Upper bound on the duration of quantum jumps - Supplementary Material

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Both the concept of quantum jumps and context it is presented on has evolved as the understanding of quantum theory and open systems has increased. Electron shelving experiments in the 1980's refocused attention on quantum jumps. In those experiments, it was observed that individual realizations of a given atom-electromagnetic field system yield random sequences of light and dark intervals in the fluorescence: strongly fluorescent transitions were suddenly interrupted by the atom being temporarily shelved in a metastable level. Most interest on the interpretation of those experiments was paid to the duration of the light and dark periods, and to the photon statistics of the emitted light.

In the scheme presented in the main text, the information regarding the time behavior of quantum jumps is obtained indirectly via time correlation of photodetection events. The photons emerge from an atomic cascade decay having a well-defined time order with signal photon being generated before the idler photon. The set-up involves two notable time scales:

(i) those related to the transition from state $|2\rangle$ to state $|3\rangle$ and of the subsequent transition to the ground state $|0\rangle$ (this corresponds to two successive quantum jumps),

(ii) those related to the delay $\tau = t_i - t_s$ between the emission of the idler photon and the signal photon.

This work reports a measurement of an upper bound to the time scales of the first kind, and focuses on the first quantum jump associated with the onset of the cascade decay.

A standard quantum optics description, summarized in Eqs. (1-7) in the main text, yields a correlation function that incorporates the monitored transition —or quantum jump— from state $|2\rangle$ to $|3\rangle$ via a discontinuous Heaviside function. A finite time duration of such a quantum jump would be manifest by the replacement of the Heaviside function by a continuous function. In this Supplemental Material section, details are given about the main steps involved in the derivation of Eqs. (1-7). This has the purpose of showing that the discontinuity of the photon two time correlation function predicted by standard quantum optics has a structural origin on the boundary conditions that specify the density matrices ρ and ρ'. The derivation helps to clarify the role of the different parameters involved in the experimental set-up ρ, while emphasizing that the quantum jumps analized here are processes of the joined atom-electromagnetic field system.

Correlation between field and atomic operators

The generation and ensuing detection of idler and signal photons is directly correlated to transitions between different atomic levels. In order to show this connection explicitly we consider the interaction between atom and the surrounding electromagnetic environment under the dipole approximation. It is described by the Hamiltonian

$$\mathcal{H} = \sum_n \omega_n \hat{\sigma}_{nn} + \sum_{\kappa,j} \omega_k \hat{a}^\dagger_{\kappa,j}\hat{a}_{\kappa,j} + \sum_{\kappa,j}\sum_{n,m}\sqrt{\frac{2\pi\omega_k}{V}}\epsilon_j\cdot\mathbf{d}_{nm}(e^{i\mathbf{k}\cdot\mathbf{x}}\hat{a}_{\kappa,j} + e^{-i\mathbf{k}\cdot\mathbf{x}}\hat{a}^\dagger_{\kappa,j})\left(\hat{\sigma}_{nm}+\hat{\sigma}^\dagger_{nm}\right) \tag{1}$$

where the electromagnetic modes are characterized by their frequency ω_κ, polarization ϵ_j, and set of dynamical variables κ (in this case wave vector); they are coupled to the atom through the coupling strengths determined by the projection of the electric field component along the dipole moment $\mathbf{d}_{nm}$. In the Heisenberg picture, the field modes satisfy

$$\dot{\hat{a}}_{\kappa,j} = -i\omega_\kappa\hat{a}_{\kappa,j} - i\sqrt{\frac{2\pi\omega_\kappa}{V}}\sum_{n,m}\epsilon_j\cdot\mathbf{d}_{nm}\left(\hat{\sigma}_{nm}+\hat{\sigma}^\dagger_{nm}\right)\,, \tag{2}$$

