Spectral analysis for cascade-emission-based quantum communication in atomic ensembles

H H Jen

Physics Department, National Tsing Hua University, Hsinchu 300, Taiwan, Republic of China
E-mail: sappyjen@gmail.com

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Abstract
The ladder configuration of atomic levels provides a source for telecom photons (signal) from the upper atomic transition. For rubidium and caesium atoms, the signal field has the range around 1.3–1.5 $\mu$m that can be coupled to an optical fibre and transmitted to a remote location. Cascade emission may result in pairs of photons, the signal entangled with the subsequently emitted infrared photon (idler) from the lower atomic transition. This correlated two-photon source is potentially useful in the DLCZ (Duan–Lukin–Cirac–Zoller) protocol for the quantum repeater. We implement the cascade emission to construct a modified DLCZ quantum repeater and investigate the role of the time-frequency entanglement in the protocol. The dependence of the protocol on the resolving and non-resolving photon-number detectors is also studied. We find that the frequency entanglement deteriorates the performance but the harmful effect can be diminished by using shorter pump pulses to generate the cascade emission. An optimal cascade-emission-based DLCZ scheme is realized by applying a pure two-photon source in addition to using detectors of perfect quantum efficiency.

1. Introduction
Quantum communication has opened up the possibility of transmitting quantum information over a long distance. A quantum repeater protocol proposed by Briegel et al [1, 2] fulfils such a long-distance system. Subsequently, Duan, Lukin, Cirac and Zoller (DLCZ) [3] suggested a long-distance quantum communication based on atomic ensembles. This scheme involves Raman scattering of incoming light from the atoms with the emission of a signal photon. The photon is then correlated with the coherent excitation of the atomic ensemble. The information may be transferred through light to another atomic ensemble or retrieved by a reverse Raman scattering process, generating an idler photon directional correlated with the signal one [4–8]. The signal and idler photons in alkali gases are in the near-infrared spectral region, which mismatches the telecommunication bandwidth optical fibre. Therefore, an alternative process that is able to generate telecom wavelength photons correlated with atomic spin excitations [9–11] would provide the essential step towards practical long-distance quantum communication.

The alkali atomic cascade transition shown in figure 1 is able to generate telecom wavelength light, the signal, from the upper transition, and a near-infrared field, the idler, from the lower one. The telecom light can travel through the fibre with minimal loss, while the near-infrared field is suitable for storage and retrieval in an atomic quantum memory element. Their use in a quantum-information system requires quantum correlations between stored excitations and the telecom field. It is interesting to assess the cascade scheme in the DLCZ protocol given that it could potentially reduce transmission losses in a quantum telecommunication system.

Correlated photon pairs may be generated by parametric down-conversion (PDC) [12–14]. The degree of entanglement can be quantified by Schmidt mode decomposition [15, 16], allowing the influence of group-velocity matching [17] to be assessed. A pure single-photon source is a basis element for quantum computation by linear optics (LOQC) [18], and it can be conditionally generated by measurement [19]. A
similar approach can be applied to the study of the transverse degrees of freedom in type II PDC [20] and PDC in a distributed microcrystal [21]. In the photonic-crystal fibre (PCF), a factorizable photon pair can be generated by spectral engineering [22]. The spectral effect has been discussed in relation to a quantum-teleportation protocol [23] as a first step towards quantum communication.

This motivates the research in this paper where we study the spectral effect of a correlated photon pair generated from the cascade atomic ensemble in the DLCZ scheme. The DLCZ scheme is based on entanglement generation and swapping, and quantum state transfer, which make up the basic elements for long-distance quantum communication. Generating entanglement is the first step in quantum-information processing, and entanglement swapping is the essence to distribute the entanglement over distant places. Quantum state transfer enables the secure transmission to eavesdropping which is therefore of great practical interest [24–26].

In this paper, we start from formulating a two-photon state generated in a cold atomic ensemble. We use Schrödinger’s equation to investigate the correlated signal and idler photons spontaneously emitted from two driving lasers via the four-level atomic structure. The essence of phase-matching in spontaneously emitted from two driving lasers via the four-wave mixing (FWM) conditions is discussed in section 2, and we calculate the second-order correlation function to show the bunching behaviour of the cascade-emitted photons. In section 3, we briefly review the Schmidt decomposition that is used to analyse the frequency entanglement and mode functions of the two-photon source. We characterize these spectral properties of the correlated two-photon state for different superradiant decay constants and study how the laser excitation pulse modifies their spectral prole. We then demonstrate the modified DLCZ scheme, a quantum repeater protocol, which employs cascade emission in section 4. We reconstruct the elements of the DLCZ scheme including entanglement generation, entanglement swapping, effective ‘polarization’ maximally entangled (PME) state projection and quantum teleportation. We investigate how frequency entanglement of the cascade photon pair influences these elements, and study their performances (fidelity, heralding and success probabilities) for two types of photon detectors (resolving photon number or not) along with the dependence of quantum efficiency. We then conclude in section 5 and discuss the alternative method of generating the telecom photon by frequency conversion. The details of the Hamiltonian and Schrödinger’s equation are described in appendix A. In appendix B, we derive the multimode two-photon state and conditional output density operators used in the modified DLCZ protocol.

