Room temperature conditional $\pi$-phase shifts mediated by simultaneously propagating single-photon level pulses

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Here we demonstrate the first room-temperature implementation of $\pi$ phase shifts in single-photon-level probe light created by a simultaneously propagating few-photon triggering signal field. The photon-photon interaction is mediated by rubidium atoms in a double $\Lambda$ atomic scheme. We use homodyne tomography and maximum likelihood estimation on the quadrature statistics of the input and phase-shifted photons to fully characterize their quantum states in the Fock state basis. For particular choices in control fields strengths and input phases, the input-output fidelity of the controlled $\pi$ phase shift operation reaches $\sim 90\%$.

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I. INTRODUCTION

Recent “grid-based” quantum processing architectures composed of many ions, Josephson-junctions, and 3D lattices of Rydberg atoms have been successful in emulating the physics of many-body Hamiltonians 1–7. A challenge of these grid-based systems is the quasi-locality of interactions between qubits, which restricts what types of quantum algorithms can be performed. While one route towards solving this challenge is to rework the theoretical architecture to improve connectivity from local interactions 8, another route is to create experimental architectures capable of nonlocal interactions.

Constructing this non-local processing grid connecting multiple quantum devices in a scalable manner is a challenge. A particularly promising architecture to achieve large quantum processing networks is to interconnect simple gas cells of warm atoms mediating photon-photon interactions. Very recently, these warm atom systems have been shown to be able to operate as quantum memories, storing and retrieving single-photon level polarization qubits with low noise and high fidelity 9. Along these lines, an entangling, photon-photon gate using warm atoms has the potential to simultaneously provide the required “all-to-all” qubit connectivity necessary for fully-programmable quantum computation while also providing an architecture in which room-temperature quantum processing can be explored. In this envisioned grid, unlike fixed locally-interacting systems, all possible pairs of qubits can be made to interact, since the inputs are flying photons that can easily be routed to interact with any gate. Simultaneously, a system of these gates could interact with room-temperature light-matter quantum memories to create a network 10–14, potentially connecting multiple distinct quantum devices, forming a “quantum internet” 15.

Given the recent developments of quantum memory systems 9 10, the remaining technological milestone for this architecture is the creation of atomic systems in which photons are made to interact to form logical operations. This is a considerable challenge because the main strength of flying photons is their typically weak coupling to the environment. One proposal for creating a strong photon-photon interaction is engineering an atomic interaction using Kerr nonlinearities created by EIT 17. However, systems mediated by Kerr nonlinearities have yet to see phase shifts higher than 15 $\mu$ radians per photon 19. Breakthrough experiments have shown that, with special preparation, it is indeed possible to obtain large phase-shifts at the single-photon level using Rydberg interactions, magneto-optical traps, and cavity quantum electrodynamics 18–21. While these experiments have demonstrated large phase-shifts at the low-light level, a quantum analysis of the output light is required to make statements about the fidelity and the process’s usefulness as a gate.

In this experiment we both demonstrate the first room-temperature implementation in which simultaneously propagating pulses of single photon level light create $\pi$ phase shifts, while also performing a quantum characterization of the phase-shifted output state.

II. DOUBLE-LAMBDA ATOMIC SYSTEM

A. Phase-Phase modulation

In this section we describe the phase control mechanism intrinsic to double-$\Lambda$ closed-loop atomic systems. While a fully quantized model of this double-$\Lambda$ system has been constructed 22, a simple semiclassical model is helpful in understanding how the atom-light interaction creates a phase shift. We investigate the four-wave mixing (FWM) generated by a 3-level electromagnetically induced transparency (EIT) system in a double-$\Lambda$ configuration.

