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Measuring the Time Atoms Spend in the Excited State Due to a Photon They Do Not 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? To answer these questions, we perform an experiment with ultracold rubidium atoms in which 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 optical depth equals 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, $(77 \pm 16)\%$ 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 180° phase flip—this happens naturally when a broadband pulse propagates through an optically thick medium with frequency-dependent absorption. These results unambiguously reveal for the first time the complex history of photons as they propagate through an absorbing medium and illustrate the power of utilizing postselection to experimentally investigate the past behavior of observed quantum systems.

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I. INTRODUCTION

When a resonant photon passes through a sample of atoms, the photon can either be absorbed ("lost") or transmitted. "Loss" is often thought of as involving two steps: excitation of an atom by a photon and subsequent decay of the atom (whether via spontaneous emission to a side mode, nonradiative decay, or some other mechanism). In free space and in the absence of stimulated emission, spontaneous emission into the forward mode by an excited atom is usually so rare as to be negligible. In the cases where a photon is successfully transmitted through the absorbing medium, should one conclude that it was transmitted because it was lucky enough to never excite an atom? Do only lost photons cause atoms to spend time in the excited state, perhaps each causing one atom to be excited for on average one spontaneous lifetime (τ_{sp})? Alternatively, one could take the point of view that light is simply an electromagnetic field that polarizes atoms—irrespective of whether a given photon is ultimately transmitted or lost—thus creating a small excitation probability wherever it is present in the medium. In this latter, "egalitarian," picture, lost and transmitted photons both excite atoms but they 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 such as these about the past behavior of a quantum particle, or about trajectories in

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quantum mechanics, have a famously controversial history with important implications for the foundations and interpretation of quantum physics [1–12]. Advances in experimental techniques for preparing and controlling quantum systems have spurred renewed interest in these questions, particularly in the context of weak measurements and continuous measurements of open quantum systems [13–15]. At the same time, new theoretical approaches for describing the dynamics of open-quantum systems are also being rapidly developed [16–18]. However, it turns out that until now, there has been no measurement (or, to our knowledge, calculation) of how much time atoms spend in the excited state due to the passage of a resonant photon that is ultimately transmitted.

II. METHODS

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 Kerr nonlinearity, widely studied for the "cross-phase shifts" (XPSs) it can generate, with applications, for instance, to quantum logic gates [19–24]. A continuous-wave, off-resonant, probe beam overlapped with the signal pulse in an atomic medium acquires a phase that depends on the population inversion, due to the different indices of refraction of the ground and excited states [$\phi(t) = (n_e - n_g)\omega L/c \times \langle P_e(t)\rangle$, where $\langle P_e(t)\rangle$ is the average probability for an atom to be in the excited state]. As the resonant signal pulse propagates through the cloud, it temporarily polarizes the atoms, briefly creating the excited-state population [$\langle P_e(t)\rangle$], thus writing a time-dependent XPS onto the probe beam. The time integral of this phase shift is proportional to the time integral of the average excited-state population induced by the signal pulse, which we refer to as the "excitation time" $\left[\tau_e \equiv \int \langle P_e(t)\rangle dt\right]$. The excitation time τ_e, which is a measure of the degree of atomic excitation induced by the signal pulse, has units of time, represents a duration, and provides an operational definition of the time atoms spend in the excited state (see Fig. 1, also [25]). 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 XPS of a few tens of microradians per excited atom (for a calibration of the XPS per photon, see Sec. II of the Supplemental Material [26]). This XPS, which lasts on the order of a 26-ns atomic lifetime, is currently far too small for us to observe on a single shot. As a result, very little information is gained about the excited-state occupation number on a single shot and the signal-atom interaction is not disturbed. However, by carrying out many such weak measurements and averaging, one can determine as precisely as necessary the average excited-state occupation number induced by the signal (or, in our implementation, the time integral of this quantity).

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 is transmitted or lost, and averaging [27]. Although there exist sources of single narrow-band photons [28–32], they are currently limited to kilohertz rates. Instead, we take 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 an interaction region illuminated by a coherent state $|\alpha\rangle$, the mean number of photons one should infer are present in the region increases by one (aside from minor corrections due to our detectors not being number resolving and dark counts [33]) 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 XPS due to a postselected single photon [22] and, shortly after, demonstrated weak value amplification of the XPS due to a postselected single photon [34]. In the current experiment, when a photon is detected after the medium, the inferred mean number of *transmitted* photons is increased from $|t\alpha|^2 \rightarrow |t\alpha|^2 + 1$, where $|t|^2$ 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 or not a photon is detected, we can isolate the effect of approximately one extra transmitted photon.