2. A correlated two-photon state

We consider N cold atoms that are initially prepared in the ground state interacting with four independent electromagnetic fields. As shown in figure 1, two driving lasers (of the Rabi frequencies Ωa and Ωb) excite a ladder configuration |0⟩ → |1⟩ → |2⟩. Two quantum fields, signal a⃗s and idler a⃗i, are generated spontaneously. The four atomic levels can be chosen as ((0), |1⟩, |2⟩, |3⟩) = (|5S1/2, F = 3⟩, |5P3/2, F = 4⟩, |4D5/2, F = 5⟩) and (|5P3/2, F = 4⟩) [9]. The atoms adiabatically follow the two excitation pulses and decay through the cascade emission of signal and idler photons. Based on the discussion in appendix A, we permit only single atomic excitations under the condition of large detuning, △1 ≫ NΩa/2. The Hamiltonian and the coupled equations of the atomic dynamics are detailed in appendix A.

To correctly describe the frequency shifts arising from dipole–dipole interactions, we need to include the non-rotating wave approximation (non-RWA) terms in the electric dipole interaction Hamiltonian. In appendix A, we consider only RWA terms for simplicity and the non-RWA terms would allow virtual transitions that in effect add to the frequency shifts in an appropriate way [27, 28]. The frequency shift has contributions from the single-atom Lamb shift and a collective frequency shift. The Lamb shift is assumed to be renormalized into the single-atom transition frequency distinguishing it from the collective shift due to the atom–atom interaction.

Writing the state vector |ψ(t)⟩ in a basis restricted to single atomic excitations, and single pairs of signal and idler photons, we can introduce the probability amplitudes

\[ C_{\mu, \lambda}(t) = \sum_{\mu=1}^{N} e^{-i\vec{k}_{\mu} \cdot \vec{r}_{\mu}} (3, \mu), k_{\mu, \lambda}, |\psi(t)\rangle, \]  

and

\[ D_{ij}(t) = \langle 0, k_{i, \lambda}, k_{j, \mu}, |\psi(t)\rangle, \]  

where \( \vec{k}_{\mu} \) here is equivalent to \( \vec{q}_{\mu} \) defined in appendix A and is denoted as \( \vec{k}_{\mu} + \vec{k}_{\mu} - \vec{k}_{i} \), from the FWM condition which will be demonstrated later. Note that \( C_{\mu, \lambda}(t) \) is an amplitude for a phased excitation of the ensemble of atoms subsequent to the signal photon emission and \( (k_{i, \mu}, \lambda, k_{j, \mu}) \) represent wave vectors and polarization indices for signal and idler fields, respectively.

After adiabatically eliminating the laser excited levels in the equations of motion, we are able to simplify and derive the amplitude \( C_{\mu, \lambda} \) and the signal–idler (two-photon) state amplitude \( D_{ij} \) as shown in appendix A.

\[ C_{\mu, k}(t) = g_{k}^{\ast} (\epsilon_{k_{\mu}, \lambda_{\mu}} \cdot \hat{a}_{\mu}) \sum_{\mu} e^{i\Delta \vec{k} \cdot \vec{r}_{\mu}} \times \int_{0}^{t} dt' e^{i(\omega_{0} - \omega_{23})t'} e^{(-\frac{\Delta_{2}}{2} + i\omega_{0})t'} b(t'), \]  

Figure 1. Four-level atomic ensemble interacting with two driving lasers (solid) with the Rabi frequencies \( \Omega_{a} \) and \( \Omega_{b} \). Cascade emissions, signal and idler photons, are labelled by \( \hat{a}_{s} \) and \( \hat{a}_{i} \), respectively, and \( \Delta_{1} \) and \( \Delta_{2} \) are the single- and two-photon laser detunings.
indicating a spectral width $\Gamma_1$ where $G_{\cal N}$ is calculated as $\frac{\Gamma_1^2}{4\Delta_2^2 N}$ in equation (6), the second-order correlation function is proportional to the product of the Rabi frequencies. The coupling constants $g_{\alpha(i)}$, polarization direction $\hat{e}_{\alpha(i)}$, and unit direction of dipole operators $\hat{d}_s$ are for signal and idler fields, respectively. Various definitions of the optical frequencies $\omega$ and laser detuning $\Delta_2$ can be found in appendix A.

The factor $\sum_{\mu} e^{i\Delta \hat{r}} \hat{n}_\mu$ reflects phase-matching of the interaction under conditions of FWM when the wavevector mismatch $\Delta k = \vec{k}_i + \vec{k}_b - \vec{k}_s - \vec{k}_i \rightarrow 0$. The radiative coupling between atoms results in the appearance of the superradiant decay constant,

$$\Gamma_3 = (N\bar{\mu} + 1) \Gamma_3,$$

where $\Gamma_3$ is the natural decay rate of the $|3\rangle \rightarrow |0\rangle$ transition, and $\bar{\mu}$ is a geometrical rate constant depending on the shape of the atomic ensemble. An expression for the collective shift $\delta\omega_0$ is given in appendix A. For a cylindrical atomic ensemble, the decay factor $N\bar{\mu} + 1$ depends on the height and radius as shown in equation (A.17). $N\bar{\mu} + 1 \approx 4$ and 6 which are comparable to the operating conditions of the experiment [9].

