Upper bound on the duration of quantum jumps

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We present a method to estimate the time scale of quantum jumps from the time correlation of photon pairs generated from a cascade decay in an atomic system, and realize it experimentally in a cold cloud of ^{87}Rb. Taking into account the photodetector response, we find an upper bound for the duration of a quantum jump of 21 ± 11 ps.

Introduction – The concept of quantum jumps is traced back to Bohr and the old quantum theory [1]. This theory found its bases on Planck's hypothesis of energy quanta and lead to successful explanations of the photoelectric effect, discrete atomic spectra, and distribution of a blackbody in thermal equilibrium. While it described the discrete states of systems accurately, the theory raised questions regarding the transition periods—or quantum jumps—between states. The periods occurred at random times and, by considering intermediate states forbidden, they had to be instantaneous. The general inadequacy of the old quantum theory appeared to be fixed by wave mechanics [2]; yet, for quantum systems under observation, it has not been possible to depart completely from the quantum jump concept [3].

The idea for early observations of quantum jumps was seeded on Dehmelt's electron shelving proposal [4], where the fluorescence of a driven two-state system is abruptly interrupted as the system transitions to a third, metastable, state. This scheme has been implemented for single trapped ions [5–7] and neutral atoms [8]. Recent quantum jump experiments involve nuclear spins [9], cavity–[10] and circuit [11, 12] quantum electrodynamics architectures. In all these demonstrations, the evolution of the monitored variable has the form of a telegraphic signal [13] presenting on and off times whose duration can be described quantitatively through a study of the waiting time-distribution [14] and applying photodetection theory [15, 16]. Since the probability for a system to be found in a particular state is conditioned to a given measurement record, the inferred transition times reflect the way the system is monitored [17, 18]. Apart from studies based on the photoelectric effect [19, 20], the time structure of the rise and descend transitions has been rarely explored at the limits of contemporary experimental capabilities [12].

In this work we focus on quantum jumps associated to spontaneous transitions involving discrete states. We consider an alternative configuration for the observation of quantum jumps in atomic systems: a monitored cascade three-level system [16, 21]. Contrary to the shelving configuration, where the transition to the metastable state is inferred from the absence of a fluorescent signal, i.e., a null measurement [22, 23], state information in a cascade system is acquired through the detection of correlated photon pairs with a well-defined time ordering. This, coupled with the fact that phase matching conditions allow for photon coincidences to be accurately measured, makes the cascade configuration ideal to experimentally investigate the discontinuous changes in atomic states.

We observe the quantum jumps in the second order time correlation function of the fluorescence generated by a cascade-based four-wave mixing (FWM) in a cold ensemble of ^{87}Rb, and model the experiment as a cascade three-level system via the adiabatic elimination of one of the intermediate states. To improve the timing precision for observing any possible jump dynamics, we take into account the measured impulse response of the single photon detectors for estimating an upper bound on the duration of the quantum jump from the measured time correlation.

Theory – Consider a four-level system in the diamond configuration as depicted in Fig. 1. For ^{87}Rb, the atomic ground state $5S_{1/2}$, level $|0\rangle$, is coupled to an excited state $5D_{3/2}$, $|2\rangle$, through a two-step excitation and decay process involving the intermediate levels $5P_{3/2}$, $|1\rangle$, and $5P_{1/2}$, $|3\rangle$. The excitation path $|0\rangle \leftrightarrow |1\rangle \leftrightarrow |2\rangle$ is coherently driven by a pair of non-resonant lasers, while coupling to the electromagnetic environment induces the de-excitation path $|2\rangle \rightarrow |3\rangle \rightarrow |0\rangle$ through spontaneous emission of idler and signal photons. Due to the collective nature of the process, a phase-matching condition of the cascade emission allows to efficiently collect the light used to monitor the atomic state.