III. EXPERIMENT

For the experiment (see Fig. 1), we use an ultracold cloud of ^{85}Rb in a magneto-optical trap (MOT) and a pair of counterpropagating overlapping beams focused to about 25 μm inside the atom cloud. We set the signal on resonance with the $F = 3$ to $F' = 4$ transition of the 780-nm D_2 line and detune the probe by $\pm$ (3–10) MHz from the same transition to monitor the excited-state population [the level scheme used is shown in Fig. 1(a)]. The signal and probe beams are frequency stabilized using Doppler-free polarization spectroscopy (for further discussion of the importance of frequency stability in the experiment, see Sec. III of the Supplemental Material [26]). The beams are spatially overlapped and separated using 90:10 beam splitters. After passing through the MOT, the probe is collected in multimode fiber. We measure the phase using

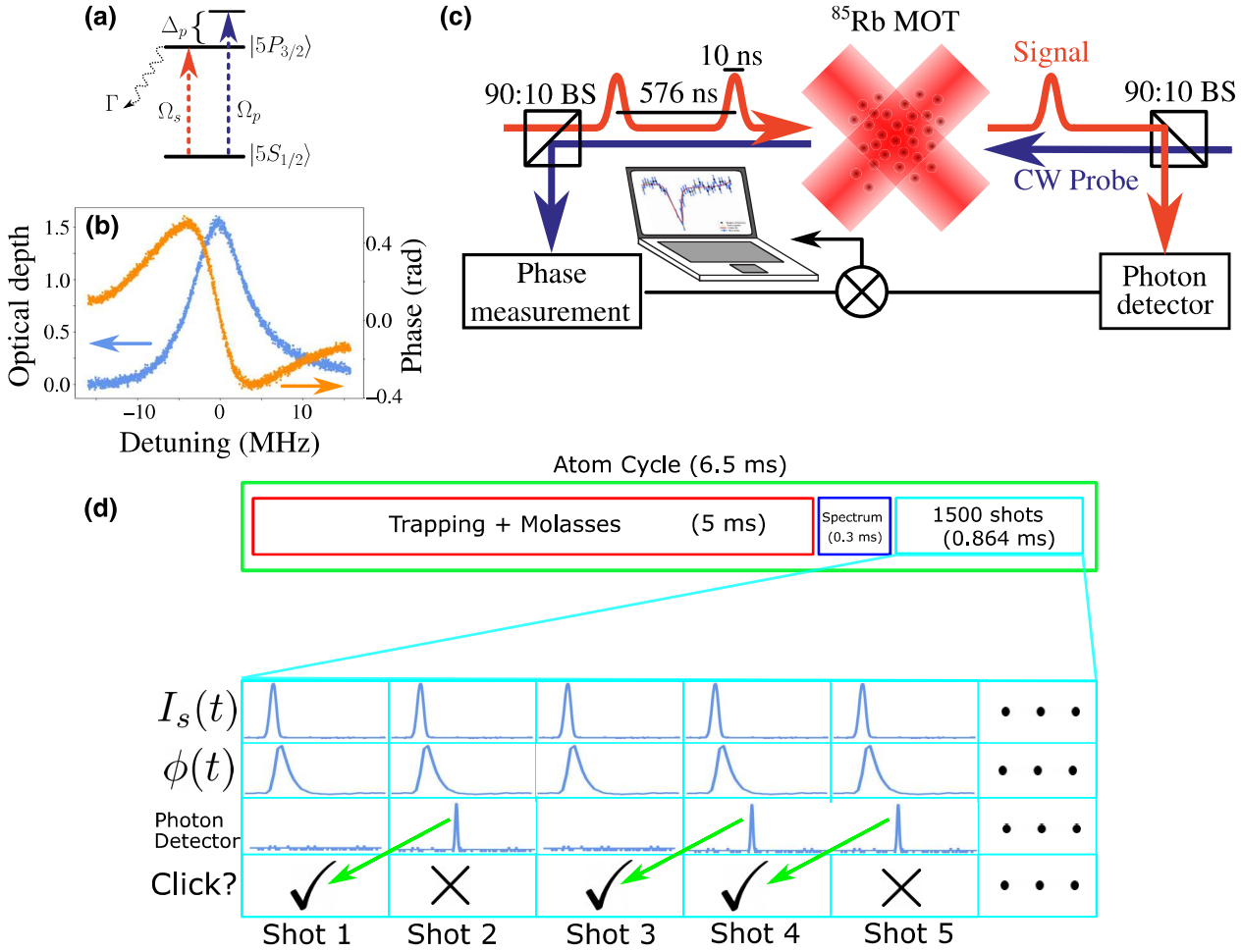

FIG. 1. (a) The level scheme used in the experiment; $\Omega_{p(s)}$ is the probe (signal) Rabi frequency, Δ_p is the probe detuning, and Γ is the spontaneous decay rate of the excited state. (b) The measured optical depth and linear phase experienced by the probe as a function of Δ_p, with the signal turned off. (c) A simplified schematic of the experimental apparatus, consisting of signal and probe beams overlapped using 90:10 beam splitters (BSs), the rubidium MOT, photon detection, and phase measurement. (d) The experimental timing sequence used in the experiment. The green arrows indicate photon detection being reassociated with the correct shot. $I_s(t)$ is the average signal intensity measured with a fast APD. $\phi(t)$ is the measured XPS during a shot. "Click?" indicates whether or not a photon is detected.