We use normalized Gaussian pulses as an example where $\Omega_a(t) = \frac{1}{\sqrt{\pi}} \Omega_b e^{-t^2/\tau^2}$, $\Omega_b(t) = \frac{1}{\sqrt{\pi}} \Omega_b e^{-t^2/\tau^2}$, so that the two pulses are overlapped with the same pulse width. $\Omega_{a,b}$ is the pulse area, and let $\Delta\omega_0 \equiv \omega_s - \omega_{23} - \Delta_2 - \delta\omega_0$, $\Delta\omega_0 \equiv \omega_s - \omega_{3} + \delta\omega_0$. In the long time limit, we have the probability amplitude $D_{\mu}$,

$$D_{\mu}(\Delta\omega_0, \Delta\omega_0) = \frac{\Omega_a \Omega_b g_\alpha^2 g_\alpha (e^{i\hat{n}_\mu} \cdot \hat{d}) (e^{i\hat{n}_\mu} \cdot \hat{d})}{4\Delta_2 \Delta_2} \times \frac{\sum_{\mu} e^{i\Delta \hat{r} \hat{n}_\mu} e^{-(\Delta\omega_0 + \Delta\omega_0)^2 t^2/8}}{\sqrt{2\pi \tau}} \frac{\Gamma_1^2}{2 - i\Delta\omega_0},$$

indicating a spectral width $\Gamma_1^2/2$ for the idler photon in a Lorentzian distribution modulating a Gaussian profile with a spectral width $2\sqrt{2/\tau}$ for the signal and idler. Energy conservation of signal and idler photons with driving fields at their central frequencies corresponds to $\omega_s + \omega_i = \omega_s + \omega_i$, which makes $\Delta\omega_0 + \Delta\omega_0 = 0$; the collective frequency shifts cancel.

Using the asymptotic form of the two-photon state given in equation (6), the second-order correlation function [29] is calculated as

$$G_{\alpha(i)}^{(2)} = |\langle \psi(\infty) | \hat{E}^\dagger_s (\vec{r}_1, t_1) \hat{E}^\dagger_i (\vec{r}_2, t_2) \hat{E}^\dagger_i (\vec{r}_2, t_2) \hat{E}^\dagger_s (\vec{r}_1, t_1) | \psi(\infty) \rangle|,$$

where

$$\hat{E}^\dagger_s (\vec{r}_1, t_1) = \sum_{k_s, i_s} \sqrt{\frac{\hbar \omega_s}{2 e_0 V}} \hat{a}_{k_s, i_s} \hat{e}_{k_s, i_s} e^{i\vec{k}_s \vec{r}_1 - \omega_s t_1},$$

$$\hat{E}^\dagger_i (\vec{r}_2, t_2) = \sum_{k_i, i_i} \sqrt{\frac{\hbar \omega_i}{2 e_0 V}} \hat{a}_{k_i, i_i} \hat{e}_{k_i, i_i} e^{i\vec{k}_i \vec{r}_2 - \omega_i t_2}.$$

$|\psi(\infty)\rangle$ denotes the state vector in the long time limit that involves the ground-state and two-photon state vectors. Free electromagnetic fields, signal and idler photons, at space $(\vec{r}_1, \vec{r}_2)$ and time $(t_1, t_2)$ are $\hat{E}^\dagger_s$ and $\hat{E}^\dagger_i$ where $\langle \dagger \rangle$ denotes their positive frequency part. For the second-order correlation function, only $D_{\mu}$ contributes to it; then following the standard procedure of $G^{(2)}_{\alpha(i)}$ calculation [29], we have

$$\sqrt{G^{(2)}_{\alpha(i)}} \propto \sum_{\mu} e^{i\Delta \hat{r} \hat{n}_\mu} e^{-2(\Delta\omega_0)^2 t^2/8} e^{-\frac{\Gamma_1^2}{2}} \Theta(\Delta\omega_0 - \Delta\omega_0),$$

where $\Delta\omega_0 \equiv t_1 - \frac{\vec{r}_1}{\epsilon}$ and $\Delta\omega_0 \equiv t_2 - \frac{\vec{r}_2}{\epsilon}$. The step function $\Theta$ shows the causal connection between signal and idler emissions and is due to the complex integral with the pole at $\Delta\omega_0 = -\frac{\Gamma_1^2}{2} - \delta\omega_0$ in the lower half plane. The emission time for the signal field $(t_1 - \frac{\vec{r}_1}{\epsilon})$ is within the pulse envelope of width $\tau$, and the idler photon decays with a superradiant constant $\Gamma_3^2/2$.

If we let $\Delta t \equiv \Delta\omega_0 - \Delta\omega_0$ and choose $\Delta\omega_0 = 0$ as the origin in time (idler gating time), then we have the second-order correlation function

$$G^{(2)}_{\alpha(i)} (\Delta t) = |\Phi_{\alpha(i)}(\Delta t)|^2 \propto e^{-\Gamma_3^2 \Delta t}$$

where $\Delta t \geq 0$. (11)

It resembles the result for the second-order correlation function in the case of the single atom, whereas here we have an enhanced decay rate due to the atomic dipole–dipole interaction. This exponential correlation function indicates the bunching property of cascade photons [30] showing an immediate emission of the idler photon following the signal one.

### 3. Schmidt decomposition

We would like to perform an analysis of the entanglement properties of our cascade-emission source. In addition to polarization entanglement, a characterization of frequency space entanglement is required to clarify its suitability in the DLCZ protocol [3].

In the long time limit, the state function is given by

$$|\psi\rangle = |0, \text{ vac}\rangle + \sum_{\alpha,i} D_{\alpha,i} |1_{k_s, i_s}, 1_{k_i, i_i}\rangle,$$

where $D_{\alpha,i}$ can be found in equation (6) and $|0, \text{ vac}\rangle$ is the joint atomic ground and photon vacuum state. The shorthand notations $s = (k_s, \lambda_s)$ and $i = (k_i, \lambda_i)$ are for different spatial modes $k_{\alpha(i)}$ and two-degree-of-freedom polarizations $\lambda_{\alpha(i)}$.