We establish a relation between the atomic system and the measurement apparatus by considering the correlation function related to the joint probability distribution of detecting a heralding photon in mode s at time t and a correlated photon in mode i at time $t + \Delta t$

$$C(t, t+\Delta t) = \langle \hat{a}_s^\dagger(t)\hat{a}_i^\dagger(t+\Delta t)\hat{a}_i(t+\Delta t)\hat{a}_s(t)\rangle \ , \quad (1)$$

with the annihilation and creation operators $\hat{a}_{s,i}$ and $\hat{a}_{s,i}^\dagger$

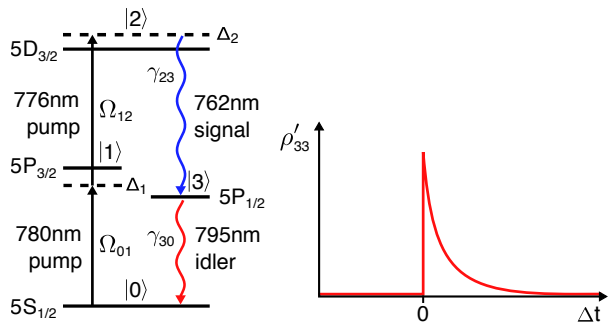

FIG. 1: (Left) Atomic level configuration in a four-wave mixing experiment. (Right) Time evolution of the population of level $|3\rangle$, conditioned on the detection of a signal photon according to Eq. (7).

of the s and i modes. For an electromagnetic environment in the vacuum state, the correlation function C is proportional to the atomic polarization correlation

$$P(t,t+\Delta t) = \langle \hat{\sigma}_{23}^{\dagger}(t)\hat{\sigma}_{30}^{\dagger}(t+\Delta t)\hat{\sigma}_{30}(t+\Delta t)\hat{\sigma}_{23}(t)\rangle\,, \quad (2)$$

with the lowering operator $\hat{\sigma}_{ij} \equiv |j\rangle\langle i|$ describing transitions from the atomic level $|i\rangle$ to level $|j\rangle$ in the Heisenberg picture. Using the quantum regression theorem [24] it is possible to show that

$$P(t,t+\Delta t) = \rho_{22}(t)\rho'_{33}(t+\Delta t)\,, \quad (3)$$

so that atomic density matrix element $\rho_{22}(t)$ represents the population of state $|2\rangle$ at time t, while $\rho'_{33}(t+\Delta t)$ that of state $|3\rangle$ at time $t+\Delta t$ under the initial condition $\rho'(t) = |3\rangle\langle 3|$. This result reflects the well-defined time order that allows the study of quantum jumps in the four-wave mixing process. A detailed analysis of the approximation adopted in this derivation, and in the following analytical treatment is included in the supplementary material [25].

It is possible to obtain an analytical expression for $P(t,t+\Delta t)$; its derivation, however, is greatly simplified if the intermediate level $|1\rangle$ is adiabatically eliminated. This elimination is valid for far-detuned lasers, which induce rapid oscillations on the probability amplitude to find the system in the intermediate level and allow us to consider its zero average value. Under this approximation, the system maps to an effective three-level cascade system with an evolution ruled by the master equation:

$$\dot{\rho} = (i\hbar)^{-1}\left[\mathcal{H}_{\rm eff},\rho\right] + \sum_{n,m}\gamma_{nm}\left[2\hat{\sigma}_{nm}\rho\hat{\sigma}_{nm}^{\dagger} - \hat{\sigma}_{nn}\rho - \rho\hat{\sigma}_{nn}\right]\,, \quad (4)$$

with the effective Hamiltonian

$$\mathcal{H}_{\rm eff} = \hbar\Delta_{\rm eff}|2\rangle\langle 2| + \sum_{i=2,3}E_i|i\rangle\langle i| + \frac{\hbar\Omega_{\rm eff}}{2}\left(|2\rangle\langle 0| + |0\rangle\langle 2|\right)\,, \quad (5)$$

and effective detuning and Rabi frequency

$$\Delta_{\rm eff} = \Delta_2 + \frac{\Omega_{01}^2}{4\Delta_1} - \frac{\Omega_{12}^2}{4\Delta_1} \quad \text{and} \quad \Omega_{\rm eff} = -\frac{\Omega_{01}\Omega_{12}}{2\Delta_1}\,. \quad (6)$$

