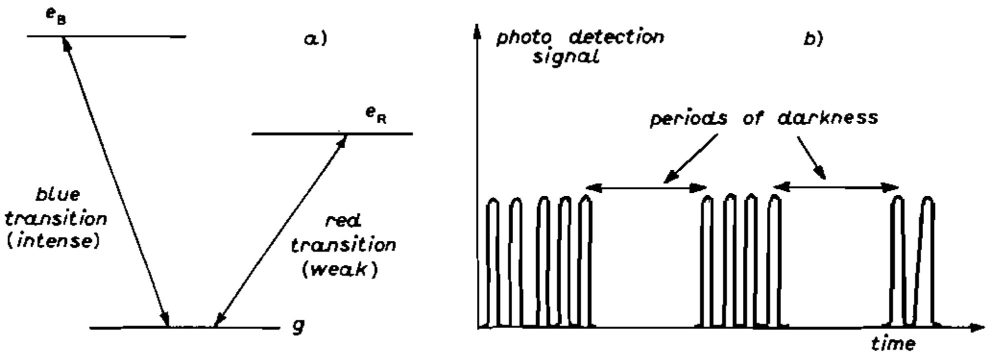

Fig. 1. – *a*) 3-level atom with two transitions starting from the gound state g. *b*) Random sequence of pulses given by a photodetector recording the fluorescence of a single atom. The periods of darkness correspond to the shelving of the electron on the metastable level e_R.

very intense $g \leftrightarrow e_B$ (which we will call for convenience the «red» and the «blue» transitions), and the suppose that two lasers drive these two transitions. When the atom absorbs a red photon, it is «shelved» on e_R, and this switches off the intense blue fluorescence for a time of the order of Γ_R^{-1}. We expect, therefore, in this case that the sequence of pulses given by the broadband photodetector recording the fluorescence light should exhibit «periods of brightness», with closely spaced pulses, corresponding to the intense blue resonance fluorescence, alternated with «periods of darkness» corresponding to the periods of shelving in r_R (fig. 1*b*)). The absorption of one red photon could thus be detected by the *absence* of a large number of blue fluorescence photons [7]. It has been recently pointed out [8] that such a fluorescence signal could provide a direct observation of «quantum jumps» between g and e_R, and several theoretical models have been presented for this effect, using rate equations and random telegraph signal theory [8], or optical Bloch equations and second (and higher) order correlation functions such as g_2 [9-12].

The purpose of this letter is to introduce another statistical function which we will call the delay function w_2 and which, in our opinion, is more suitable than g_2 for the analysis of signals such as the one of fig. 1*b*). We define $w_2(\tau)$ as the probability, if one has detected one photon at time t, to detect *the next one* at time $t+\tau$ (and not *any other one*, as it is the case for g_2) [13]. We suppose for the moment that the detection efficiency is equal to 1, so that w_2 and g_2 refer also to emission processes. The delay function $w_2(\tau)$ is directly related to the repartition of delays τ between two *successive* pulses and thus provides simple evidence for the possible existence of periods of darkness. We would like also to show in this letter that $w_2(\tau)$ is very simple to calculate and is a very convenient tool for extracting all the spectroscopic information contained in the sequence of pulses of fig. 1*b*).

We first introduce, in parallel with $w_2(\tau)$, a related function $P(\tau)$ defined by

$$P(\tau) = 1 - \int_0^\tau d\tau' \, w_2(\tau') \, . \tag{1}$$

From the definition of w_2, it is clear that $P(\tau)$ is the probability for not having any emission of photons between t and $t+\tau$, after the emission of a photon at time t. $P(\tau)$ starts from 1 at $\tau = 0$ and decreases to zero as τ tends to infinity. We now make the hypothesis that P and w_2 evolve in time with at least two very different time constants. More precisely, we suppose that $P(\tau)$ can be written as

$$P(\tau) = P_{\text{short}}(\tau) + P_{\text{long}}(\tau) \, , \tag{2}$$

where

$$P_{\text{long}}(\tau) = p \exp[-\tau/\tau_{\text{long}}], \tag{3}$$

and where $P_{\text{short}}(\tau)$ tends to zero very rapidly, *i.e.* with one (or several) time constant(s) τ_{short} much shorter than τ_{long}. We shall see later on that this splitting effectively occurs for the three-level system described above.

