Measuring the time atoms spend in the excited state due to a photon they don’t absorb

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When a resonant photon traverses a sample of absorbing atoms, how much time do atoms spend in the excited state? Does the answer depend on whether the photon is ultimately absorbed or transmitted? In particular, if it is not absorbed, does it cause atoms to spend any time in the excited state, and if so, how much? In an experiment with ultra-cold Rubidium atoms, we simultaneously record whether atoms are excited by incident (“signal”) photons and whether those photons are transmitted. We measure the time spent by atoms in the excited state by using a separate, off-resonant “probe” laser to monitor the index of refraction of the sample – that is, we measure the nonlinear phase shift written by a signal pulse on this probe beam – and use direct detection to isolate the effect of single transmitted photons. For short pulses (10 ns, to be compared to the 26 ns atomic lifetime) and an optically thick medium (peak OD = 4, leading to 60% absorption given our broad bandwidth), we find that the average time atoms spend in the excited state due to one transmitted photon is not zero, but rather (77 ± 16)% of the time the average incident photon causes them to spend in the excited state. We attribute this observation of “excitation without loss” to coherent forward emission, which can arise when the instantaneous Rabi frequency (pulse envelope) picks up a phase flip – this happens naturally when a broadband pulse propagates through an optically thick medium with frequency-dependent absorption [1]. These results unambiguously reveal the complex history of photons as they propagate through an absorbing medium and illustrate the power of utilizing post-selection to experimentally investigate the past behaviour of observed quantum systems.
When a resonant photon passes through a sample of atoms, the photon can either be absorbed (“lost”) or transmitted. “Loss” in fact involves two steps: excitation of an atom by a photon, and subsequent decay of the atom (whether via spontaneous emission to a side mode, non-radiative decay, or other). In free space and in the absence of stimulated emission, re-emission into the forward mode might seem such an unlikely event as to suggest that atomic excitation is *almost always* accompanied by loss. In other words, in the cases where a photon is successfully transmitted through the absorbing medium, should we conclude it was transmitted because it was lucky enough never to excite an atom? On the other hand, one could take the point of view that photons are simply an electromagnetic field which polarizes atoms – irrespective of whether they are ultimately to be transmitted or lost – thus creating a small excitation probability wherever they are present in the medium. In this latter, “egalitarian,” picture, lost and transmitted photons still behave differently, because the former are on average scattered within the first optical decay length, while the latter have the opportunity to polarize the entire sample. Does this mean that transmitted photons cause atoms to cumulatively spend more time in the excited state than do absorbed ones? Questions about the past behaviour of a quantum particle, or about trajectories in quantum mechanics, have a famously controversial history with important implications for the foundations and interpretation of quantum physics [2][12]. Renewed interest in these questions in the context of open quantum systems has also motivated new theoretical approaches [13][15] and experiments [16][18].

To study the degree of excitation produced in a cloud of cold $^{85}$Rb atoms by an incident signal pulse, we use a nonlinear optical effect known as a resonant Kerr nonlinearity, widely studied for the “cross-phase shifts” (XPS) it can generate, with applications for instance to quantum logic gates [19][24]. A continuous-wave, off-resonant, probe beam overlapped with – but counter-propagating with respect to – the signal pulse acquires a phase that depends on the population inversion, due to the different indices of refraction of the ground and excited states. The time-integral of the phase shift relative to its value when no atoms are excited is proportional to the “excitation time,” by which we mean the expectation value of the total time spent cumulatively by all atoms in the excited state: the time-integral of the mean occupation number in the excited state. In principle, exciting a single atom would change the phase of a resonant laser beam focused down to one atomic cross-section by an amount on the order of a radian. In our experiment, the probe is focused to approximately $10^4$ cross-sections, and acquires a phase of a few tens of $\mu$rad per excited atom. This phase shift, which lasts on the order of a 26-ns atomic lifetime, is currently far too small for us to observe on a single shot.