The simplest, intuitive model of our system is quantum interference of the coherence terms generated by EIT and Stokes(anti-Stokes) four-wave mixing. The Stokes
process generates coherent light in the signal frequency \((\omega_c - \omega_s + \omega_p = \Delta + \omega_p = \omega_s)\), while the anti-Stokes generates it in the probe frequency \((\omega_c - \omega_s - \omega_s = \Delta - \omega_s = \omega_p)\). The influence of the four-wave-mixing on the overall coherence can be identified by investigating the polarizability created by the atomic coherences generated in the double-Λ system. Since \(\Delta \ll \omega\), we can make an adiabatic approximation for the contribution of the other fields, assuming that their effect is to create slowly-varying amplitudes (on the order of \(\Delta \ll \omega\)). Identifying that our additional time-dependencies are slowly varying relative to the field’s frequency (similar to a standard EIT system) the solutions can be found via the Optical-Bloch equations:

\[
\rho_{13} = \frac{i\Omega_i}{2i\Delta_2 + \Gamma} - \frac{\Omega_i|\Omega_j|^2}{\gamma(2i\Delta_2 + \Gamma)^2} \tag{1}
\]

\[
\rho_{12} = \frac{i\Omega_i\Omega_j^*}{\gamma(2i\Delta_2 + \Gamma)^2} \tag{2}
\]

Where \(\Omega_i = \Omega_{c1} e^{i\phi_{c1}} + \Omega_{c2} e^{i\Delta_1 t + \phi_{c2}}\) and \(\Omega_i = \Omega_{p1} e^{i\phi_{p1}} + \Omega_{p2} e^{i\Delta_1 t + \phi_{p2}}\) are effective Rabi frequencies containing the time dynamics of both fields. Explicitly plugging in these time dependent relations, and solving for frequency terms that match the probe and signal, we obtain the following relations for the output polarizability:

\[
P(\omega_p) = \frac{i\mu_{13}}{2i\Delta_2 + \Gamma} + \frac{\mu_{13} \mu_{23}^* E_p E_{c1} E_{c2}}{\gamma(2i\Delta_1 + \Gamma)^2} e^{i(\omega_p t + \phi_p - \phi_{c1})}
+ \frac{\mu_{13} \mu_{23}^* E_p (E_{c1}^2 + E_{c2}^2)}{\gamma(2i\Delta_1 + \Gamma)^2} \tag{3}
\]

\[
P(\omega_s) = \frac{i\mu_{13}}{2i\Delta_2 + \Gamma} + \frac{\mu_{13} \mu_{23}^* E_p E_{c1} E_{c2}}{\gamma(2i\Delta_1 + \Gamma)^2} e^{i(\omega_p t + \phi_p + \phi_{c2} - \phi_{c1})}
+ \frac{\mu_{13} \mu_{23}^* E_p (E_{c1}^2 + E_{c2}^2)}{\gamma(2i\Delta_1 + \Gamma)^2} \tag{4}
\]

Where \(P(\omega_p)\) and \(P(\omega_s)\), obtained from \(P_{out} = Tr[\rho P]\), are the terms proportional to \(e^{i\omega_p t}\) and \(e^{i\omega_s t}\) respectively. Here we see that the typical four-wave mixing terms generate phase-controllable quantum coherence in the atom. Similar large cross-phase modulations have been seen in a closed-loop four-level system for cold atoms\[21\]. This phase-controllable output is consistent with fully-quantum theoretical predictions \[22\].

The geometrical phase shift can be obtained from finding the argument of the output:

\[
\Delta \phi_{\omega_p} = \arctan \left( \frac{\text{imag}(P_{out})}{\text{real}(P_{out})} \right) \tag{5}
\]

\[
\Delta \phi_{\omega_s} = \arctan \left( \frac{\text{imag}(P_{out})}{\text{real}(P_{out})} \right) \tag{6}
\]

A particularly noteworthy feature of this system is its independence on signal input power. Unlike a Kerr Non-linearity, which has an increasing phase shift per photon added to the signal field, this observed phase shift is independent of signal power, as long as the probe and signal are of equal strengths.