Our main point is that this form for $P(\tau)$ proves the existence of bright and dark periods in the photodetection signal, and furthermore allows the calculations of all their characteristics (average duration, repetition rate, ...). Our analysis directly follows the experimental procedure that one would use in order to exhibit such dark and light periods in the signal. We introduce a time delay θ such as

$$\tau_{\text{short}} \ll \theta \ll \tau_{\text{long}} \tag{4}$$

and we «store» the intervals Δt between successive pulses in two «channels»: the interval Δt is considered as short, if $\Delta t < \theta$, as long, if $\Delta t > \theta$. We now evaluate quantities such as the probability Π for having a long interval after a given pulse and the average durations T_{long} and T_{short} of long and short intervals. If none of these three quantities depends (in first approximation) on θ, this clearly demonstrates the existence of bright periods (*i.e.* succession of short intervals) and of dark ones (*i.e.* occurence of a long interval).

The probability Π for having an interval Δt larger than θ is directly obtained from the function $P: \Pi = P(\theta)$. Using the double inequality (4), we get $P_{\text{short}}(\theta) \simeq 0$ and $P_{\text{long}}(\theta) \simeq P_{\text{long}}(0) = p$, so that

$$\Pi = p\,. \tag{5}$$

The average durations T_{long} and T_{short} of long and short intervals are given by

$$\begin{cases} T_{\text{long}} = \dfrac{1}{\Pi} \displaystyle\int_\theta^\infty \mathrm{d}\tau\, \tau w_2(\tau)\,, \\[2ex] T_{\text{short}} = \dfrac{1}{1-\Pi} \displaystyle\int_0^\theta \mathrm{d}\tau\, \tau w_2(\tau)\,. \end{cases} \tag{6}$$

After an integration by parts, and using again the double inequality (4), this becomes

$$\begin{cases} T_{\text{long}} = \tau_{\text{long}}\,, \\[2ex] T_{\text{short}} = \dfrac{1}{1-p} \displaystyle\int_0^\infty \mathrm{d}\tau\, P_{\text{short}}(\tau)\,. \end{cases} \tag{7}$$

We see that the average length of long intervals is just the long-time constant of $P(\tau)$, while the average length of short intervals is related to the rapidly decreasing part of $P(\tau)$. None of the three quantities obtained in (5) and (7) depends on θ, which indicates the intrinsic existence of dark periods and of bright ones. The average duration of a dark period $\mathcal{T}_{\text{D}}$ is just τ_{long}, while the average duration of a bright period $\mathcal{T}_{\text{B}}$ is the product of the duration of a short interval T_{short} by the average number $\bar{N}$ of consecutive short intervals [2]:

$$\mathcal{T}_{\text{D}} = \tau_{\text{long}} \, , \tag{8a}$$

$$\mathcal{T}_{\text{B}} = T_{\text{short}} \bar{N} \, . \tag{8b}$$

This average number $\bar{N}$ can be written $\sum_N NP_N$, where $P_N = (1-p)^N p$ is the probability for having N short intervals followed by a long one. Actually, the notion of «brightness» for a period has a sense only if it contains many pulses. We are then led to suppose $p \ll 1$, so that

$$\bar{N} = \frac{1-p}{p} \simeq \frac{1}{p} \gg 1 \, . \tag{9}$$

Using (7) and (8b), the length of a bright period can finally be written as

$$\mathcal{T}_{\text{B}} = \frac{1}{p} \int_0^\infty \mathrm{d}\tau \, P_{\text{short}}(\tau) \, . \tag{10}$$

Note that if the efficiency of the detection ε is not 100%, results (8a) and (10) are still valid, provided certain conditions hold. Remark first that in a bright period, the mean number of pulses is multiplied by ε, and that the interval between two successive pulses is divided by ε. In order to still observe dark and bright periods, one has to detect many pulses in a given bright period, and the average delay between two detected pulses must be much shorter than the length of a dark period:

$$\begin{cases} 1 \ll \varepsilon N \, , \\ T_{\text{short}}/\varepsilon \ll T_{\text{long}} \, . \end{cases} \tag{11}$$

Provided these two inequalities are satisfied, it is still possible to detect dark and bright periods, whose lengths are again given by (8a) and (10).