frequency-domain interferometry, which involves beating the probe against a co-propagating reference beam, which is shifted off resonance by 100 MHz and hence nearly unaffected by the atoms.

Each cycle of the experimental timing sequence [Fig. 1(d)] consists of approximately 5 ms of MOT trapping, molasses, and free expansion, and a 1.152-ms "measurement" stage. During this last stage, both the magnetic fields and the MOT beams are turned off, but the repumper is left on to avoid forming a dark state in $F = 2$. During the first 288 μs of each measurement stage, the probe phase and transmission are measured while the frequency of the probe (and reference) is scanned across resonance to extract the optical depth (OD) and the linear phase as a function of the detuning [for a representative measurement taken at reduced OD, see Fig. 1(b)]. For the remaining 864 μs, the probe is left on continuously while signal pulses pass through the medium. The signal pulses have a 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 XPS acquired by the probe within a shot [$\phi(t)$] is plotted for a signal pulse containing $|\alpha|^2 \approx 34$ photons in Fig. 2(a) for a red-detuned probe (−5.6 MHz, negative XPS) and in Fig. 2(b) for a blue-detuned probe (+4.7 MHz, positive XPS). The signal photon number is measured using an neutral-density (ND) filter calibrated with a Thorlabs power meter and a Thorlabs avalanche