The field operators obtained from this mode decomposition can then be written as a sum of free-field operators $\mathbf{E}_f^{(+)}$ obtained for zero coupling, and source-field operators $\mathbf{E}_s^{(+)}$ accounting for the radiated field by the atom. The corresponding mode operators $\hat{a}_{\kappa,j}$ reflect such an structure,

$$\hat{a}_{\kappa,j}(t) = e^{-i\omega_\kappa t}\hat{a}_{\kappa,j}(t_0) - i\sqrt{\frac{2\pi\omega_\kappa}{V}}\sum_{n,m}\epsilon_i\cdot\mathbf{d}_{nm}\int_{t_0}^{t}d\tau e^{-i\omega_\kappa(t-\tau)-i\mathbf{k}\cdot\mathbf{x}}\left(\hat{\sigma}_{nm}(\tau)+\hat{\sigma}_{nm}^\dagger(\tau)\right). \quad (3)$$

The explicit effect of the source contributions can be obtained by considering a large environment from which the sum over modes is changed into an integral which can be solved directly. Using the Born-Markov and rotating wave approximations it is possible to solve for the resulting spatial and temporal integrals [1, 2]. The result will cause for field modes centered at the atomic frequency and displaying nonzero projection of the electric field to the dipole moment to be the only modes populated by the source term.

The correlation function given by Eq. (1) in the main text,

$$\mathcal{C}(t,t+\tau) = \langle\hat{a}_s^\dagger(t)\hat{a}_i^\dagger(t+\tau)\hat{a}_i(t+\tau)\hat{a}_s(t)\rangle,$$

is then proportional to atomic polarization correlation

$$\mathcal{P}(t,t+\tau) = \langle\hat{\sigma}_{23}^\dagger(t)\hat{\sigma}_{30}^\dagger(t+\tau)\hat{\sigma}_{30}(t+\tau)\hat{\sigma}_{23}(t)\rangle \quad (4)$$

for fields initially in the vacuum state, as a consequence of Eq. (3) and the above mentioned approximations. Thus establishing the connection between Eq. (1) and (2) in the main text, and a first relation of the transitions to the photodetectors.

The time evolution of the atomic state, due to the spontaneous character of the transitions of interest, is described by an atomic density matrix satisfying a Lindblad like equation. Using the quantum regression theorem it is possible to transform Eq. (4) into

$$\mathcal{P}(t,t+\tau) = \rho_{22}(t)\rho_{33}'(\tau)\,, \quad (5)$$

where $\rho_{22}(t)$ represents the population of state $|2\rangle$ at time t, while $\rho_{33}'(\tau)$ gives the population of state $|3\rangle$ at time $t+\tau$ for an auxiliary density matrix that resets the origin of time to the value t and satisfies the initial condition $\rho'(\tau=0)=|3\rangle\langle 3|$. This condition underlines the well-defined time order that allows us to study quantum jumps in the four-wave mixing process. Notice that $\mathcal{P}(t,t+\tau)$ also represents, for a single atom, the probability of detecting the next photon emitted by the system, *i.e.* the waiting time distribution function.

Due to the relevance of Eq. (5) we review its derivation assuming the existence of a unitary evolution operator $\hat{U}$. Consider that at the initial time t_0 $\hat{\sigma}_{ij}(t_0)=|j\rangle\langle i|$ and the general properties of the trace,

$$\begin{aligned}
\mathcal{P}(t,t+\tau) &= \langle\hat{\sigma}_{23}^\dagger(t)\hat{\sigma}_{30}^\dagger(t+\tau)\hat{\sigma}_{30}(t+\tau)\hat{\sigma}_{23}(t)\rangle \qquad (6)\\
&= \mathrm{Tr}\left[\hat{\sigma}_{23}^\dagger(t)\hat{\sigma}_{30}^\dagger(t+\tau)\hat{\sigma}_{30}(t+\tau)\hat{\sigma}_{23}(t)\hat{\rho}(t_0)\right]\\
&= \mathrm{Tr}\left[\hat{U}(t+\tau,t_0)\hat{\sigma}_{30}^\dagger(t_0)\hat{\sigma}_{30}(t_0)\hat{U}^{-1}(t+\tau,t_0)\right.\\
&\cdot\ \left.\hat{U}(t,t_0)\hat{\sigma}_{23}(t_0)\hat{U}^{-1}(t,t_0)\hat{\rho}(t_0)\hat{U}(t,t_0)\sigma_{23}^\dagger(t_0)\hat{U}^{-1}(t,t_0)\right]\\
&= \rho_{22}(t)\mathrm{Tr}\left[\hat{U}(t+\tau,t_0)\hat{\sigma}_{30}^\dagger(t_0)\sigma_{30}(t_0)\hat{U}^{-1}(t+\tau,t_0)\hat{U}(t,t_0)|3\rangle\langle 3|\hat{U}^{-1}(t,t_0)\right]\\
&= \rho_{22}(t)\mathrm{Tr}\left[\hat{U}(t+\tau,t)\hat{|}3\rangle\langle 3|\hat{U}^{-1}(t+\tau,t)|3\rangle\langle 3|\right]\\
&= \rho_{22}(t)\rho_{33}'(t+\tau)\,.
\end{aligned}$$