The spatial correlation of the two-photon state in the FWM condition can be eliminated by pinholes or by coupling to a single-mode fibre, so we consider only the continuous frequency space. For some specific polarizations $\lambda_s$ and $\lambda_i$, we have the state vector $|\Psi\rangle$,

$$|\Psi\rangle = \int f(\omega_s, \omega_i) a^\dagger_s (\omega_s) a^\dagger_i (\omega_i) |0\rangle d\omega_s d\omega_i,$$

where

$$f(\omega_s, \omega_i) = e^{-i(\Delta\omega_0 + \Delta\omega_0)^2 t^2/8}. $$
Following the theoretical work on two-photon pulses generated from down-conversion by Law et al. [15], the quantification of entanglement can be determined in the Schmidt basis where the state vector is expressed as
\[ |\Psi\rangle = \sum_n \sqrt{\lambda_n} \hat{b}_n^{\dagger} \hat{c}_n^{\dagger} |0\rangle, \]
(15)
where \( \hat{b}_n^{\dagger} \) and \( \hat{c}_n^{\dagger} \) are the effective creation operators and \( \lambda_n \) (no confusion with the polarization index \( \lambda_n \)) are the probabilities in the corresponding two-photon emission modes \( n \). If \( \lambda_1 = 1 \), it means a pure two-photon emission. The eigenfunctions \( \psi_n \) and \( \phi_n \) are the solutions of the eigenvalue equations,
\[ \int K_1(\omega, \omega') \psi_n(\omega') d\omega' = \lambda_n \psi_n(\omega), \]
(18)
\[ \int K_2(\omega, \omega') \phi_n(\omega') d\omega' = \lambda_n \phi_n(\omega), \]
(19)
where \( K_1(\omega, \omega') = \int f(\omega, \omega') f^*(\omega', \omega) d\omega_1 \) and \( K_2(\omega, \omega') = \int f(\omega_2, \omega) f^*(\omega_2, \omega') d\omega_2 \) are the kernels for the one-photon spectral correlations [15, 16]. The orthogonality of eigenfunctions is \( \int \psi_i(\omega) \psi_j(\omega) d\omega = \delta_{ij} \), \( \int \phi_i(\omega) \phi_j(\omega) d\omega = \delta_{ij} \), and the normalization of the quantum state requires \( \sum_n \lambda_n = 1 \).

In the Schmidt basis, the von Neumann entropy may be written as
\[ S = -\sum_{n=1}^{\infty} \lambda_n \ln \lambda_n. \]
(20)
If there is only one non-zero Schmidt number \( \lambda_1 = 1 \), the entropy is zero, which means no entanglement and a factorizable state. For more than one non-zero Schmidt number, the entropy is larger than zero and the bipartite entanglement is present.

The kernel in equation (14) has all the frequency entanglement information; the entanglement means that \( f(\omega_x, \omega_y) \) cannot be factorized in the form \( g(\omega_x) h(\omega_y) \), a multiplication of two separate spectral functions. By inspection, the Gaussian profile of signal and idler emission is a source of correlation. The joint spectrum \( \Delta \omega/\Delta \omega_0 + \Delta \omega_0 \) is confined within the width of the order of \( 1/\tau \). The Lorentzian factor associated with the idler emission has a width governed by the superradiant decay rate.

In figure 2, we show the Schmidt decomposition of the spectrum. We use a moderate superradiant decay constant \( N\bar{\mu} + 1 = 5 \), comparable to [9], and a nanosecond pulse duration \( \tau = 0.25 \) (in units of \( 1/10 \approx 26 \) ns), and \( \Gamma_3/2\pi = 6 \) MHz. Due to slow convergence associated with the Lorentzian profile, we use a frequency range up to \( \pm 1200 \) (in unit of \( \Gamma_3 \)) with \( 2000 \times 2000 \) grid. The numerical error in the eigenvalue calculation is estimated to be about 1% error. In this case, the largest Schmidt number is 0.8 and the corresponding signal mode function has a FWHM Gaussian profile \( \sqrt{2} \ln(2)/\tau \approx 19\Gamma_3 \). The idler mode function \( \phi_i \) reflects the Lorentzian profile in the spectrum at the signal peak frequency \( \Delta \omega_0 = 0 \),
\[ f(\Delta \omega_0 = 0, \Delta \omega_0) = e^{-(\Delta \omega_0)^2/8} \]
(21)
where a relatively broad Gaussian distribution is overlapped with a narrow spread of the superradiant decay rate (FWHM \( > (N\bar{\mu} + 1)\Gamma_3/2 \)).

Figure 3 shows that the cascade-emission source is more entangled if the superradiant decay constant or the pulse duration increases. We note that the Gaussian profile aligns...
the spectrum along the axis $\Delta \omega_2 = -\Delta \omega_3$ and the spectral width for the signal photon at the centre of the idler frequency distribution ($\Delta \omega_1 = 0$) is determined by the pulse duration $\tau$. For a shorter pulse $\tau^{-1} > (N\tilde{\mu}+1)^{1/2}$, the joint Gaussian profile has a larger width, and the spectrum is cut off by the Lorentzian idler distribution. A larger width leads to a less entangled source and distributes the spectral weight mainly along the crossed axes $\Delta \omega_1 = 0$ and $\Delta \omega_2 = 0$. A narrow Lorentzian profile cuts off the entanglement source $e^{-|\Delta \omega_1 + \Delta \omega_3|^2/8}$ tilting the spectrum along the line $\Delta \omega_2 = 0$. In the opposite limit, $\tau^{-1} < (N\tilde{\mu}+1)^{1/2}$, the spectrum is highly entangled corresponding to tight alignment along the axis $\Delta \omega_3 = -\Delta \omega_1$ (figure 3(c)).