We now tackle the problem of the calculation of w_2 and P for the 3-level atom described above, for which we shall use a dressed-atom approach. Immediately after the detection of a first fluorescence photon at time t, the system is in the state $|\varphi_0\rangle = |g, N_{\text{B}}, N_{\text{R}}\rangle$, *i.e.* atom in the ground state in the presence of N_{B} blue photons and N_{R} red photons. Neglecting antiresonant terms, we see that this state is only coupled by the laser-atom interactions to the two other states $|\varphi_1\rangle = |e_{\text{B}}, N_{\text{B}}-1, N_{\text{R}}\rangle$ and $|\varphi_2\rangle = |e_{\text{R}}, N_{\text{B}}, N_{\text{R}}-1\rangle$ (the atom absorbs a blue or a red photon and jumps from g to e_{B} or e_{R}). These three states form a nearly degenerate 3-dimensional manifold $\mathcal{E}(N_{\text{B}}, N_{\text{R}})$ from which the atom can escape only by emitting a second fluorescence photon. The detection of this photon then projects the atom in a lower manifold. Consequently, the probability $P(\tau)$ for not having any emission of photon between t and $t+\tau$ after the detection of a photon at time t is simply equal to the population of the manifold $\mathcal{E}(N_{\text{B}}, N_{\text{R}})$ at time $t+\tau$ knowing that the system starts from the state $|\varphi_0\rangle$ at time t.

(2) We treat here durations of intervals between pulses as independent variables. This is correct, since two successive intervals are uncorrelated. At the end of a given interval, the detection of a photon projects the atom in the ground state, so that any information concerning the length of this interval is lost. This is to be contrasted with the fact that two successive pulses are correlated (antibunching effect for example).

In order to calculate this population, we look for a solution for the total wave function of the form

$$|\psi(t+\tau)\rangle = \sum_{i=0,1,2} a_i(\tau)|\varphi_i\rangle \times |0 \text{ fluorescence photon}\rangle + \\ + \sum_j b_j(\tau)\ |j\text{: states involving fluorescence photons}\rangle \tag{12}$$

with $a_0(0)=1$, all other coefficients being equal to zero at time t. From (12), we then extract P:

$$P(\tau) = \sum_i |a_i(\tau)|^2 . \tag{13}$$

The equations of motion for the a_i's read

$$\begin{cases} i\dot{a}_0 = \dfrac{\Omega_\mathrm{B}}{2}\, a_1 + \dfrac{\Omega_\mathrm{R}}{2}\, a_2 , \\ i\dot{a}_1 = \dfrac{\Omega_\mathrm{B}}{2}\, a_0 - \left(\Delta_\mathrm{B} + \dfrac{i\Gamma_\mathrm{B}}{2}\right) a_1 , \\ i\dot{a}_2 = \dfrac{\Omega_\mathrm{R}}{2}\, a_0 - \left(\Delta_\mathrm{R} + \dfrac{i\Gamma_\mathrm{R}}{2}\right) a_2 , \end{cases} \tag{14}$$

where Ω_B and Ω_R represent the blue and red Rabi frequencies, $\Delta_\mathrm{B}(\Delta_\mathrm{R})$ the detuning between the blue (red) laser and the blue (red) atomic transition, and where Γ_B and Γ_R are the natural widths of levels e_B and e_R. This differential system is easily solved by Laplace transform, and each $a_i(\tau)$ appears as a superposition of 3 (eventually complex) exponentials. The main result is then that, provided Γ_R and Ω_R are small enough compared to Γ_B and Ω_B, $P(\tau)$ can be written as in (2)-(3): this proves the existence of periods of darkness in the photodetection signal.

We shall not give here the details of the general calculations, and we shall only investigate the two limiting cases of weak and strong blue excitations.