Master equations for the diamond level configuration.

We consider a four-level system in the diamond configuration as depicted in Fig. 1 in the main text. Here, the atomic ground state $5S_{1/2}$ is coupled to an excited state $5D_{3/2}$ through a two-step excitation (spontaneous decay) process involving the intermediate $5P_{3/2}$ ($5P_{1/2}$) level. The intra-level coupling is mediated by the electromagnetic field, composed of a quantized electromagnetic environment and the classical fields produced by a pair of lasers. In particular, the non-resonant laser fields drive the $5S_{1/2} \leftrightarrow 5P_{3/2}$ and $5P_{3/2} \leftrightarrow 5D_{3/2}$ transitions, while coupling to the vacuum modes induce the $5D_{3/2} \rightarrow 5P_{1/2} \rightarrow 5S_{1/2}$ de-excitation path.

Interaction with the nuclear spin gives rise to hyperfine levels which are selectively populated using driving lasers with adequate bandwidth and polarization. The relative populations are determined by the field polarization and atomic selection rules. The Rabi frequencies and decay rates defining each path can be determined using the Wigner-Eckhard theorem, we consider an unpolarized laser beam in the calculations.

The evolution of the system in the prescribed diamond configuration is ruled by the master equation

$$\dot{\rho} = (i\hbar)^{-1}\left[\mathcal{H},\rho\right] + \mathcal{L}_{\rm spon}\rho \tag{7}$$

with Hamiltonian

$$\begin{aligned}\mathcal{H} = & \sum_{n=1}^{3}\{\hbar\omega_n\hat{\sigma}_{nn}\} + [\frac{\Omega_{01}}{2}e^{-i\omega_{L1}}\hat{\sigma}_{01} + \text{H.c.}] \\ & +[\frac{\Omega_{12}}{2}e^{-i\omega_{L2}}\hat{\sigma}_{12} + \text{H.c.}]\,,\end{aligned} \tag{8}$$

where the transition frequencies ω_n and laser-mediated interactions with Rabi frequency Ω_{ij} are included, while spontaneous emissions are considered through the Lindblad term

$$\mathcal{L}_{\rm spon} = \sum_{n,m}\gamma_{nm}\left[2\hat{\sigma}_{nm}\rho\hat{\sigma}^{\dagger}_{nm} - \hat{\sigma}_{nn}\rho - \rho\hat{\sigma}_{nn}\right]\,. \tag{9}$$

with γ_{nm} the inverse decay rate from level n to m. We neglect Doppler broadenning effects and other dephasing factors.

The experimental conditions allow for the adiabatic elimination of the level $|1\rangle$ as described in the main text. This is related to the fact that the experimental set up yields a majoritary population of the excited level $5D_{3/2}$ F= 3, and due to the selection of detunings and laser powers

$$|\Delta_{nl}|, \Omega_{0n}, \Omega_{nm}, \gamma_{nm}, \gamma_{10} \gg \gamma_{21}\,, \tag{10}$$

the decay to the ground state is mainly through the $5P_{1/2}$ F=2 state. The validity of the adiabatic elimination in this regime was confirmed by numerical simulations involving the four levels.