Note that the short pulse duration ($\tau \geq 0.25$ (6.5 ns)) should not violate the assumption of adiabaticity $\tau \gg 1/\Delta_1$ or $1/\Delta_2$.

The Schmidt analysis and calculation of von Neumann entropy show that signal–idler fields are more entangled if the ensemble is more optically dense, corresponding to stronger superradiance. For the DLCZ protocol, we wish to avoid the frequency entanglement. The superradiance may be reduced with smaller atomic densities but good qubit storage and retrieval efficiency require a moderate optical thickness [9]. A better approach involves using the short pulse excitation $\tau^{-1} > (N\tilde{\mu}+1)^{1/2}$. We will investigate the spectral properties in more detail for the DLCZ scheme in the next section. Note that there has been a development in the setting of spontaneous PDC to generate frequency-uncorrelated entangled photons by using shorter pump pulses for scalable all-optical quantum-information processing [31].

4. DLCZ scheme with cascade emission

In the DLCZ protocol, a weak pump laser Raman scatters a single photon generating a quantum correlated spin excitation in the ensemble. By interfering the Raman photons generated from two separate atomic ensembles on a beam splitter (B.S.), the DLCZ entangled state $(|01\rangle + |10\rangle)/\sqrt{2}$ is prepared conditioned on one and only one click of the detectors after the B.S. Hence, $|0\rangle$ and $|1\rangle$ represent the state of zero or one collective spin excitation stored in the hyperfine ground-state coherences. This state originates from indistinguishable photon paths. The error from multiple excitations can be made negligible if the pump laser is weak enough.

As shown in figure 4, we consider instead that one of the ensembles employs cascade emission. The idea is for cascade emission to generate a telecom photon ($\hat{\alpha}_s$) for transmission in the optical fibre, and an infrared photon that interferes locally with the Raman photon generated in the $\Lambda$-type atomic ensemble. In this way, the interference of the infrared photons generates the entangled state,

$$|\Psi\rangle = \frac{1}{\sqrt{2}}(|01\rangle_\alpha,s + |10\rangle_\alpha,s),$$

similar to the conventional DLCZ entanglement generation scheme. Now, however, instead of a stored spin excitation, we generate a telecom photon. Here we denote $|\Psi\rangle$ as a matter–light entangled state where ($\alpha,s$) represent an atomic collective spin excitation and a telecom photon, respectively.

$$|\Psi\rangle_{\text{DLCZ}} = \frac{S_\alpha^s + S_\alpha^r}{\sqrt{2}} |0\rangle_{\alpha,B}. $$

Figure 4. Entanglement generation in the DLCZ scheme using the cascade and Raman transitions in two different atomic ensembles. The large white arrows represent laser pump excitations corresponding to the dashed lines in either cascade or Raman level structures. Here $\hat{\alpha}_s$ represents the emitted telecom photon. B.S. means the beam splitter that is used to interfere the incoming photons measured by the photon detector D. Label A refers to the pair of ensembles for later reference.

Figure 5. Entanglement swapping of the DLCZ scheme using the cascade transition. Site $A$ is described in detail in figure 4 and equivalently for site $B$. The telecom signal photons are sent from both sites and interfere by B.S. midway between with detectors represented by $c_1^s$ and $c_2^r$. Synchronous single clicks of the detectors from both sites ($m^s_{1,2}$, $n^r_{1,2}$) and the midway detector ($c^s_{1,2}$) generate the entangled state between lower atomic ensembles at sites $A$ and $B$. The locally generated entanglement is swapped to distantly separated sites in this cascade-emission-based DLCZ protocol.

The entanglement swapping with the cascade emission may be implemented as shown in figure 5, and will be discussed in detail in the next section. The initial state is a tensor product of two state vectors generated locally at sites $A$ and $B$:

$$|\Psi\rangle_{AB} = (\sqrt{1 - \eta_{1A}}|0\rangle + \sqrt{\eta_{1A}}|1\rangle_1^s) \otimes (\sqrt{1 - \eta_{2A}}|0\rangle + \sqrt{\eta_{2A}}|1\rangle_2^s) \otimes (\sqrt{1 - \eta_{1B}}|0\rangle + \sqrt{\eta_{1B}}|1\rangle_1^r) \otimes (\sqrt{1 - \eta_{2B}}|0\rangle + \sqrt{\eta_{2B}}|1\rangle_2^r),$$

where ($s$, $i$) represent the signal and idler photons from the cascade emission, and ($r$, $a$) are the Raman scattered photon and the collective spin excitation. Here $\eta_1$ and $\eta_2$ are the efficiencies to generate cascade and Raman emission. Since
\( \eta_1 \) and \( \eta_2 \ll 1 \), multiple atomic excitations or multi-photon generation can be excluded.