In these expressions, Ω_{ij} denote the bare Rabi frequencies for the induced $|i\rangle \leftrightarrow |j\rangle$ transitions, γ_{ij} the spontaneous decay rates, and Δ_i the detuning between the driving lasers and the atomic resonances $(E_i - E_j)/\hbar$. The master equation evolution is equivalent to a solvable set of algebraic equations obtained using the Laplace transform, allowing for the calculation of analytical expressions that describe the evolution of the density matrix. With the initial condition $\rho'_{33}(t_0) = 1$ where t_0 is the time at which the signal photon is detected, and for the experimental parameters shown below one finds

$$\begin{aligned} &\rho'_{33}(t_0+\Delta t) = 0 && \text{for } \Delta t < 0\,, \\ &\rho'_{33}(t_0+\Delta t) \approx e^{-\left(\gamma_{30}+\frac{\gamma_{23}}{2}\right)\Delta t} && \text{for } \Delta t > 0\,, \end{aligned} \quad (7)$$

while $\rho_{22}(t)$ is a continuous function of t. Thus, the correlation function $P(t,t+\Delta t)$ exhibits a discontinuity at $\Delta t = 0$ that reflects the breaking of time symmetry induced by the quantum jump associated to the transition $|2\rangle \to |3\rangle$. For $\Delta t > 0$ the atom can perform a second jump to the ground state with statistics given by ρ'_{33} in Eq. (7).

Signal and idler photons impinge on photon detectors that, while able to distinguish between either mode, are unable to determine with certainty the hyperfine level they were emitted from. This indeterminacy causes that the atomic state conditioned to the detection of a signal photon is given by a superposition of the hyperfine states of level $|1\rangle$, i.e., $5P_{1/2}$ $F = 1$ and $F = 2$ states. This yields oscillations of $\rho'_{33}(t_0+\Delta t)$ for $\Delta t > 0$ with a frequency determined by the beating frequencies of these hyperfine levels [26]. These quantum beats can be well reproduced with our model by numerically solving the Bloch equations for a density matrix that involves all hyperfine sublevels of the $|i\rangle$, i=0,1,2,3 states. The frequency and amplitude of the quantum beats are highly sensitive to the polarization, intensity and detuning of the pump lasers. Details of the calculations will be provided in a forthcoming publication. Important to notice here is that the observation of quantum beats provides a striking evidence of quantum coherence.

Experiment – Figure 2 shows schematically the experimental setup for generating time-ordered photon pairs by four-wave mixing in a cold ensemble of ^{87}Rb atoms. Pump beams at 780 nm and 776 nm excite atoms from the $5S_{1/2}, F = 2$ ground level to the $5D_{3/2}, F = 3$ level via a two-photon transition. The signal (762 nm) and idler (795 nm) photons emerge from a cascade decay back to the ground level through the $5P_{1/2}$ level, and are coupled to single mode fibers. Phase matching between pump and target modes is ensured with all four modes propagating in the same direction. The two pump modes

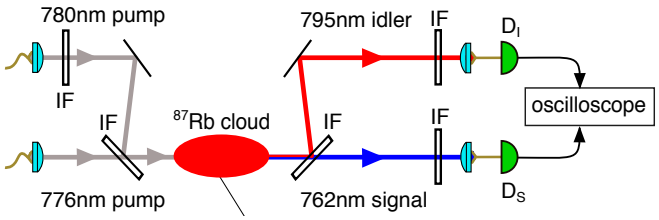

FIG. 2: Schematic of the four-wave mixing experiment. IF: interference filters to combine pump beams and to separate the photons pairs; D_S, D_I: silicon avalanche photodiodes (APD).