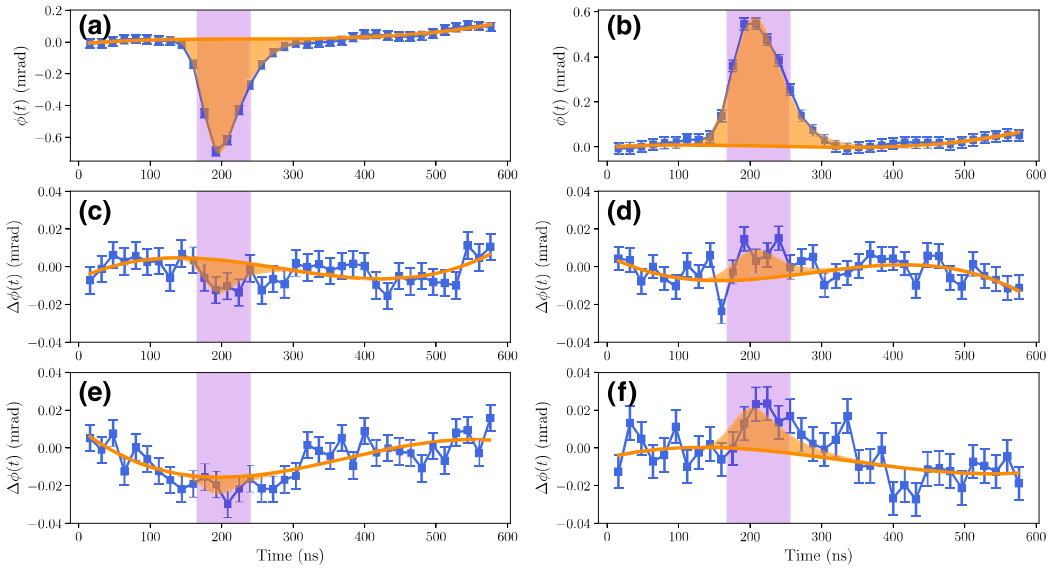

FIG. 2. (a),(b) The probe cross-phase shift and (c)–(f) the probe cross-phase shift differences versus time, averaged over approximately 6 billion shots, in the presence of a 10-ns signal pulse. The probe XPS $[\phi(t)]$ is recorded for pulses containing $|\alpha|^2 \approx 34$ photons incident at approximately $t = 200$ ns for probe detunings of (a) -5.6 MHz and (b) $+4.7$ MHz. The purple rectangle indicates the window of time wherein the XPS occurs. The shaded gold curve is a fit to a cubic polynomial (to model the background drift) plus a peaked function. The solid gold line shows only the cubic fit to background. Plots (c) and (d) show $\Delta\phi(t)$ for pulses containing $|\alpha|^2 \approx 34$ photons and plots (e) and (f) show $\Delta\phi(t)$ for pulses containing $|\alpha|^2 \approx 134$ photons. As before, the probe detuning is negative on the left [(c),(e)] and positive on the right [(d),(f)]. Here, the shaded gold region indicates a fit to a cubic polynomial plus a function with the shape of the peaks fitted to the relevant average XPS [e.g., for (c), the XPS in (a) is used, while the XPS used for (e) and (f) is not shown] but with an adjustable amplitude. As before, the solid gold line shows only the cubic background fit. The phase is sampled every 16 ns and a 25-MHz measurement bandwidth is used. The phase noise on a single sample is approximately 100–200 mrad and is determined by the sampling rate, the measurement bandwidth, and the probe power, which is set to approximately 5 nW in order to be much less than saturation (about 50 nW). The error bars shown are the standard error of the mean.

photodiode in order to take into account the temporal shape of the signal pulses and the reported mean photon numbers are estimated to be accurate within approximately 10% (for information about how the signal photon number is calibrated in the experiment, see Sec. I of the Supplemental Material [26]; for an estimate of faster intensity fluctuations, see Sec. III of the Supplemental Material [26]).

In both Figs. 2(a) and 2(b), the 576-ns trace is averaged approximately 6 billion times and the XPS due to the 34-photon signal pulse is clearly visible, with a peak of ± 600 μrad. The pulse enters at about 175 ns into the shot and $\phi(t)$ grows quickly as the signal pulse enters the medium (with a slight delay due to the 25-MHz bandwidth of our data acquisition), peaks around 200 ns, and decays exponentially over a slightly longer time scale set by the atomic lifetime (26.5 ns) after the signal pulse exits the medium. The peak XPS per photon, ϕ_0, is found by dividing the peak excursion of $\phi(t)$ by the average number of photons in the signal pulse. ϕ_0 is -20.0 ± 0.4 μrad per photon and $+16.4 \pm 0.4$ μrad per photon in Figs. 2(a) and 2(b), respectively.

Once it exits the medium, the transmitted portion of the signal is collected in a single-mode fiber and detected with a single-photon-counting module (SPCM) [see Fig. 1(c)] 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 fiber, the signal collection efficiency is further reduced using a variable ND filter so that background counts are suppressed to about 1% per shot (about 20 000 counts per second). The number of input signal photons is adjusted to maintain the signal-detection probability between 20% and 25% (typical mean photon numbers are $|\alpha|^2 \approx 34$ and 134). In order to prevent electrical crosstalk, the signal detection is optically delayed by about 600 ns. The detection events are digitized in parallel with the phase and associated (realigned) with the correct shot during analysis [see Fig. 1(d)].

IV. ANALYSIS AND RESULTS

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. $\phi(t)$ for all the shots "with a click" is then averaged—this is called $\phi_C(t)$—and subtracted from the average of all the shots "with no click"—this is called $\phi_{NC}(t)$. This difference

in cross-phase shifts, $\Delta\phi(t) = \phi_C(t) - \phi_{NC}(t)$, isolates the effect of a transmitted single photon on the XPS (that is, the effect of a transmitted single photon on the excited-state population). But $\Delta\phi(t)$ 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 $\Delta\phi(t)$. Moreover, because the effect of a single photon is inherently weak, even small spurious correlations produced by a shared or "common" cause have the potential to reduce, swamp, or even masquerade as the effect of a single transmitted photon.