### 4.1. Entanglement swapping

Before we proceed to expand the product state of equation (23) and investigate the spectral effects of cascade emission on the modified DLCZ scheme, we would like to address the intrinsic errors from the protocols. Consider the product state generated from the entangled states of \( A \) and \( B \) as in figure 5,

\[
|\Psi\rangle_A \otimes |\Psi\rangle_B = \left\langle \frac{|0\rangle_{\text{as}} + |1\rangle_{\text{as}}}{\sqrt{2}} \right| \otimes \left\langle \frac{|0\rangle_{\text{as}} + |1\rangle_{\text{as}}}{\sqrt{2}} \right|_B
\]

where the subscript \( (a) \) represents a stored local atomic excitation, and \( (s) \) means a telecom photon propagating towards the B.S. in the middle. We can tell from this effective state that the first component \( |1010\rangle_{\text{as}} \) contributes no telecom photons at all (two local excitations) and can be ruled out by measuring a `click’ at one of the middle detectors. The second and the third components have the components of the entangled state of quantum swapping, and the fourth one is the source of error if the photodetector cannot resolve one from two photons. The error could be corrected by using a photon-number resolving detector (PNRD) if other drawbacks like dark counts, photon losses during propagation and detector inefficiency are not considered.

Now we will formulate the entanglement swapping including the spectral effects discussed in section 3. We ignore pump-phase offsets, assuming 50/50 B.S. and a symmetric set-up \( \eta_{1a} = \eta_{1b} = \eta_1, \eta_{2a} = \eta_{2b} = \eta_2 \) for simplicity. Expand the previous joint state, equation (23) and keep the terms up to the second order of \( \eta_{1,2} \) that can contribute to the detection events \( \hat{m}_{1,2}, \hat{n}_{1,2} \).

\[
|\Psi\rangle_{\text{eff}} = \eta_1 (1 - \eta_2) |1\rangle_1 |1\rangle_1 |1\rangle_1 |1\rangle_1 B + \eta_2 (1 - \eta_1) |1\rangle_1 |1\rangle_1 |1\rangle_1 |1\rangle_1 B \\
+ \sqrt{\eta_1 \eta_2 (1 - \eta_1) (1 - \eta_2)} |1\rangle_1 |1\rangle_1 |1\rangle_1 |1\rangle_{\text{as}} B + \sqrt{\eta_1 \eta_2 (1 - \eta_1) (1 - \eta_2)} |1\rangle_1 |1\rangle_1 |1\rangle_{\text{as}} |1\rangle_{\text{as}} B \\
+ \sqrt{\eta_1 \eta_2 (1 - \eta_1) (1 - \eta_2)} |1\rangle_1 |1\rangle_1 |1\rangle_{\text{as}} |1\rangle_{\text{as}} B, \tag{25}
\]

where the cascade-emission state \( |1\rangle_1 |1\rangle_1 \) is fulfilled by measuring three clicks from the three pairs of the detectors respectively \( \hat{m}_{1,2}, \hat{n}_{1,2}, \hat{c}_{1,2} \). The quantum efficiency of the detector is considered in the protocol, and we describe a model for quantum efficiency in appendix B.1. We then use this model to describe photodetection events registered by non-resolving photon detectors (NRPD). Starting with the input density operator \( \hat{\rho}_0 = |\Psi\rangle_{\text{eff}} \langle \Psi| \), we derive the projected density operator, equation (B.16), conditioned on the three clicks of \( \hat{m}_{1,2}, \hat{n}_{1,2} \) and \( \hat{c}_{1,2} \) in appendix B.2. We use the Schmidt decomposition of the projected density operator and assume a single mode for the Raman scattered photon. We find the unnormalized density operator \( \hat{\rho}_{\text{out}}^{(2)} \) given in equation (B.16),

\[
\hat{\rho}_{\text{out}}^{(2)} = \frac{\eta_1^2 (1 - \eta_2)^2}{16} (2 - \eta_1) \eta_1 \eta_\text{eff} \left( 1 + \sum_j \lambda_j^2 \right) |0\rangle \langle 0| \\
+ \frac{\eta_1 \eta_2 (1 - \eta_1) (1 - \eta_2)}{8} \hat{S}_B + \frac{\hat{S}_A}{8} |0\rangle \langle 0| \hat{S}_A + \sum_j \lambda_j \int \phi_j(\omega) \phi_j^*(\omega) \Phi(\omega) \Phi^*(\omega) d\omega d\omega, \\
\times \left( \hat{S}_B^\dagger |0\rangle \langle 0| \hat{S}_A + \hat{S}_A^\dagger |0\rangle \hat{S}_B \right), \tag{26}
\]

where \( \eta_1 \) and \( \eta_\text{eff} \) are the quantum efficiencies of the detectors at the telecom and infrared wavelengths, respectively. \( \lambda_j \)'s are the Schmidt eigenvalues derived in section 3. The first term in equation (26) is the atomic vacuum state at sites \( A \) and \( B \) and contributes an error to the output density operator. The second term contains the components of the DLCZ entangled state. We can define the fidelity \( F \) as the projection density operator to the entangled state \( |\Psi\rangle_{\text{DLCZ}} = (\hat{S}_A^\dagger + \hat{S}_B^\dagger) / \sqrt{2} \) and calculate the success probability \( P_s \) of entanglement swapping of the entangled state and the heralding probability \( P_H \) for the third click as [33]

\[
F = \frac{\text{Tr} (\hat{\rho}_{\text{out}}^{(2)} |\Psi\rangle_{\text{DLCZ}} \langle \Psi|)}{\text{Tr} (\hat{\rho}_{\text{out}}^{(2)})}, \tag{27}
\]

\[
P_H = P_1 + P_2, \quad P_1 = P_2 = \frac{\text{Tr} (\hat{\rho}_{\text{out}}^{(2)})}{N}, \tag{28}
\]

\[
P_s = P_1 \times F_1 + P_2 \times F_2, \quad F_1 = F_2 = F, \tag{29}
\]

where \( P_{1,2} \) is the heralding probability of the single click from the mid-way detector \( c_{1,2} \) as shown in figure 5, and a trace (Tr) is taken over atomic degrees of freedom. The normalization factor \( N \) is calculated in equation (B.9) and is given by

\[
N = \frac{\eta_1^2 (1 - \eta_2)^2}{4} \eta_\text{eff} + \frac{\eta_1 \eta_2 (1 - \eta_1) (1 - \eta_2)}{2} \eta_\text{eff} \\
+ \frac{\eta_2^2 (1 - \eta_1)^2}{4} \eta_\text{eff}. \tag{30}
\]

