Answer to the Second Referee Report

We thanks the Referee for this insightful review. We answer his/her comments as follows.

I am not at all knowledgeable about optical experiments, so I cannot comment on the novelty or value of this experiment to that field. However, I would like to comment on the conceptual framework that motivated this work, "quantum jumps," especially since the authors cite the paper by John Bell "Are there quantum jumps," whose title he chose to be identical to that of a paper by Schrodinger.

The point made by Schrodinger and Bell is that, if one describes, such an experiment as the authors perform, using standard quantum theory, and includes the apparatus in the state vector description, one ends up with a state vector which is a superposition of the various possible outcomes, including the recording by the apparatus of each outcome. The problem is that one or another outcome actually occurs in the laboratory, and standard quantum theory does not describe how this takes place.

Both Schrodinger and Bell objected to the Copenhagen school's "collapse postulate," which simply states that this transition has to take place sometime, somehow, but is silent on both these issues. Both wished for a transition of the state vector from the superposition to the observed state to be described by theory, not by postulate. In his paper, Bell approvingly wrote about a modification of quantum theory, a new theory which provides such a description, giving a characteristic time for this evolution.

We agree with the Referee that standard quantum mechanics does not provide a satisfactory description of how the transition from the superposition of all possible states to the actual measurement outcome.

The quantum jump notion emerged from the old quantum theory, it was introduced by Bohr to treat the transition from one stationary state to another one. Both the concept and context it is refered to has evolved as the understanding of quantum theory and open systems has increased. Schrödinger emphasized that the idea of jumps appeared to be in sharp conflict with the continuity of wave mechanics. Already in 1930, quantum jumps were reinterpreted as state reduction caused by measurement. Also in that year, Wigner and Weisskopf derived the exponential decay of spontaneous emission from the coupling of the atomic dipole to the continuum of electromagnetic field modes without requiring the hypothesis of quantum jumps. Later, master equation methods were developed for dealing with the irreversible dynamics of such open quantum systems. In this way, standard quantum mechanics was shown to successfully describe the electromagnetic field-atom dynamics providing the probabilities for the different outcomes of a given process.

However, quantum jump models for atoms were never entirely forgotten and the electron shelving experiments in the eigthies refocussed attention on that concept. 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 interrupted by the atom being temporarily shelved in a metastable level. Most attention in 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.

A link between continuous quantum measurement theory and stochastic quantum evolution for the pure state of the system plus an environment was considered by many authors. Carmichael showed that quantum jumps are ubiquotuous to standard photodetection theory. Dalibard, Castin, and Mølmer designed Monte-Carlo wavefunction simulations as an efficient method for numerically work out moderately large quantum systems that originally involved N-level material systems and a quantized electromagnetic field. A key point is that such a successful approach for photodetection assume the presence of stochastic quantum processes that include quantum jumps.

The theory that Bell had in mind in the manuscript “Are there quantum jumps?” differs from the quantum trajectory formalisms mentioned in the last paragraph. Ghirardi, Rimini, and Weber introduced a nonlinear and stochastic modification of Schrödinger’s equation to dynamically implement a continuous dynamical reduction of the state vector onto mutually orthogonal subspaces.

All these works confirm that the ’quantum jump’ concept is a relevant one, that deserves to be studied. We believe that experimental bounds on the timescale involved in the transitions from the superposition of all possible states to the actual measurement outcome in different physical scenarios, are a fundamental step to improve our understanding of the limits of quantum mechanics, and how to modify or extend it.

The point is that the time involved in such a quantum jump is not what the authors conceive it to be. It is the time it takes the superposition of such macroscopic states to evolve to one of those observed states.

What the authors have experimentally done is measure the time between reception of a “signal” photon emitted by an electron dropping to a state ($|3\rangle$ in their notation) and the subsequent emission of an “idler” photon as the electron drops to the ground state ($|0\rangle$ in their notation). Their theoretical analysis concerns only the microscopic system, does not include the apparatus, so this has nothing to do with the notion of quantum jumps discussed above. Their measurement is not the time it takes the superposition of possible outcomes to become one outcome.

We do not agree with the referee about a misconception of the time involved in the quantum jump. We neither claim to have found a way to perform a direct measurement of the transition time of the state vector from all possible states to the measured state. What we have done is to identify and to implement a particular experimental scheme where an upper bound for such a time can be measured regardless the specific dynamics of the transition itself.

I would add that, while the authors concentrate their interest upon their data relative to the shortest time the electron stays in state $|3\rangle$, the measurement of any time in the range of times given by the decay curve they present involves a “quantum jump,” a transition from the superposition of possibilities to the actuality.

The scheme we have chosen to study quantum jumps shares similarities with that used in the seminal works of Dehmelt, Toschek and Wineland. In it, the information of the time behavior of quantum jumps is obtained indirectly via time correlation of detection events. In our scheme, the photons emerge from an atomic cascade decay having a well-defined time order: the first photon of the cascade (signal) is generated before the photon resonant with the ground state (idler). In close analogy with the shelving experiment, each set-up involves different time scales

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

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

We are interested in an upper bound to the time scales (i) and focus 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 manuscript, 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. Notice that Eq.(3) reflects that before time t the atom -field can be considered to be in the 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 and an atomic state $|3\rangle$ with a single photon in one of these modes.

"Now, concerning their analysis, they arrive at the conclusion that standard quantum theory predicts a step-function jump in the time between reception of the signal photon and the earliest idler photon. Again, I have no expertise at all in this area, but I would be surprised if a more careful analysis did not moderate that step function a certain amount.