Spurious correlations due to a common cause can arise from two sorts of effects: (1) fluctuations that affect both the *linear* phase experienced by the probe and the transmission of the signal and that occur on short enough time scales that our background subtraction fails to eliminate them; and (2) fluctuations that affect the *magnitude* of the *nonlinear* XPS 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 small, because the linear phase picked up by the probe passing through the atom sample is on the order of a radian [see Fig. 1(b)]—1000 times larger than the XPS imparted by a signal pulse containing approximately 100 photons. For example, if fluctuating background magnetic fields are present, they produce Zeeman shifts that affect both signal and probe detuning resulting in a spurious "common-cause" correlation between signal transmission and the linear phase of the probe that can appear in $\Delta\phi(t)$. In our experiment, we observed features in $\Delta\phi(t)$, which we attribute to correlations of this kind because of their particular dependence on signal detuning (antisymmetric around resonance) and probe detuning (symmetric around resonance). We additionally observed that the time dependence of these features differs from that of the nonlinear XPS, displaying damped oscillatory behavior with a period of approximately 500 ns and an amplitude that diminishes with decreasing pulse length. (If there is a background fluctuation on a time scale longer than the pulse length, no correlations build up between transmission and linear phase.) We therefore carry out the experiment with short pulses so that any effects on $\Delta\phi(t)$ due to spurious correlations that remain due to the signal not being perfectly on resonance (where their effect disappears) will be both suppressed compared to, and distinguishable from, the sharply peaked XPS due to a transmitted photon. In Figs. 2(c)–2(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 the amplitude of which is left as a free parameter ϕ_T but the shape of which is constrained to match the shape of $\phi(t)$ (gold shading) (see Eq. 5 in Sec. III of the Supplemental Material [26]). Figures 2(c) and 2(e) show $\Delta\phi(t)$ for signal pulses with $|\alpha|^2 \approx 34$ and $|\alpha|^2 \approx 134$ photons, respectively, both of which are carried out with a negative probe detuning (and therefore a negative ϕ_0). Figures 2(d) and 2(f) also depict $\Delta\phi(t)$ taken with $|\alpha|^2 \approx 34$ and $|\alpha|^2 \approx 134$ photons but are taken with a positive probe detuning (and therefore a positive ϕ_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 100 h 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 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. However, such fluctuations produce a correlation in $\Delta\phi(t)$ that is proportional to the average number of photons and can therefore be separately calibrated by varying the average photon number. To assess the total effect of all types of proportional noise in our experiment, we measure $\Delta\phi(t)$ for signal pulses with approximately 588, 898, 1527, and 3040 photons. From these measurements, we can separate out the XPS due to a single transmitted photon from the effects of proportional noise (see Section III of the Supplemental Material [26]).

In Fig. 3(a), all of the data from Figs. 2(c)–2(f) are combined and normalized to the relevant ϕ_0 for each curve. Below, Fig. 3(b) shows the peak amplitude ϕ_T, after correction for the effects of proportional noise, for both the positive- and negative-detuning cases (see Sec. III of the Supplemental Material [26]). The shaded region shows the null results of checks for systematic effects that 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 (+)] (for further explanation of how checks for systematic effects are performed, see Sec. V of the Supplemental Material [26]). In Fig. 3(c), 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:

