

Dear Editor,

first of all, let me thank you and the referees for the positive and constructive comments. Our replies to the specific points raised are as follows:

Reviewer 1:

1.

Regarding the experiment - it is indeed unfortunate that it explores only photons with BW larger than the natural BW of Rb, exploring only one side of the curve  $BW > \Gamma_0$ . Additionally, in these experimental parameters, as the authors indeed mention, there is no difference between a coherent state with mean  $n=1$  and their single photon source because of small spatial overlap, and it is certainly not clear, even if there was a measurable difference, why not just use a weak coherent state ( $n \ll 1$ ) and post select on a 'click' (which will project the state to mostly 1 with negligible probability for 2 photons or more).

Reply:

Our motivation to use Fock state single photons is twofold:

1a. Light-atom interfaces are developed for quantum networks. Quantum networks operate with single photons, hence they should be tested with single photons.

1b. Light-atom interaction is EXPECTED to be different for coherent and Fock state excitation. It is correct that for our current parameters we did not expect this effect to be significant and a corresponding experiment using coherent pulses with mean photon number of one is likely to give similar results.

The reviewer's suggestion of using a weak coherent beam in conjunction with a postselection scheme to project the light field into a Fock  $N=1$  state is not suited for our situation of scattering light by a two level system. In detail, post-selection is based on the existence of a second observable which measurement result is considered depending on the outcome of the post-selection observable. Such a second observable could be a second photon or the internal state of the atom. However, when scattering single photons or a weak coherent pulse on a two-level system there is no second observable as there is at most one photon in the system. The detected photon is the observable of interest and cannot be used to project the light into a Fock state.

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2.

Regarding the motivation - the temporal/BW aspect is indeed very important for single photon  $\leftrightarrow$  single quantum emitter interactions, yet two level systems are not enough for quantum information processing, and storage/processing is done with two ground states of 3-level systems or more. Accordingly, deterministic coupling with such systems does not involve just a single transition, thereby avoiding this BW issue.

Reply: We agree that two level systems are not enough for quantum information processing. However, a two level system interacting with a single photon is the most fundamental case of

light-matter interaction and a thorough understanding is a requisite for more complicated interaction schemes. The interaction dependence on the photon BW is also highly relevant in 3-level systems and has been theoretically studied by Pinotsi and Imamoglu in [19].

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Reviewer 2:

1.

Between Eqs. (1) and (2) the authors introduce the ‘scattering probability  $\epsilon$ ’. They should define the term ‘scattering probability’ more clearly as the probability to scatter a photon out of the mode of the incident light and highlight its relation to the extinction of the incident light. Otherwise, readers not working in the same field might have problems to understand the meaning of this parameter. For the same reasons: The overlap parameter  $\Lambda$  in Eq. (2) combines the mode structure of the focused light as well as the solid angle used for focusing [12,13]. Since the authors detect the transmitted photons via focusing onto a single mode fiber, and I assume the used lens and fiber are of the same type as used for guiding the signal photons to the single atom setup, the authors project the detected photons onto the same mode as incident onto the atom. This circumstance should be highlighted already here and not only later when describing the setup, because without projection onto the incident mode, e.g. by focusing onto a detector directly, the  $(1-\Lambda)$  term in Eq. (2) would have to be modified.

Reply: We refined our definition of the term ‘scattering probability’ and the relationship to the observed extinction by adding the following sentences to Section 2:

"Here, the scattering probability  $\epsilon$  is the probability to scatter the incident photon into a spatial mode different from the excitation mode. In the absorption experiment presented in Sec. 3, we determine  $\epsilon$  by detecting the photons in the excitation mode after passing the atom. The scattering probability  $\epsilon$  is then equal to the extinction, that is the reduction of detected photons due to the interaction with the atom."

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2.

page 3 bottom, beginning of experimental section: I fully appreciate the balanced and comprehensive way the authors relate to the work of others. However, to my best knowledge Refs. [31] and [32] of the manuscript do not report on a measurement of the transient excitation of an atom. I suggest to cite only Ref. [13,33] here.

Reply:

We agree and remove Refs. [31] and [32].

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3.

on page 4, bottom: The authors should clarify the mean photon number of the coherent state pulse used in the experiments.

Reply: We added a statement giving the number of photons in the pulse:

"Our heralded photon source can not efficiently prepare photons with a bandwidth below  $2\Gamma_0$ . Therefore, we simulate narrowband photons with 100 ms long pulses of weak laser light. Each pulse contains on average  $\approx 1000$  photons, corresponding to an intensity well below saturation."

-----Pre-Production Review-----

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- Done.

With this, we hope to have addressed all the issues pointed out by the referees and editors, and are looking forward for a reply.

With Best Regards on behalf of all authors,

Christian Kurtsiefer