To resolve such a small effect, one could imagine carrying out this measurement on a sequence of single incident photons, binning the measurements of excitation time according to whether the photon was transmitted or lost, and averaging [25]. Although there exist sources of single, narrow-band photons [26][30], they are currently limited to kHz rates. Instead, we took advantage of the faster rates (approximately 2 MHz) that can be achieved using pulsed coherent states by relying on a novel quantum effect discovered by our group several years ago [22]. In this effect, the mean number of photons one should infer were present in an interaction region illuminated by a coherent state $|\alpha\rangle$ increases by one (aside from minor corrections due to saturation and dark counts) whenever a photon is detected. (In the limit of small incident mean photon number $|\alpha|^2 \ll 1$, it is clear that each detection event revises one’s estimate to at least 1; more generally, when a coherent state is split into two modes – e.g., the “detected” and “undetected” modes for an inefficient photon-counter – their states remain separable, meaning that a measurement collapses the “detected” mode to have 0 or 1 photons while leaving the mean number in the “undetected” mode unchanged.) We first employed this effect to isolate the cross-phase shift due to a post-selected single photon [22], and shortly after, demonstrated Weak Value Amplification of the cross-phase shift due to a post-selected single photon [31]. In the current experiment, when a photon is detected after the medium, the inferred mean number of transmitted photons is increased from $|\alpha|^2 \to |\alpha|^2 + 1$, where $|f|^2$ is the probability for a photon to be transmitted and $|\alpha|^2$ is the mean number of resonant photons entering the medium. By binning our measurements of excitation time in terms of whether a photon is detected or not, we can isolate the effect of a transmitted
have the potential to reduce, swamp, or even masquerade as the effect of a single transmitted photon.

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Detection events are digitized in parallel with the phase, and associated (re-aligned) with the correct

600

were

34

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1%

20

background counts are suppressed to about

70%

700

s, the probe is left on continuously while signal pulses pass through the medium. Signal pulses have

gaussian temporal profile with an rms width of 10 ns and are separated by 576 ns (such that the 864 µs

acquisition window contains 1500 pulses). Each 576 ns window is referred to as a “shot,” and contains 36

phase measurements taken at 16 ns intervals, enabling us to separate the phase shift due to the signal pulse

from slower variations in the index of refraction.

The average phase acquired by the probe within a shot (φ(t)) in the presence of a 34-photon signal

detected with a single-photon counting module (SPCM) (see Fig. 1h) with a detector efficiency of about

70%. The total path efficiency is about 20%, but to reduce spurious counts from probe light leaking into

the fibre, the signal collection efficiency is further reduced using a variable neutral-density filter so that

background counts are suppressed to about 1% per shot (about 20,000 cps). The number of input signal

photons is adjusted to maintain signal detection probability between 20 – 30% (typical photon numbers

were 34 and 134). In order to prevent electrical cross-talk, signal detection is optically delayed by about

600 ns. Detection events are digitized in parallel with the phase, and associated (re-aligned) with the correct

shot during analysis (see Fig. 1d).

We analyze our data by binning all shots into “shots with a click” or “shots without a click” depending

on whether or not the SPCM detected a photon associated with that shot. φ(t) for all the shots “with a click”
is then averaged – this is called φC(t) – and subtracted from the average of all the shots “with no click” –
this is called φNC(t). This difference, Δφ(t) = φC(t) − φNC(t), isolates the effect of a transmitted single

photon on the probe’s phase (which is to say, on the excited-state population). But Δφ(t) is not sensitive

only to the effect of a transmitted photon. Any correlation between the phase of the probe and the click

probability (i.e., the signal transmission) will show up in Δφ(t) – even small spurious correlations, which

have the potential to reduce, swamp, or even masquerade as the effect of a single transmitted photon.