### B. Experimental Setup

To implement this scheme, we use a double-Λ atomic system, in which both individual Λ subsystems exhibit EIT. Here, two hyperfine ground states are coupled to an excited state by weak probe fields called probe and signal (\(\Omega_p\) and \(\Omega_s\)) and two strong control fields (\(\Omega_{c1}\) and \(\Omega_{c2}\)). We utilize two external cavity diode lasers phase-locked
at 6.8 GHz corresponding to the $^{87}\text{Rb}$ ground state splitting. For the first $\Lambda$ system, the probe and control field are 400 MHz red detuned from the $|5S_{1/2}, F = 1\rangle \Rightarrow |5P_{1/2}, F = 1\rangle$ and the $|5S_{1/2}, F = 2\rangle \Rightarrow |5P_{1/2}, F = 1\rangle$ transition respectively. The signal and control field for the second $\Lambda$ system are 80 MHz red detuned from the probe and first control field respectively, see Fig. 1. Both control fields are fixed to the same linear polarization, while probe and signal fields are set to have the same orthogonal polarization to allow for convenient separation of the control fields after atomic interaction. The intensities of the probe and signal fields are temporally modulated by means of acousto-optical modulators (AOM). For our medium we use a 7.5 cm long glass cell with anti-reflection coated windows containing isotopically pure $^{87}\text{Rb}$ with 10 Torr of Ne buffer gas kept at a temperature of 336 K. In this medium, 2 $\mu$s long single-photon pulses can be transmitted through the EIT medium with 80% transmission in the presence of the first control field $\Omega_1 = 45mW$. The addition of the second control field $\Omega_2 = 5mW$ results in a total transmission of 50%.

III. PHASE MEASUREMENT

A. Homodyne Detection of Pulse Sequence

A homodyne detector can obtain phase information of light through interference with a strong coherent source, producing an output voltage of the form:

$$V_{\text{homo}} = E_pE_{LO}\cos(\phi_{LO} - \phi_p) = E_pE_{LO}\cos(\omega_{pz}t - \phi_p)$$

where $E_p$ ($E_{LO}$) and $\phi_p$ ($\phi_{LO}$) are the amplitude and phase respectively of the probe (local oscillator). The phase of the probe, $\phi_p$, relative to the local oscillator can be obtained by scanning the local oscillator phase (with a frequency of $\omega_{pz}$) and fitting the resultant voltage $V(t) \propto \cos(\omega_{pz}t - \phi_p)$ to a cosine curve to extract the offset, $\phi_p$.

While the homodyne detector measures phase information relative to the local oscillator, more information is required to relate the light’s phase change in the probe generated by the signal field. To obtain this phase shift, we subtract the obtained phase (relative to the LO) when both the probe and signal are on, with a reference phase (relative to the LO) when the signal is off:

$$\Delta \phi = (\phi_{ref} - \phi_{LO}) - (\phi_{out} - \phi_{LO}) = \phi_{out} - \phi_{ref}$$

In order to have an accurate measurement of $\Delta \phi$, $\phi_{out}$ and $\phi_{ref}$ must both be measured within a window smaller than experimental fluctuations of the phase (such as slight path differences created by air fluctuations).

This can be achieved by sending an alternating pulse sequence. By sending pulses of light and extracting the peaks of the pulses, an identical voltage $V(t)$ to the DC case can be obtained. Then by alternating the phase-shifted case (in which the signal and probe pulses are simultaneously on) with the reference case (in which the signal pulse is off), the phase shift can be obtained simply by comparing the fits of peaks of the two pulse sequences, as shown in Figure 2a.

The experiment was repeated every 60$\mu$s, and a sequence of three 2$\mu$s long probe pulses with 20$\mu$s interpulse delay were directed to the medium. The second probe pulse was sent with an identically-sized and temporally-overlapping signal pulse, while the first and last pulses contained simply probe and signal fields. Control fields 1 and 2 were on during the whole cycle, therefore, the second probe pulse went through the double-$\Lambda$ system and picked up large optical phase shift due to the presence of the signal field, whereas the first probe pulse experienced electromagnetically-induced transparency EIT and was considered as the reference, Fig1b.
B. Analysis of Input-phase vs. Phase-shift

As shown in Equation 6, we theoretically expect the output phase-shift $\Delta \phi$ due to influence of the signal field to be a function of the input phases of both the probes ($E_p$, $E_s$) and the control fields ($E_{c1}$, $E_{c2}$). To experimentally observe this phase-phase relationship we linearly vary the phase of one of our control fields, $E_{c2}$, with a piezo actuator. By choosing the frequency of the piezo changing the phase of the local oscillator to be much larger than the frequency of the piezo changing the input control phase $\omega_{pz}^{LO} \gg \omega_{pz}^{C1}$, the phase of this phase-varying control field can be approximated as constant in each individual $2\pi$ cycle of the local oscillator phase. Specifically, we scan the phase of our local oscillator at $\omega_{pz}^{LO} = 200Hz$ and our control field at $\omega_{pz}^{C1} = 5Hz$. Each $2\pi$ repetition of the homodyne’s LO phase provides a particular “shot” in which the output phase $\Delta \phi = \phi_{out} - \phi_{ref}$ can be obtained by fitting the offsets of sinusoids formed by the peaks of the first and second pulses in the pulse sequence (as discussed in the previous section).