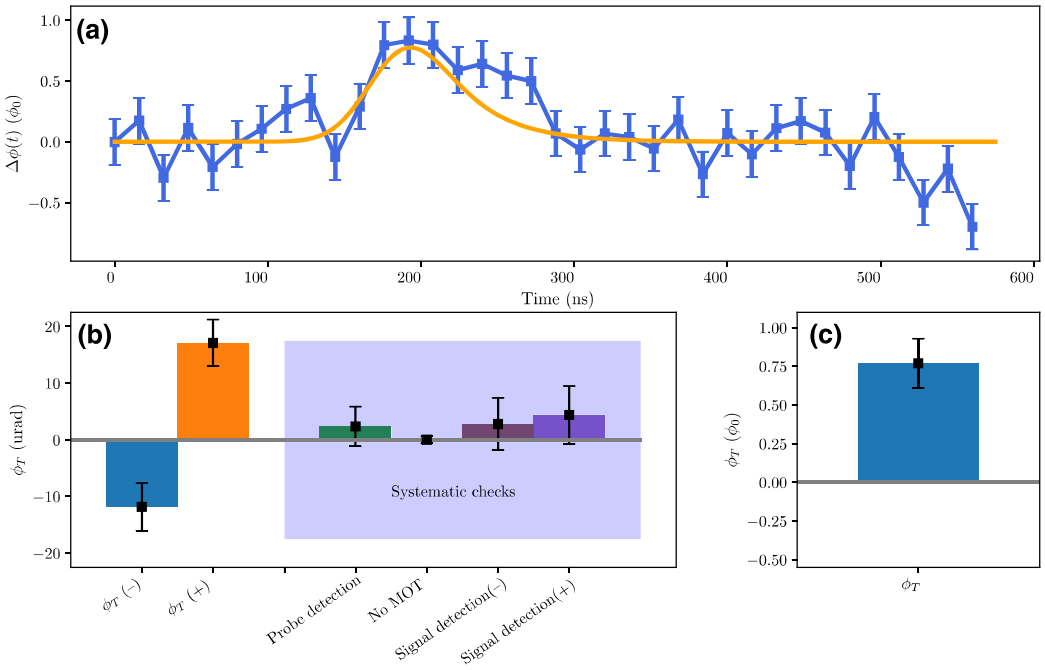

FIG. 3. (a) A weighted average of $\Delta\phi(t)$ with 34 and 134 photons and with positive and negative probe detunings, in units of ϕ_0. The gold line represents our measurement of the effect of a transmitted photon, with the amplitude and shape taken from fits to $\phi(t)$ and the effect of proportional noise subtracted off (see Sec. III of the Supplemental Material [26]). (b) The phase shift due to a transmitted photon (ϕ_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: ϕ_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 (ϕ_T/ϕ_0), found by combining measurements from (b) at negative and positive detunings.

we find $\phi_T/\phi_0 = 0.77 \pm 0.16$. The unequivocal implication of this result is that even photons that are ultimately transmitted spend a significant portion of time as atomic excitations.

V. DISCUSSION

As unexpected as "excitation without loss" might be, even less intuitive is the implication that lost photons are scattered long before the spontaneous lifetime of the atom has elapsed. This conclusion follows directly from the fact that measurements of ϕ_0 and ϕ_T are reflective of the time atoms spend in the excited state (τ_0 and τ_T) and the fact that spontaneous decay occurs at a constant rate $\Gamma = 1/\tau_{\rm sp}$, making the probability that a photon will be lost proportional to the total time it causes atoms to spend in the excited state: $P_L = \Gamma\tau_0 = \tau_0/\tau_{\rm sp}$. In other words, the cumulative time spent in the excited state due to an average incident photon (τ_0) is equal to the product of the loss probability (P_L) and the atomic lifetime $\tau_{\rm sp}$. But, of course, τ_0 must also be equal to the average of the excitation times for lost and transmitted photons, τ_L and τ_T, weighted by the loss and transmission probabilities (i.e., $\tau_0 = \tau_{\rm sp}P_L$ but also $\tau_0 = \tau_L P_L + \tau_T P_T$). Thus, any nonzero (and positive) τ_T immediately implies that $\tau_L < \tau_{\rm sp}$, which can only occur if atoms spend on average less than one atomic lifetime in the excited state before spontaneously decaying.

For a broadband pulse passing through an optically thick medium, $\tau_L < \tau_{\rm sp}$ can be understood in terms coherent deexcitation of atoms. When a broadband pulse propagates through an absorbing medium, the envelope acquires a phase flip that 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 [35–37]; or in terms of temporal beats created once the central part of the spectrum is absorbed; and/or as atomic emission that persists long after the passage of the initial pulse, but is 180° out of phase with it [38]. 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 [39]—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 $\tau_{\rm sp}$ (which drives the nonlinear effect of lost photons below what would be naively expected).