Spurious correlations can arise from two sorts of effects: (1) fluctuations which affect both the linear
phase shift experienced by the probe and the transmission of the signal, and which occur on short enough timescales that our background subtraction fails to eliminate them; and (2) fluctuations which affect the magnitude of the nonlinear cross-phase shift and the transmitted signal power (whether they are fast or slow).

Fluctuations of the first kind can have a huge effect, even if they are quite small, because the linear phase picked up by the probe passing through the atom sample is on the order of a radian – 1000 times larger than the XPS imparted by a signal pulse containing approximately 100 photons. In our experiment, we observed features in \( \Delta \phi(t) \), which we attributed to correlations of this kind because of their particular dependence on signal detuning (anti-symmetric around resonance) and probe detuning (symmetric around resonance). We additionally observed that the time dependence of these features differed from that of the nonlinear cross-phase shift, displaying damped oscillatory behavior with a period of approximately 500 ns and an amplitude that diminished with decreasing pulse length. (If there is a background fluctuation on a timescale longer...
of averaging together roughly 6 billion shots (and as many signal pulses), requiring a total of 900 photons, but were taken with a positive probe detuning (and therefore a positive $\phi$ function (gold line), plus a peaked function whose amplitude is left as a free parameter $\phi$). The residual effects of this kind of spurious correlation appear as a slowly varying background in $\Delta \phi(t)$ (mrad) compared to, and distinguishable from, the sharply-peaked cross-phase shift due to a transmitted photon. In out the experiment with short pulses so that any effects on $\alpha$ than the pulse length, no correlations build up between transmission and linear phase.) We therefore carried out the experiment with short pulses so that any effects on $\Delta \phi(t)$ due to spurious correlations which remain due to the signal not being perfectly on resonance (where their effect disappears) would be both suppressed compared to, and distinguishable from, the sharply-peaked cross-phase shift due to a transmitted photon. In Fig. 3-f, the residual effects of this kind of spurious correlation appear as a slowly varying background in $\Delta \phi(t)$. We reject this slowly-varying background by fitting all 36 phase difference measurements to a cubic function (gold line), plus a peaked function whose amplitude is left as a free parameter $\phi_T$ but whose shape is constrained to match the shape of $\phi(t)$ (gold shading). Figures 2c and 2e show $\Delta \phi(t)$ for signal pulses with $|\alpha|^2 = 34$ and $|\alpha|^2 = 134$ photons respectively, both of which were carried out with a negative probe detuning (and therefore a negative $\phi_0$). Fig. 2d and 2f also depict $\Delta \phi(t)$ taken with $|\alpha|^2 = 34$ and $|\alpha|^2 = 134$ photons, but were taken with a positive probe detuning (and therefore a positive $\phi_0$). Each plot is the result of averaging together roughly 6 billion shots (and as many signal pulses), requiring a total of 900 billion phase measurements and over one hundred hours of data acquisition.

Fluctuations of the second kind can contribute even if they are very slow, as they lead to the XPS being correlated with detection probability, and are therefore not eliminated by our background-subtraction
FIG. 3. (a) A weighted average of $\Delta \phi(t)$ with 34 and 134 photons, and with positive and negative probe detunings, in units of $\phi_0$. The gold line is the shape of the cross-phase shift taken from the fits to $\phi(t)$, and is meant to guide the eye. (b) The phase shift due to a transmitted photon ($\phi_T$) for $-5.6$ MHz and $+4.7$ MHz is shown. In the shaded region, the null results of checks for systematic effects are shown: $\phi_T$ with the probe detuned far from resonance, with the MOT turned off, and with the signal detuned far from resonance (with the probe detuning set to be above (+) and below (-) resonance). (c) Our best estimate of the relative phase shift due to a transmitted photon ($\phi_T/\phi_0$), found by combining measurements from (b) at negative and positive detunings.