Temporal evolution of the effective three-level system

We now present a theoretical analysis that properly describes the temporal evolution of the effective three-level system evolving under the action of Eq. (4) in the main text. From this evolution, the coefficients $\rho_{ij} = \langle i|\rho|j\rangle$ satisfy a set of coupled linear differential equations. This set of equations can be transformed into an algebraic one using the Laplace transform

$$\tilde{\rho}_{ij}(s) \equiv \mathcal{L}[\rho_{ij}(t)] \equiv \int_0^\infty dt\rho_{ij}(t)e^{-st}, \tag{11}$$

with s a complex variable, since the Laplace transform of the time derivative $\dot{\rho}_{ij}(t)$ with a predetermined boundary condition at a given initial time $t = 0$ satisfies the relation [3]

$$\begin{aligned}\mathcal{L}[\dot{\rho}_{ij}(t)] &= \rho_{ij}(t)e^{-st}\Big|_0^\infty + s\int_0^\infty dt\rho_{ij}(t)e^{-st} \\ &= -\rho_{ij}(0) + s\tilde{\rho}(s)\,.\end{aligned} \tag{12}$$

Being interested in the correlation function Eq.(5), we solve this set of equations for $\tilde{\rho}_{22}$ and $\tilde{\rho}'_{33}$ subject to different initial conditions; then, we apply the inverse Laplace transform to obtain the time dependence of these elements. For an initial state $\rho_{00}(t=0) = 1$ the algebraic equations lead to

$$Q(s)\tilde{\rho}_{22}(s) = \frac{1}{2}\Omega^2_{\rm eff}\left(s+\gamma_{30}\right)\left(s+\frac{1}{2}\gamma_{23}\right)s^{-1}\,, \tag{13}$$

$$Q(s)\tilde{\rho}_{33}(s) = \frac{1}{2}\Omega^2_{\rm eff}\gamma_{23}\left(s+\frac{1}{2}\gamma_{23}\right)s^{-1}\,, \tag{14}$$

$c_{22}^{(1)} = -0.01546$	$c_{22}^{(2)} = 0$	$c_{22}^{(3)} = 0.49333$	$c_{22}^{(4)} = 0.00718$
$c_{33}^{(1)} = -0.96753$	$c_{33}^{(2)} = -0.00034$	$c_{33}^{(3)} = 0.00001$	$c_{33}^{(4)} = 0.00035$

TABLE I: Numerical coefficients that determine $\rho_{22}(t)$, Eq. (22) and $\rho'_{33}(\tau)$, Eq.(22).

with the fourth-order polynomial

$$Q(s) = \left((s + \frac{\gamma_{23}}{2})^2 + \Delta_{\text{eff}}^2\right)(s+\gamma_{23})(s+\gamma_{30}) + \Omega_{\text{eff}}^2(s + \frac{\gamma_{23}}{2})(s + \gamma_{30} + \frac{\gamma_{23}}{2})\,. \tag{15}$$

Similarly, for $\rho'_{33}(\tau = 0) = 1$ one obtains

$$Q(s)\tilde{\rho}'_{22}(s) = \frac{1}{2}\gamma_{30}\Omega_{\text{eff}}^2(s + \frac{1}{2}\gamma_{23})s^{-1}\,, \tag{16}$$

$$Q(s)\tilde{\rho}'_{33}(s) = \left[\left((s + \frac{1}{2}\gamma_{23})^2 + \Delta_{\text{eff}}^2\right)(s+\gamma_{23})s + \Omega_{\text{eff}}^2\left(s + \frac{1}{2}\gamma_{23}\right)^2\right]s^{-1}\,. \tag{17}$$

The time dependence of the density matrix elements will be given by the nature of the roots of Eqs. (13-16).