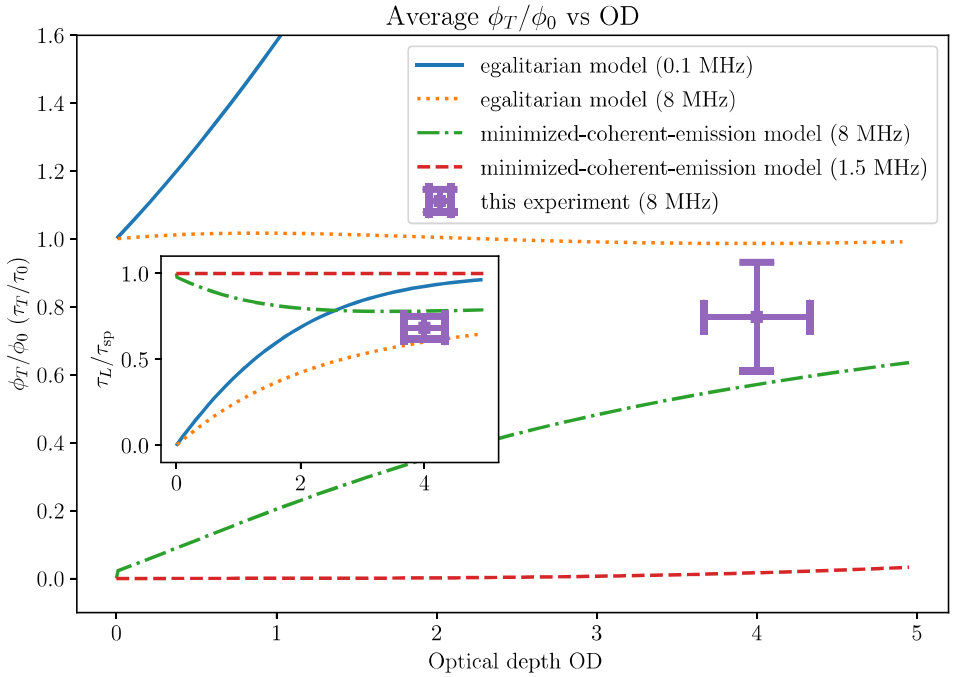

FIG. 4. A comparison of our experimental results with the predictions of various semiclassical models. The predictions of a toy model (see Sec. IV of the Supplemental Material [26]) in which coherent forward emission occurs as a result of an interference effect arising when broadband small-area pulses 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 narrow-band (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, τ_L/τ_{sp} is plotted for all four models, along with the inferred result of our measurement.

In Fig. 4, we compare our experimental result [purple] with several simple theoretical models: in solid blue for narrow-band pulses (dotted orange 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 described in more detail in Sec. IV of the Supplemental Material [26], 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 of such a minimized-coherent-emission model for a narrow-band 50-ns pulse. The data clearly rule out $\phi_T \propto \tau_T = 0$ and are consistent with a range of values that includes both the egalitarian model and the minimized-coherent-emission prediction. These models, however, make strikingly different predictions for τ_T in the narrow-bandwidth limit and for τ_L in the low-OD limit (see the inset).

The "egalitarian" model predicts that $\tau_T \neq 0$ and hence that $\tau_L < \tau_{sp}$. Its implications, however, become extreme in the small-OD limit. Here, $P_L = 1 - e^{-OD} \to$ 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 that $\tau_0 = P_L\tau_{sp}$. This would imply that $\tau_T/\tau_{sp} \to$ OD and $\tau_L/\tau_{sp} \to$ OD/2. So this model not only requires a mechanism to pull atoms out of the excited state faster than Γ but also 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, in the minimum-coherent-emission model, which we base on 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 narrow-band pulses, $\tau_L \to \tau_{sp}$.

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 evolution of the atom. A complete

quantum description of an itinerant pulse of light interacting with an ensemble of two-level atoms is a topic of current research, as the problem requires, in Klaus Mølmer's words, a "vastly complex expansion of the possibly entangled multiphoton quantum state on a large number of modes" [40]. He goes on to say that while formal solutions to this computationally hard many-body problem are typically intractable, numerical methods including matrix product states [41,42] and multiphoton scattering theory in temporal mode bases have shown promising results [43]. An alternative approach, which we are currently pursuing and that will be the subject of a future publication [44], involves the theory of weak and/or continuous measurements to probe the trajectories of interacting open quantum systems [13,16–18,45–47].

VI. CONCLUSION

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 do not 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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Note added.—While this work has been under review, further progress has led to a preliminary theoretical result predicting a value of 0.40 for a peak OD of 4 [44].

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[26] See the Supplemental Material at http://link.aps.org/supplemental/10.1103/PRXQuantum.3.010314 for details regarding the dependence of the per-photon cross-phase shift on pulse length, the size of the cross-phase shift per photon, assessing the total amount of proportional noise in the signal, the minimum-coherent-emission model, systematic effects, and data availability.

[27] This is clearly closely related to the weak measurements of Aharonov, Albert, and Vaidman, as it involves a pre- and postselection and a weak coupling to a probe system; for other examples of pre- and postselected systems including from cavity QED, see Refs. [45,48–50]).

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