We have used the following properties for the calculation of \( \hat{\rho}_{\text{out}}^{(2)} \) and \( N \),

\[
\int d\omega d\omega' d\omega' / f(\omega, \omega') \Phi(\omega) \Phi^*(\omega) = 1, \tag{31}
\]

where orthonormal relations in the mode functions are used, and

\[
\int d\omega d\omega' d\omega' / f(\omega', \omega') f^*(\omega', \omega) f(\omega, \omega) f^*(\omega, \omega) = \sum_j \lambda_j^2. \tag{32}
\]

Note that the single-mode spectral function for the Raman photon satisfies \( \int d\omega / \Phi(\omega) = 1 \).

The fidelity, heralding and success probabilities become

\[
F = 1 + \sum_j \lambda_j \int \phi_j(\omega)\phi_j^*(\omega) \Phi(\omega) \Phi^*(\omega) d\omega d\omega' \frac{\eta_1(2 - \eta_1)(1 + \sum_j \lambda_j^2)/2 + 2}{\eta_1(2 - \eta_1)(1 + \sum_j \lambda_j^2)/2 + 2}. \tag{33}
\]
from equation (37) and the success probability from
PH
PH
\begin{equation}
P_H = \frac{\eta_t \eta_i (2 - \eta_t) (1 + \sum \lambda_j^2) / 2 + 2 \eta_t}{(\sqrt{\eta_t} + 1/\sqrt{\eta_t})^2}.
\end{equation}
(34)

\begin{equation}
P_S = \frac{1 + \sum \lambda_j \int \phi_j(\omega_i) \phi_j^*(\omega_i') \Phi^*(\omega_i) \Phi(\omega_i') \, d\omega_i \, d\omega_i'}{(\sqrt{\eta_t} + 1/\sqrt{\eta_t})^2},
\end{equation}
(35)

where \( \frac{1 - \eta_t}{\eta_t} \approx 1 \) and \( \eta_t = \eta_i / \eta_t \).

The fidelity depends on a sum of square of Schmidt numbers in the denominator and the mode mismatch between the idler and Raman photons in the numerator. Let us assume that the Raman photon mode is engineered to be matched with the idler photon mode of the largest Schmidt number (\( \phi_i(\omega_i) \) in our case), which is required to have a larger fidelity (so is the success probability) compared to other modes. We may also compare the NRPD with the performance of PNRD in the midway detectors; then we have the fidelity, heralding and success probabilities,

\begin{equation}
F = \begin{cases}
1 + \lambda_1, & \text{NRPD} \\
\eta_t (2 - \eta_t) (1 + \sum \lambda_j^2) / 2 + 2 \eta_t, & \text{PRND}
\end{cases}
\end{equation}
(36)

\begin{equation}
\begin{cases}
\eta_t \eta_i (2 - \eta_t) (1 + \sum \lambda_j^2) / 2 + 2 \eta_t, & \text{NRPD} \\
\eta_t \eta_i (1 - \eta_t) (1 + \sum \lambda_j^2) + 2 \eta_t, & \text{PRND}
\end{cases}
\end{equation}
(37)

\begin{equation}
\begin{cases}
\eta_t (1 + \lambda_1), & \text{NRPD} \\
\eta_t (1 + \lambda_1) / (\sqrt{\eta_t} + 1/\sqrt{\eta_t})^2, & \text{PRND}
\end{cases}
\end{equation}
(38)

When the relative efficiency is made arbitrarily small, the fidelity approaches \( (1 + \lambda_1) / 2 \) for both types of detectors. It reaches 1 if a pure cascade-emission source is generated (von Neumann entropy \( F = 0 \) and \( \lambda_1 = 1 \)). When \( \eta_t = 1 \) with a pure source using NRPD with a perfect quantum efficiency, \( F = 2/3, P_H = 3/4, P_S = 1/2 \), which coincide with the results of [33] (with perfect quantum efficiency).

We discuss the frequency entanglement for various pulse widths and superradiant decay rates in section 3. We find that for shorter driving pulses and smaller superradiant decay rates, the cascade-emission source is less spectrally entangled. That means when \( \eta_t \) is fixed, a shorter driving pulse heralds a higher fidelity DLCZ entangled state.

In figure 6, we numerically calculate the entropy and plot out the fidelity from equation (36), the heralding probability from equation (37) and the success probability from equation (38) as a function of the relative efficiency \( \eta_t \). With a perfect detection efficiency \( (\eta_t = 1) \), we find that at a smaller \( \eta_t \), the less entangled source gives us a higher fidelity DLCZ entangled state but with a smaller success probability. Small generation probability for cascade emission \( (\eta_t < 1) \) reduces the error of NRPD from the interference of two telecom photons, but it reduces the successful entanglement swapping at the same time.