The author's calculation involves a certain amount of idealization, does not deal with the spatial nature of the atomic state within the Rb atom. I would think that the state vector describing the population of state $|2\rangle$, its depopulation with concurrent photon emission, the population of state $|3\rangle$ and then its depopulation with concurrent photon emission, would end up giving a superposition which would not contain a step function jump between signal photon emission and idler photon emission. However, it might be an alteration that is so small as to be experimentally undetectable. "

The spontaneous character the transitions of interest is manifest in the usage of a density matrix satisfying a Lindblad like equation, which in fact gives rise to the connection between Eqs. (1) and (2) of the manuscript. Adopting the Heisenberg picture the equations of motion of the field operators can be expanded as a sum of free-field operators $\mathbf{E}_f^{(+)}$ (the field of the electromagnetic environment without coupling to the system) and $\mathbf{E}_s^{(+)}$ the source-field operators accounting for the radiated field by the atom. Under the Born-Markov approximation the source operator at a time t can be shown to be proportional to the transition operator $\sigma_{nm}(t - r/c)$ for frequencies close to the atomic transition and average to zero otherwise. For fields initially in the vacuum state, the time- and normal-ordered correlation function given in Eq. (1) can be written in terms of source terms only. Thus establishing the connection between Eq. (1) and (2) in the manuscript, and, in so doing, a first relation to the photodetectors.

Equation (3) allows a direct interpretation of the cascade transitions in terms of quantum jumps. The correlation function involves two density matrices that satisfy the same Lindblad equation with different boundary conditions: the first photon is emitted with a probability proportional to $\rho_{22}(t_s)$, but the correlated observation of the photon pair requires a density matrix ρ' which at the same time t must satisfy the condition $\rho'(t)_{33} = 1$. The idler photon is emitted at a later time t_s with a probability proportional to $\rho_{33}(t_s + \tau)$.

The detailed atomic structure of Rb atom is sintetized in the electric dipole moments for both stimulated and spontaneous transitions according to standard quantum mechanics applied to atom-electromagnetic field systems. This atomic property links the finite sized atom to the quantum electromagnetic field. In the reported model and in order to simplify the description, stimulated transitions we modeled using the rotating wave approximation and, taking into account the lasers detunings, the first level $|1\rangle$ was adiabatically eliminated. These approximations do not alter the structural condition that gives rise to the presence of the Heaviside function wich is the time boundary conditions on ρ and ρ'. We must mention that this has been corroborated by numerical calculations that, in fact, involve not just the four levels here mentioned, but all the levels involved in their hyperfine structure.

Regardless, if the apparatus is included in the state vector, there certainly would be, as the authors suggest in their concluding remarks, a moderation of this step function behavior due to the behavior of the photodetectors. Indeed, that seems to be what they are measuring.

An experimental characterization of the actual correlation function requires a proper incorporation of the photodetector response time to the arrival of photons. As a consequence, in the reported experiment a careful study of the detector response was performed. The results are summarized in Figure 3. To "substract" the detector effects on the measurement of the correlation function $C(t, t+\Delta t)$, a deconvolution of the response function on the direct experimental measurements was carried out.

We thank the Referee for acknowledging that we include the measurement time characteristics in our analysis. We would like to stress that we explicitly clarified that this measurement sets an upper bound limited by the measurement apparatus technical limitation. As opposed to previous approaches, this is a technical and not fundamental limitation, offering the prospective of tighter upper bounds as detection technology improves.

In conclusion, I cannot speak to the value for the photonic community of this experimental upper limit on the briefest correlation time between the signal and idler photons, effectively an upper limit on the amount of time an electron spends in an excited state before dropping to its ground state. It may be that this is a valuable result. However, I object to this being called an experiment that measures the time duration of a quantum jump, since my understanding of the meaning of the time duration of a quantum jump is the time it takes the superposition of possible outcomes (what Sch ödinger called in his 1935 paper the "catalog of expectations") given by Schrödinger's equation to become one, actual, outcome.

There are two key points for a proper interpretation of the results reported in the manuscript:

(i) The close link between the spontaneous emission of a photon and a given atomic transition (addressed above).

(ii) Causality effects in cascade transitions.

In the experiment under consideration

(i) The photodetector used in the experiment allows a characterization—up to the detector capabilities—of the correlation function $C(t, t + \Delta t)$.

(ii) The regression theorem allows for $P(t, t + \Delta t)$ to be written in terms of population matrix elements ρ_{22} and ρ'_{33} with strict and in general different time-boundary conditions, Eq. (3).

This equation reflects the idea that the emission of a signal photon at time t signals the occupation of state $|3\rangle$ at the same time, taken into consideration by $\rho'_{33}(t) = 1$. The probability for this event to occur is proportional to the occupation of $\rho_{22}(t)$ at the time t. In terms of the referee's comment the density matrix ρ' must be chosen taking *the superposition of possible outcomes given by Schrödinger's equation to become one, actual, outcome* at time t. This boundary condition is fundamental in the presence of the $\Theta(t)$ function in $P(t, t + \Delta t)$.

The Referee sees our measurements as the minimum time the electron stays in level $|3\rangle$ before decaying. Even under this interpretation, setting an upper bound on the shortest time the electron stays in state $|3\rangle$ is indeed a way to explore the contrast between a continuous dynamical description expected by Schrödinger and Bell, and the discontinuity predicted by a now standard quantum optics description.

Summarizing, the time correlation between signal and idler photons is composed of two parts. The exponential decay, predicted by standard quantum mechanics, is an accurate characterization of the amount of time an electron spends in an excited state before dropping to its ground state. The focus of our work is instead the sharp rise of the correlation function. This rise is the result of the transition from the atom being in any possible state to a single outcome, i.e., the state $|3\rangle$. We expect the dynamics of this transition to be faster than out detection apparatus.

In order to clarify the relevance of Eq. (3), a supplemental material has been added where the common master equations for ρ and ρ' are given and their differences derived from the time boundary conditions are emphasized.