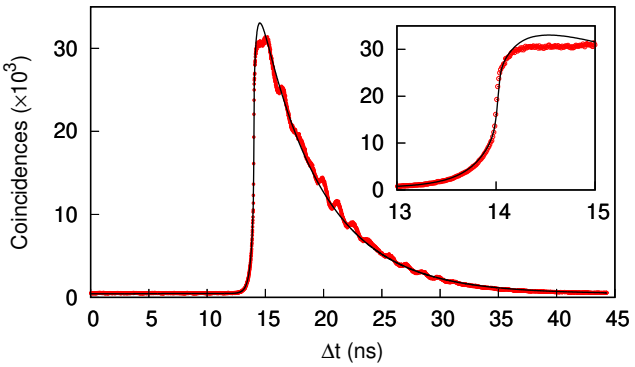

FIG. 3: Histogram $G_{\mathrm{FWM}}(\Delta t)$ of detection time differences for photons pairs generated by four-wave mixing in the cold cloud of ^{87}Rb. The continuous line shows the result of the best fit of Eq. (10). Inset: detail of the sharp rise corresponding to a quantum jump.

are almost collimated and have a Gaussian beam waist of about 400 μm in the atomic cloud. The linearly polarized pump mode at 780 nm is red-detuned by 40 MHz from the $5S_{1/2}, F = 2$ to $5P_{3/2}, F = 3$ transition and has an optical power of 0.5 mW. The orthogonally polarized pump mode at 776 nm shares the same optical mode, has an optical power of 6.5 mW, and is tuned such that the two-photon excitation is blue-detuned by 4 MHz from the difference between the ground state and the $5D_{3/2}, F = 3$ level. We record the detection event time differences of the photon pairs with a digital oscilloscope (sampling rate $4 \times 10^{10}\,\mathrm{s}^{-1}$) with an effective time resolution below 10 ps; the single photon avalanche detectors themselves have a nominal timing jitter around 50 ps FWHM.

Figure 3 shows the histogram $G_{\mathrm{FWM}}(\Delta t)$ of signal and idler photodetection time differences Δt into 10 ps wide bins. The expected exponential decay starts at $\Delta t_0 \approx 14$ ns due to technical delays through fibers and cables. The decay time constant is shorter than the natural decay time $1/\gamma_{30} \approx 27$ ns and determined by the number of atoms involved and the detuning of the pump fields [27, 28]. The oscillations on top of the exponential decay are due to quantum beats between the transition of interest and alternative decay paths involving nearby

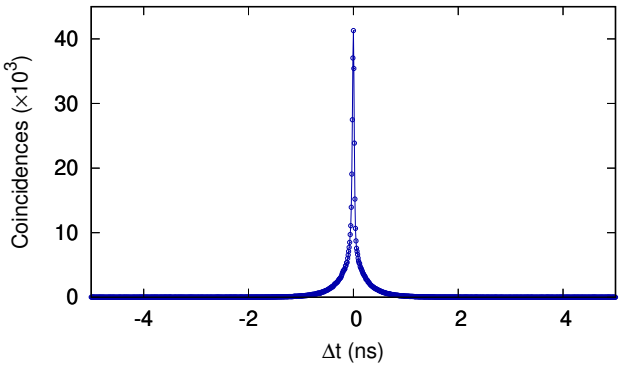

FIG. 4: Histogram $G_{\mathrm{D}}(\Delta t)$ of the difference in detection time for photons pairs generated by spontaneous parametric down-conversion in a nonlinear optical crystal. The bandwidth of the photon pair exceeds 20 nm, corresponding to a coherence time of about 0.1 ps. Thus, we can take the measured time correlation as a reasonable approximation for the impulse response of the two-detector system.

hyperfine levels [26]. The onset of the fluorescence in the idler mode, reflecting the quantum jump, shows a rise time much faster than the time scales of the transitions involved, commensurate with the time jitter of the avalanche photodetectors (see inset of Fig. 3).