procedure. Two such mechanisms include (1) signal intensity fluctuations beyond those inherent to a coherent state, and (2) variations in atom number (OD). If some signal pulses are brighter than others, they will naturally lead to higher detector click rates and to larger nonlinear phase shifts. Since this is an increase in the phase shift written by all of the $|\alpha|^2$ photons in the pulse, its effect can easily exceed the signal of interest due to an individual transmitted photon. Similarly, if the number of atoms fluctuates, periods of high optical depth will be correlated with both low signal transmission and high nonlinear phase shifts. Such correlations, unlike those of the first kind, cannot be distinguished from the effect of a transmitted photon through their dependence on detuning, or their time dependence, both of which are identical to that of the XPS. Instead, the degree of noise must be independently characterized. During the experiment, we continuously monitored the signal optical depth and observed optical depth fluctuations of less than 5%. To assess the total effect of proportional noise in our experiment, we measure $\Delta \phi(t)$ for signal pulses with 588, 898, 1527 and 3040 photons, and from these measurements estimate the total proportional noise due to the combined effects of OD variation and signal intensity fluctuations beyond that of a coherent state to be less than or equal to about 3%, substantially less than Poisson fluctuations for coherent states with average photon number of 34 or 134.

In Fig. 3a all of the data from Figures 2 c-f are combined and normalized to the relevant $\phi_0$ for each curve. Below, Fig. 3b shows the peak amplitude $\phi_T$, after correction for the effects of proportional noise, for both the positive- and negative-detuning cases. The shaded region shows the null results of checks for systematic effects which might masquerade as that of a single photon: data taken with the probe detuned far
from resonance; with the MOT turned off; and with the signal detuned far from resonance (with the probe detuned to the red (−) and to the blue (+)). In Fig. 3, measurements at positive and negative detunings are combined to produce our best estimate of the XPS due to a transmitted photon as a fraction of the XPS due to an average incident photon: we find $\phi_T/\phi_0 = 0.77 \pm 0.16$. The unequivocal implication of this result is that even photons which are ultimately transmitted spend a significant portion of time as atomic excitations.

As unexpected as “excitation without loss” might be, even less intuitive is the implication that lost photons are scattered long before the atom’s spontaneous lifetime has elapsed. This conclusion follows directly from the fact that measurements of $\phi_0$ and $\phi_T$ are reflective of the time atoms spend in the excited state ($\tau_0$ and $\tau_T$) and the fact that spontaneous emission occurs at a constant rate $\Gamma = 1/\tau_{sp}$, making a photon’s probability of being lost proportional to the total time it causes atoms to spend in the excited state: $P_L = \Gamma \tau_0 = \tau_0/\tau_{sp}$. In other words, the total time spent in the excited state due to an average incident photon is equal to the product of the loss probability and the atomic lifetime $\tau_{sp}$. But of course $\tau_0$ must also be equal to the average of the excitation times for lost and transmitted photons, $\tau_L$ and $\tau_T$, weighted by the loss and transmission probabilities. Thus, any non-zero (and positive) $\tau_T$ immediately implies that $\tau_L < \tau_{sp}$, which can only occur if atoms spend on average less than one atomic lifetime in the excited state before spontaneously emitting.

For a broadband pulse passing through an optically thick medium, $\tau_L < \tau_{sp}$ proves to be easy to understand: when such a pulse propagates through an absorbing medium, the envelope acquires a phase flip which causes the pulse area – the angle through which this pulse will rotate an atomic Bloch vector – to decay quickly to zero, even in regions where the pulse energy is not decaying rapidly. (This can be understood through the pulse area theorem [32, 33]; or in terms of temporal beats created once the central part of the spectrum is absorbed; and/or as atomic emission which persists long after the passage of the initial pulse, but is 180° out of phase with it [1].) Regardless of the perspective one takes, the consequence for the atoms is that portions of the pulse – even in the linear, or single-photon, regime [35] – coherently drive excited atoms back to the ground state. We believe that this coherent forward emission process underlies both the nonlinear phase shift associated with signal photons that are not lost, and the concomitant reduction of the mean time spent in the excited state below $\tau_{sp}$ (which drives the nonlinear effect of lost photons below what would be naively expected). While such semiclassical arguments support the notion that coherent emission must be present, they are incapable of quantifying precisely how much of it occurs, and provide only a lower bound. Moreover, they are of course silent on questions of entanglement between the final state of the signal photon and the time spent by an atom in the excited state.