According to the experimental data, and using the electronic structure of ^{87}Rb atoms,

$$\begin{aligned} \Omega_{\text{eff}} &= 163.04\text{MHz} \quad , \quad \Delta_{\text{eff}} = -16.2\text{MHz} \\ \gamma_{23} &= 2.436\text{MHz} \quad , \quad \gamma_{30} = 35.92\text{MHz}. \end{aligned} \tag{18}$$

This results in that two of the roots of $Q(s)$ are real while the other two are complex numbers, the latter being complex conjugate of each other:

$$r_1 = -37.025\text{MHz} \sim -(\gamma_{30} + \gamma_{23}/2) \tag{19}$$

$$r_2 = -1.2306\text{MHz} \sim -(\gamma_{23}/2) \tag{20}$$

$$\begin{aligned} r_{3,4} &= -1.2445 \pm 163.984i\text{MHz} \\ &= -\tilde{\gamma} \pm i\tilde{\Omega} \\ &\sim -\frac{\gamma_{23}}{2} \pm \sqrt{\Omega_{\text{eff}}^2 + \Delta_{\text{eff}}^2 + \frac{\gamma_{23}}{2}(\gamma_{30} - \frac{\gamma_{23}}{2})}i \end{aligned} \tag{21}$$

These roots determine the general features of $\rho_{22}(t)$ and $\rho'_{33}(\tau)$ that translate into the exponential and oscillatory aspects of the correlation function $\mathcal{P}(t, t+\tau)$. Since the two real roots of $Q(s)$ are negative

$$\rho_{22}(t) = 0, \qquad \text{if} \quad t < 0,$$

$$\rho'_{33}(\tau) = 0, \qquad \text{if} \quad \tau < 0.$$

For $t > 0$

$$\begin{aligned} \rho_{22}(t) &= \quad c_{22}^{(1)}(1 - e^{r_1 t}) + c_{22}^{(2)}(1 - e^{r_2 t}) + c_{22}^{(3)}(1 - \cos(\tilde{\Omega}t)e^{\tilde{\gamma}t}) \\ &+ \ c_{22}^{(4)}\sin(\tilde{\Omega}t)e^{\tilde{\gamma}t}\,, \end{aligned} \tag{22}$$

so that $\rho_{22}(t \to 0^+) = 0$. For $\tau > 0$

$$\begin{aligned} \rho'_{33}(\tau) &= 1 + c_{33}^{(1)}(1 - e^{r_1\tau}) + c_{33}^{(2)}(1 - e^{r_2\tau}) + c_{33}^{(3)}(1 - \cos(\tilde{\Omega}\tau)e^{\tilde{\gamma}\tau}) \\ &+ \ c_{33}^{(4)}\sin(\tilde{\Omega}\tau)e^{\tilde{\gamma}\tau}. \end{aligned} \tag{23}$$

The coefficients resulting from the experimental set-up are shown in Table I. From these values it is expected that asymptotically $\rho'_{33}(\infty) \to 0.03207$ and $\rho_{22}(\infty) \to 0.47787$. The ideal asymptotic $\mathcal{P}(t;\infty)$ function corresponds to state with a population of about 3% for level $|3\rangle$ and about 0.48% for level $|2\rangle$. At long times ρ'_{33} has an oscillatory behaviour with a small amplitude $\sim c_{33}^{(4)}$ and frequency $\tilde{\Omega}$.

While $\rho'_{33}(\tau)$ exhibits a discontinuity at $\tau = 0$, $\rho_{22}(t)$ is continuos at $t = 0$. The discontinuity of $\rho'_{33}(\tau)$ gives rise to the implicit presence of a Heaviside function in Eq. (7) of the main text. It reflects that before time t the atom–field can be considered to be in a superposition of states that involves, among other states, that of an atom in state $|2\rangle$ and no photons in the EM modes associated to the transition $|2\rangle \rightarrow |3\rangle$ and an atomic state $|3\rangle$ with a single photon in one of these modes.

The discontinuity, due to the time boundary condition on ρ', has a structural character. This was verified by numerical calculations that were performed involving not just the four levels here mentioned, but all their hyperfine sublevels.

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[1] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics* (Cambridge University Press, 1995), Sec. 15.5.4.
[2] H. J. Carmichael, *Statistical Methods in Quantum Optics 1: Master Equations and Fokker-Planck Equations* (Springer-Verlag, 1999), Chap. 2.
[3] Whenever $\rho_{ij}(t)e^{-st} \rightarrow 0$ for $t \rightarrow \infty$.