We begin by the low intensity limit ($\Omega_\mathrm{B} \ll \Gamma_\mathrm{B}$, blue transition not saturated). We suppose the blue laser tuned at resonance ($\Delta_\mathrm{B}=0$) and we consider first $\Delta_\mathrm{R}=0$. The system (14) has 3 time constants, 2 short ones τ_1 and τ_2, and a long one τ_3:

$$\frac{1}{\tau_1} = \frac{\Gamma_\mathrm{B}}{2}, \tag{15a}$$

$$\frac{1}{\tau_2} = \frac{\Omega_\mathrm{B}^2}{2\Gamma_\mathrm{B}}, \tag{15b}$$

$$\frac{1}{\tau_3} = \frac{\Gamma_\mathrm{R}}{2} + \frac{\Gamma_\mathrm{B}}{2}\,\frac{\Omega_\mathrm{R}^2}{\Omega_\mathrm{B}^2}. \tag{15c}$$

The weight of τ_2 is predominant in $P_\mathrm{short}(\tau)$ and we find

$$T_\mathrm{short} = \tau_2/2 . \tag{16}$$

Physically, $2/\tau_2$ represents the absorption rate of a blue photon from g to e_B. It can be interpreted as the transition rate given by the Fermi golden rule, with a matrix element

$\Omega_B/2$ and a density of final state $2/\pi\Gamma_B$, and corresponds to the width of the ground state induced by the blue laser. On the other hand, the long time constant in $P(\tau)$ is proportional to τ_3:

$$T_{\text{long}} = \tau_3/2\,. \tag{17}$$

Physically, $2/\tau_3$ represents the departure rate from e_R, due to both spontaneous (first term of (15c)) and stimulated (second term of (15c)) transitions. The second term of (15c) can be written $(\Omega_R/2)^2\tau_2$ and then appears as a Fermi golden rule expression. It gives the stimulated emission rate of a red photon from e_R (matrix element $\Omega_R/2$) to the ground state g broadened by the blue laser (density of states τ_2/π). Note that the condition $T_{\text{long}} \gg T_{\text{short}}$ implies

$$\Gamma_R\,,\ \Omega_R \ll \frac{\Omega_B^2}{\Gamma_B}\,. \tag{18}$$

From now on, we choose Ω_R such that the two spontaneous and stimulated rates of (15c) are equal, and we calculate from (8a) and (10) the variation with the red detuning Δ_R of the ratio $\mathcal{T}_D/\mathcal{T}_B$. We find that this ratio exhibits a resonant variation with Δ_R (fig. 2a))

$$\frac{\mathcal{T}_D}{\mathcal{T}_B} = \frac{1}{2 + (\tau_2\Delta_R)^2}\,. \tag{19}$$

This shows that it is possible to detect the $g - e_R$ resonance by studying the ratio between the lengths of dark and bright periods. Note that this ratio can be as large as $\frac{1}{2}$ (for $\Delta_R = 0$) and that the width of the resonance is determined by the width of the ground state induced by the laser. We have supposed here that $\Delta_B = 0$; if this were not the case, one would get a shift of the resonance given in (19) due to the light shift of g.

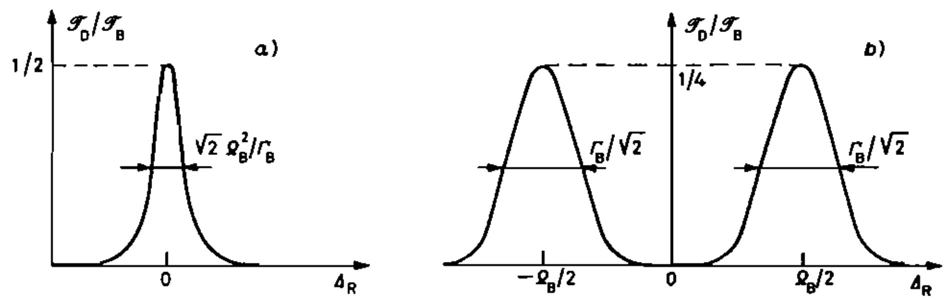

Fig. 2. – Variation with the red laser detuning Δ_R of the ratio $\mathcal{T}_D/\mathcal{T}_B$ between the average lengths of dark and bright periods. a) Weak-intensity limit, b) high-intensity limit.