To improve the time resolution for observing the quantum jump, we characterize the detector response function. For this, we direct photon pairs with a large optical bandwidth generated by spontaneous parametric down-conversion (SPDC) in a nonlinear optical crystal onto the two avalanche photodetectors. The SPDC source is based on β-Barium Borate, cut for non-degenerate type-I phase matching. Pumped with light at 405 nm, it generates time-correlated photon pairs at 770 nm and 854 nm [29]. The measured bandwidths (23 nm and 32 nm, respectively) correspond to a coherence time of the down-converted photon pairs below 0.1 ps, a negligible contribution to $G_{\mathrm{D}}(\Delta t)$ in comparison to the detector response time scale. The resulting calibration coincidence histogram $G_{\mathrm{D}}(\Delta t)$ into 10 ps wide bins is shown in Fig. 4. Despite the wavelength difference, $G_{\mathrm{D}}(\Delta t)$ does not show any appreciable asymmetry. From this, we infer that the timing response of the detectors does not vary significantly over this wavelength range, and we thus expect that the measured behavior is a good approximation of the detector timing characteristics for photons at 762 nm and 795 nm.

Result – While the structure of $G_{\mathrm{FWM}}(\Delta t)$ away from $\Delta t = 0$ is well understood, we have no model for a possible dynamic of the jump itself. In order to associate a time scale to the transient behavior, we join the two parts of Eq. (7) with a smooth heuristic transition

$$\sigma(x) = \frac{1}{1 + \exp(-x)}\,. \qquad (8)$$

The choice of $\sigma(x)$ is not inspired by a specific dynamical model, its only purpose is to establish a time scale for the transient. This particular form is attractive since it admits the step function as a limiting scenario. The atomic state, with the exponential decay with time constant τ described by Eq. (7), is then enclosed in the monitor function

$$y(\Delta t;\alpha,\tau) \;=\; \sigma\!\left(\frac{\Delta t}{\alpha}\right)\exp\left(-\frac{\Delta t}{\tau}\right), \qquad (9)$$

where the jump timescale is characterized by α.

By convolving the monitoring function $y(\Delta t)$ in Eq. (9) with the normalized measured detector response $g_{\rm D}(\Delta t) = G_{\rm D}(\Delta t)/\sum G_{\rm D}(\Delta t)$, we construct a model Y for the *observed* $G_{\rm FWM}(\Delta t)$ in Fig. 3,

$$Y(\Delta t) \;=\; A\, y(\Delta t-\Delta t_0;\alpha,\tau) * g_{\rm D}(\Delta t) + Y_0 \qquad (10)$$

with amplitude A, decay time τ, accidental coincidence background Y_0, time delay Δt_0, and characteristic time α for the jump. We then use A, Y_0, Δt_0, and α as free parameters to fit $Y(\Delta t)$ to our measured time difference distribution. This fit, shown as continuous line in Fig. 3, results in $\alpha = 4.7 \pm 2.5\,$ps corresponding to a 10%–90% rise time associated with the jump of $21 \pm 11\,$ps [30].

Conclusion – We establish a bound for the duration of a quantum jump based on the observed onset of time correlations between photons emitted from an atomic cascade decay in a cold cloud of ^{87}Rb. We find a value that is about three orders of magnitude shorter than the natural lifetime of the involved atomic states, and four orders of magnitude longer than an optical cycle.

In comparison with other techniques [5–12], there seems to be no fundamental limit to the time resolution of this method down to the time scale of the photoelectric effect. We believe our measurement is still limited by the uncertainty in the time response of the avalanche photodetectors, and potentially far from the timescale of quantum jumps – should there be one. Adoption of faster and better characterized detectors, for example from a recent generation of superconducting nanowires [31], has the potential to significantly improve the time resolution of such an experiment, and possibly establish or abandon a resolvable time scale for quantum jumps.