Using a Spectrum M2i.2031 digitizer card with 2 GSamples of onboard memory, we store continuous homodyne data in bursts of 1.3 seconds, with an internal sampling clock of 100MHz. We acquire these large bursts of data, split the data into individual 5ms “shots” (corresponding to the time to make a complete $2\pi$ cycle of the local oscillator phase), and obtain the phase-shift per shot (by fitting the phase shift). These phase shifts can then be plotted to observe the output phase shift as a function of input phase.

As expected, the output phase shift follows the control field phase, with some additional variation in the phase on a slower timescale due to air fluctuations. This can be seen in Figure 2c and d, for equal and unequal powers of the probe and signal pulses.

As discussed in the semiclassical model, while the phase shift applied on the probe pulses $\Delta \phi$ and on the four-wave mixing pulses $\Delta \phi_{FWM}$ depends on the overall input phases of the four fields, the phase shift does not scale with probe and signal power if the strength of the two fields are scaled equally. We observe similar results to Figure 2c when using input photon numbers ranging from hundreds of photons to only 0.95 photons.

We have repeated the experiment for the situation where the probe field is substantially stronger than the signal field. In this case, the probe pulse contains 17.5 photons on average while signal pulse has only 0.64 photons. Consequently, the probe field is dominant in the medium and therefore the double-$\Lambda$ phase shift does not follow the input overall phase shift and FWM phase shift $\Delta \phi_{FWM}$, Fig 2d.

Additionally, to obtain statistical information to be used for an estimation of the quantum state, we then collect individual “shots” (corresponding to the time to make a complete $2\pi$ cycle of the local oscillator phase) into bins of output phase shifts. Each bin contains mul-

Additionally, since four-wave mixing (FWM) is very relevant in understanding this system, we add a third pulse to the sequence in which the signal field pulse is on and the probe is off, creating pulsed four-wave-mixing with an associated phase shift, $\Delta \phi_{FWM} = (\phi_{FWM} - \phi_{ref})$. The pulse sequences used for the experiment can be seen in Fig. 1b.

FIG. 3: Reconstructed density matrices in the case of $\pi$-phase shifted states. a) probe field reconstructed on homodyne without Rb cell. b) Reconstructed from first pulse in sequence in which signal field is off c) Reconstructed from third pulse in sequence in which probe field is on and FWM is generated by signal d) Reconstructed from third pulse in sequence in which fully double-$\Lambda$ scheme is used.
multiple 2π “shots,” and each individual “shot” has a set of voltage and time values associated with the peaks of the pulses. These voltage and time values, can then be converted into measurements of the electric field quadrature $X(\theta)$.

C. Quantum State Reconstruction using post-selected quadratures

To estimate the quantum state of the phase shifted output light measured by the homodyne detector, we reconstruct the quantum state using the assembled, post-selected quadrature statistics $X(\theta)$.

The voltage output of the homodyne is quantum mechanically a measurement of the generalized quadrature:

$$X_\theta = a e^{-i\theta} + a^\dagger e^{i\theta}$$

Using the assembled quadrature statistics extracted from our pulse sequences and the techniques for identifying and binning by output phase shifts, we can perform maximum-likelihood estimation to construct density matrices of the underlying quantum states, sorted by their post-selected, phase-shifted values.

We perform a quantum state reconstruction on each of the states associated with each pulse sequence: EIT, double-Λ, and FWM. There is an associated reconstructed density matrix for each post-selected bin of output phase shift. Additionally we also measure and reconstruct the density matrix for the probe and signal fields without the cell. These density matrix elements, in the Fock state basis, are plotted for the post-selected phase shift of $\pi \pm .4$ in Figure 3.