Consider now the high-intensity limit ($\Omega_B \gg \Gamma_B$, blue transition saturated). We still suppose $\Delta_B = 0$. The two short time constants τ_1 and τ_2 of (14) are now equal to $4/\Gamma_B$, so that $T_{\text{short}} = 2/\Gamma_B$. The corresponding two roots r_1 and r_2 of the characteristic equation of (14)

$$r_1 = -\frac{\Gamma_B}{4} - i\,\frac{\Omega_B}{2}\,, \tag{20a}$$

$$r_2 = -\frac{\Gamma_B}{4} + i\,\frac{\Omega_B}{2}\,, \tag{20b}$$

have now an imaginary part $\pm i\Omega_B/2$, which describes a removal of degeneracy induced in the manifold $\mathscr{E}$ (N_B, N_R) by the atom blue laser coupling: the two unperturbed states $|\varphi_0\rangle$ and $|\varphi_1\rangle$ of $\mathscr{E}$ (N_B, N_R), which are degenerate for $\Delta_B = 0$, are transformed by this coupling into two perturbed dressed states

$$|\psi_\pm\rangle = \frac{1}{\sqrt{2}}\left(|\varphi_0\rangle \pm |\varphi_1\rangle\right), \tag{21}$$

having a width $\Gamma_B/4$ and separated by the well-known dynamical Stark splitting Ω_B [14]. The interaction with the red laser couples the third level $|\varphi_2\rangle$ to $|\psi_\pm\rangle$ with matrix elements $\pm\,\Omega_R/2\sqrt{2}$. This coupling is resonant when $|\psi_2\rangle$ is degenerate with $|\psi_+\rangle$ or $|\psi_-\rangle$, *i.e.* when $\Delta_R = \pm\,\Omega_B/2$. Such a resonant behaviour appears on the general expression of the slow-time constant τ_3 of (14)

$$\frac{1}{\tau_3} = \frac{\Gamma_R}{2} + \frac{\Omega_R^2}{32}\,\frac{\Omega_B^2 \Gamma_B}{(\Omega_B^2/4 - \Delta_R^2)^2 + \Delta_R^2 \Gamma_B^2/4}, \tag{22}$$

which reaches its maximum value

$$\frac{1}{\tau_3} = \frac{\Gamma_R}{2} + \frac{\Omega_R^2}{2\Gamma_B} \tag{23}$$

for $\Delta_R = \pm\Omega_B/2$. As in (15*c*), the first term of (22) or (23) represents the effect of spontaneous transitions from e_R. The second term of (23) can be written as $(\Omega_R/2\sqrt{2})^2 \cdot (4/\Gamma_B)$ and appears as a stimulated emission rate of a red photon from e_R to the broad $|\psi_+\rangle$ or $|\psi_-\rangle$ states. If, as above, we choose Ω_R such that the 2 rates of (23) are equal, we get for $\mathscr{T}_D/\mathscr{T}_B$ the double-peaked structure of fig. 2*b*). The two peaks have a maximum value of $\frac{1}{4}$ and a width $\Gamma_B/\sqrt{2}$ (for $\Delta_R =$ $\mathscr{T}_D/\mathscr{T}_B = \Gamma_B^4/2\Omega_B^4 \ll 1$, so that the weight of the dark periods becomes very small). This shows that measuring in this case the ratio between the lengths of dark and bright periods gives the possibility to detect, on a single atom, the Autler-Townes effect induced on the weak red transition by the intense blue laser excitation.

In conclusion, we have introduced in this paper new statistical functions which allow a simple analysis of the electron shelving scheme proposed by DEHMELT for detecting very weak transitions on a single trapped ion. We have shown that there exist, in the sequence of pulses given by the photodetector recording the fluorescence light, periods of darkness. The average length $\mathscr{T}_D$ of such dark periods, which is determined by the spontaneous and stimulated lifetimes of the shelving state, can reach values of the order of the average length $\mathscr{T}_B$ of the bright periods. They should then be clearly visible on the recording of the fluorescence signal. We have also shown that it is possible to get spectroscopic information by plotting the ratio $\mathscr{T}_D/\mathscr{T}_B$ *vs.* the detuning of the laser driving the weak transition. The smallest width obtained in this way is the width of the ground state due to the intense laser. Note that this width is still large compared to the natural width of the shelving state. It is clear that, in order to get resonances as narrow as possible, the two lasers should be alternated in time.

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