In Figure 4, we compare our experimental result (purple) with several simple theoretical models: in blue [solid] for narrowband pulses (orange [dotted] for broadband pulses), the egalitarian model introduced earlier, in which absorbed photons affect the medium only up until the point where they are scattered, whereas transmitted photons interact with the full length of the medium; in green [dot-dash], a model based on the minimum amount of coherent forward emission consistent with the semiclassical Bloch-state evolution for a 10-ns pulse like the one used in our experiment; and in red [dashed], for comparison, the predictions such a minimized-coherent-emission model would make for a narrowband, 50-ns pulse. The data clearly rule out $\phi_T \propto \tau_T = 0$, and are consistent with a range of values which includes both the egalitarian model and the minimized-coherent-emission prediction. These models, however, make strikingly different predictions for $\tau_T$ in the narrow-bandwidth limit; and for $\tau_L$ in the low-OD limit (see inset).

The “egalitarian” model predicts that $\tau_T \not= 0$, and hence that $\tau_L < \tau_{sp}$. Its implications, however, become extreme in the OD → 0 limit. Here $P_L \to OD$, and $\tau_L \to \tau_T/2$ (because the average position for a photon to be scattered is halfway through the medium). As the transmission probability approaches 100%, $\tau_0 \to \tau_T$; and yet we know $\tau_T = P_L \tau_{sp}$. This would imply that $\tau_T/\tau_{sp} \to OD$ and $\tau_L/\tau_{sp} \to OD/2 \to 0$. So this model not only requires a mechanism to pull atoms out of the excited state faster than $\Gamma$, but needs the speed of this mechanism to diverge as 1/OD, which seems impossible to justify on physical grounds. More natural would be the expectation that at least in the low-OD regime, $\tau_L \to \tau_{sp}$, implying $\tau_T \to 0$, as occurs for instance
A comparison of our experimental results to the predictions of various semi-classical models. The predictions of a toy model in which coherent forward emission occurs as a result of an interference effect arising when broadband small-area pulses propagate through an optically thick medium are shown both for signal pulses of 8 MHz bandwidth as in our experiment (green dot-dashed line), and for narrowband (1.5 MHz) ones (red dashed line). An “egalitarian” model in which photons affect the medium up until the point where they are scattered (or transmitted) is shown for the 8 MHz (orange dotted line) and 0.1 MHz (blue solid line) cases. Our experimental results are also shown (purple), with the peak on-resonant OD inferred from the optical depth seen by the probe (about 4), consistent with the amount of attenuation experienced by a resonant pulse with a bandwidth of 8 MHz. In the inset, $\tau_L/\tau_{sp}$ is plotted for all four models, along with the inferred result of our measurement.

in the minimum-coherent-emission model we base on the solutions to the Maxwell-Bloch equations. The latter model has extreme sensitivity not only to bandwidth but also to pulse shape and duration (which is unusual for calculations of the effect of a single photon), but it reproduces the intuition that for low OD or narrowband pulses, $\tau_L \rightarrow \tau_{sp}$.

The experiment described in this work constitutes an important milestone: the first measurement of the time atoms spend in the excited state due to photons they don’t ultimately absorb. We find that this time is nonzero, and can even be on the order of the atomic lifetime. These results cry out for a fully quantized theoretical treatment as well as further experiments varying the optical depth, bandwidth, and pulse shape in order to further elucidate the strange history of transmitted photons.

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