IV. HIGH FIDELITY QUANTUM STATE RETRIEVAL

By comparing the quantum state of the phase-shifted output of the double-Λ, $\rho_{DL\theta}$, with the quantum state of the light without the cell, $\rho_{in\theta}$, the fidelity can be calculated.

$$F_\theta = tr(\sqrt{\sqrt{\rho_{in\theta}} \rho_{DL\theta} \sqrt{\rho_{in\theta}}})$$

This fidelity is plotted as a function of output phase shift. As seen in Figure 5c, the fidelity reaches its highest value of 94% at $\Delta \phi = 1.29$. The fidelity is also observed to reach $F = 88\%$ at $\Delta \phi = 3.5$, indicating that, despite being mediated by a room-temperature FWM process, the phase-shifted quantum state at the single-photon level appears accurately resembles the input state.

The reconstructed density matrices can additionally be mapped to a Wigner function representation. Figure 4 illustrates an input-output visualization of how the phase-shifted output state moves across phase-space for different reconstructed states. In particular, in Figure 4c, a π phase-shifted input probe, consisting of an average of 0.64 photons, is created by a signal pulse of 4.5 photons.

While our system’s average behavior is consistent with a simple semiclassical model, our quantum state reconstruction goes beyond this semiclassical characterization in quantifying the quantum state of the light. Despite this, most of the fidelity’s relationship to input phase can still be understood by this semiclassical model. As illustrated in Figure 5b, the phase of the four-wave-mixing is controlled by the relative phase of the input fields. This relationship on phase can be seen illustrated in Figure 5e. When the FWM is in-phase with the probe, the fidelity is lower because the quantum interference is constructive, producing a probe output that is larger than its input. When the relative phase of the system is out-of-phase, the amplitude of the output flips in sign because of the unbalanced size of the four-wave-mixing and the probe.

V. DISCUSSION: TOWARDS QUANTUM GATES

We have shown that a room-temperature atomic system can produce π conditional phase shifts in single photon level fields triggered by few-photon signal fields. In
addition, we have shown that the $\pi$ phase-shifted quantum states, reconstructed from the quadrature statistics, still posses 88% fidelity with respect to the original input. The primary mechanism for the phase-shift is due to phase-controllable frequency conversion via the nonlinear process. This is unlike traditional “giant Kerr” schemes, in which the $\chi^3$ nonlinearity modifies the dispersive properties of the medium. While quantum interference in our system is generated by a $\chi^3$ nonlinearity, this is because the quantum coherence terms in $\rho_{12}$ generating four-wave mixing interfere linearly with respect to the input fields.

The focus of our analysis has been primarily on the Fock-state reconstruction of a single output mode. Extending our analysis to multiple modes will provide us with further insights both from a fundamental and applied perspective. Extending our analysis to a quantized multi-temporal-mode framework could provide a better fundamental understanding of quantum processes. For example, cross-phase modulation schemes using the Kerr effect have been shown to have fundamental limitations for quantum computation, such as large phase shifts requiring low temporal fidelity due to pulse distortion [23]. In our work, analyzing the quantum state at the peaks of pulses instead of continuous waves, allowed us to gain gained extra information regarding the relationship between quantum temporal modes and their fidelities. A full picture of the inner workings of these fundamental photon-photon interactions could be obtained by additionally reconstructing the temporal modes and investigating different temporal wave-function widths and shapes. We think our results provide for a first time a framework towards a full characterization of an atom mediated photon-photon process.

Moreover, we have analyzed our system by looking at the quantum state of the probe, after the phase-shift is triggered by a coherent state at the single photon level. A natural extension to our experiment will be to induce the phase-shift using a true single-photon Fock state. In order to characterize both the probe and signal quantum states after interaction, we could use two homodyne detectors to perform multi-spatial mode tomography. We can then perform multi-mode process tomography, thus gaining unparallel access to the creation of quantum correlations between the modes.

By incorporating phase-stabilization to our control fields, we envision our system to find immediate application as an all-optical switch. Such a switch would simultaneously work for much lower light levels than current implementations while simultaneously requiring much less experimental overhead.

VI. ACKNOWLEDGMENTS

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