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[1] N. Bohr, The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science **26**, 1 (1913).
[2] E. Schrödinger, Br J Philos Sci **III**, 233 (1952).
[3] J. Bell, "Are there quantum jumps?" in *Schrödinger: Centenary Celebration of a Polymath*, edited by C. W. Kilmister (Cambridge University Press, 1987) pp. 41–52.
[4] H. G. Dehmelt, Bull. Am. Phys. Soc. **20**, 60 (1975).
[5] W. Nagourney, J. Sandberg, and H. Dehmelt, Phys. Rev. Lett. **56**, 2797 (1986).
[6] T. Sauter, W. Neuhauser, R. Blatt, and P. E. Toschek, Phys. Rev. Lett. **57**, 1696 (1986).
[7] J. C. Bergquist, R. G. Hulet, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **57**, 1699 (1986).
[8] M. A. Finn, G. W. Greenlees, and D. A. Lewis, Opt. Commun. **60**, 149 (1986).
[9] P. Neumann, J. Beck, M. Steiner, F. Rempp, H. Fedder, P. R. Hemmer, J. Wrachtrup, and F. Jelezko, Science **329**, 542 (2010).
[10] S. Gleyzes, S. Kuhr, C. Guerlin, J. Bernu, S. Deléglise, U. B. Hoff, M. Brune, J.-M. Raimond, and S. Haroche, Nature **446**, 297 (2007).
[11] R. Vijay, D. H. Slichter, and I. Siddiqi, Phys. Rev. Lett. **106**, 110502 (2011).
[12] Z. K. Minev, S. O. Mundhada, S. Shankar, P. Reinhold, R. Gutierrez-Jauregui, R. J. Schoelkopf, M. Mirrahimi, H. J. Carmichael, and M. H. Devoret, arXiv (2018), 1803.00545 .
[13] R. J. Cook and H. J. Kimble, Phys. Rev. Lett. **54**, 1023 (1985).
[14] C. Cohen-Tannoudji and J. Dalibard, EPL **1**, 441 (1986).
[15] R. J. Glauber, Phys. Rev. **130**, 2529 (1963).
[16] J. Javanainen, Phys. Scr. **1986**, 67 (1986).
[17] H. Carmichael, *An open systems approach to quantum optics*, Lecture Notes in Physics Monographs (Springer, Berlin, 1991).
[18] H. M. Wiseman and G. J. Milburn, Phys. Rev. A **47**, 1652 (1993).
[19] M. Ossiander, F. Siegrist, V. Shirvanyan, R. Pazourek, A. Sommer, T. Latka, A. Guggenmos, S. Nagele, J. Feist, J. Burgdörfer, R. Kienberger, and M. Schultze, Nature Physics **13**, 280 (2016).
[20] M. Ossiander, J. Riemensberger, S. Neppl, M. Mittermair, M. Schäffer, A. Duensing, M. S. Wagner, R. Heider, M. Wurzer, M. Gerl, M. Schnitzenbaumer, J. V. Barth, F. Libisch, C. Lemell, J. Burgdörfer, P. Feulner, and R. Kienberger, Nature **561**, 374 (2018).
[21] B. H. W. Hendriks and G. Nienhuis, Journal of Modern Optics **35**, 1331 (1988).
[22] D. T. Pegg, Physics Letters A **153**, 263 (1991).
[23] M. Porrati and S. Putterman, Phys. Rev. A **36**, 929 (1987).
[24] M. Lax, Phys. Rev. **172**, 350 (1968).
[25] See Supplemental Material [URL will be inserted by publisher] for a detailed analysis of the assumptions in the analytical model.
[26] G. K. Gulati, B. Srivathsan, B. Chng, A. Cerè, and C. Kurtsiefer, New J. Phys. **17**, 093034 (2015).
[27] B. Srivathsan, G. K. Gulati, B. Chng, G. Maslennikov, D. Matsukevich, and C. Kurtsiefer, Phys. Rev. Lett.

111, 123602 (2013).
[28] A. Cerè, B. Srivathsan, G. K. Gulati, B. Chng, and C. Kurtsiefer, Phys. Rev. A **98**, 023835 (2018).
[29] A. Villar, A. Lohrmann, and A. Ling, Opt. Express **26**, 12396 (2018).
[30] A. Cerè, "Quantum jump time estimation from FWM in cold 87Rb," (2018).
[31] V. Shcheslavskiy, P. Morozov, A. Divochiy, Y. Vakhtomin, K. Smirnov, and W. Becker, Rev. Sci. Instrum. **87**, 053117 (2016).