The optimal success probability occurs by using the same excitation efficiency for both cascade and Raman configurations. For PNRD, the fidelity is higher than NRPD, and the heralding probability is the same independent of the degree of frequency space entanglement. The success probabilities for both types of detectors are equal. The advantage of PNRD shows up in the fidelity of quantum swapping.

In figure 7, we show that the measures improve monotonically with the quantum efficiency \( (\eta = \eta_t) \) of the detector at the telecom wavelength, with \( \eta_t = 0.5 \). The success
ensembles (entangled pairs of DLCZ entangled states, and another two possible detection events of \( \hat{\Psi} \))

quantum-teleportation protocol (b), another two ensembles (\( \hat{\Psi} \))

used to generate two DLCZ entangled states at \( \hat{\Psi} \) by applying the phases \( \phi_1 \) and \( \phi_2 \) to sides (A, C) and (B, D) in figure 8(a), respectively, through single-bit operations \( \hat{\Psi} \). For cascade-emission-based quantum communications, the spectral effect of the cascade emission we implement here reduces the success rate for the generation of the PME state because of the frequency entanglement in the source where \( \lambda_1 < 1 \).

For an arbitrary quantum state transfer to long distance, the quantum-teleportation scheme may be used. Another two ensembles \( (I_1, I_2) \) are introduced \( \hat{\Psi} \), and the quantum state can be described by \( |\Psi\rangle = (d_0 S_{1i}^1 + d_1 S_{1i}^2) |0\rangle \) with \( |d_0|^2 + |d_1|^2 = 1 \).

The joint density matrix for quantum teleportation is

\[
\hat{\rho}_{\Omega,T} = (d_0 S_{1i}^1 + d_1 S_{1i}^2) |0\rangle \langle 0| (d_0 S_{1i}^1 + d_1 S_{1i}^2) + 4 \left[ \eta_2 (2 - \eta_2) (1 + \sum_{\alpha} \lambda_{\alpha}^2) + 4 \right].
\]

For \( \eta_2 \ll 1 \), \( \hat{\rho}_{\Omega,T} \) reaches the maximum of 1/2 when a pure source \( (\lambda_1 = 1) \) is used. Compared with the original DLCZ proposal \( \hat{\Psi} \), the success probability. The normalized density matrix for the PME state to enable the quantum cryptography and Bell inequality measurement by applying the phases \( \phi_1 \) and \( \phi_2 \) is

Figure 8. Effective PME projection (a) and quantum teleportation (b) in the DLCZ scheme. Four atomic ensembles \( (A, B, C, D) \) are used to generate two DLCZ entangled states at \( \hat{\Psi} \).

The PME state is projected probabilistically conditioned on four possible detection events of \( (D_i \) or \( D_j \)) in (a). In the quantum-teleportation protocol (b), another two ensembles \( (I_1, I_2) \) are used to prepare a quantum state that is teleported to the atomic ensembles \( B \) and \( D \) conditioned on four possible detection events of \( (D_i \) or \( D_j \)) and \( (D_i \) or \( D_j \)).

probabilities for both types of detectors are the same and again the advantage of PNRD shows up in the fidelity.

4.2. Effective PME state and quantum teleportation

In figure 8, we illustrate a scheme for probabilistic and effective PME state preparation and quantum teleportation. The term ‘polarization’ is used as an analogy \( \hat{\Psi} \) to the entangled photons in the polarization degree of freedom and note that what is actually prepared here is the entangled photons in path modes. Four ensembles \( (A, B, C, D) \) are used to generate two entangled pairs of DLCZ entangled states, and another two ensembles \( (I_1, I_2) \) are used to prepare a quantum state to be teleported.

With the conditional output density matrix from equation (B.16), we proceed to construct the PME state \( |\Psi\rangle_{\text{PME}} = \frac{1}{\sqrt{2}} (S_{1i}^A S_{1i}^D + S_{1i}^B S_{1i}^C)|0\rangle \) where \( (C, D) \) represents another parallel entanglement connection set-up, figure 8(a). This PME state is useful in entanglement-based communication schemes \( \hat{\Psi} \), and we will here calculate its success probability. The normalized density matrix for the AB system is from equation (26) \( \eta_2 \).

\[
\hat{\rho}_{\text{PME}} = \frac{1}{\sqrt{2}} (S_{1i}^A S_{1i}^D + S_{1i}^B S_{1i}^C)|0\rangle \langle 0| (S_{1i}^A S_{1i}^D + S_{1i}^B S_{1i}^C) + \lambda_1 (S_{1i}^A S_{1i}^D)|0\rangle \langle 0| S_{1i}^A + \lambda_1 (S_{1i}^B S_{1i}^C)|0\rangle \langle 0| S_{1i}^B),
\]

where the largest Schmidt number \( \lambda_1 \) of mode overlap is chosen and \( a \equiv \eta_2 (2 - \eta_2) (1 + \sum_{\alpha} \lambda_{\alpha}^2) \).

A parallel pair of entangled ensembles \( (C, D) \) is introduced, and the joint density operator is \( \hat{\rho}_{\text{PME}} = \hat{\rho}_{\text{PME}} \otimes \hat{\rho}_{\text{PME}} \).

The latter expression is developed mathematically in appendix B.3.

With the projection of the PME state, we have the post-measurement success probability (a click from each side; the side of (A or C) and (B or D)),

\[
P_{\text{PME}} = \langle \Psi | \hat{\rho}_{\text{PME}} \otimes \hat{\rho}_{\text{PME}} | \Psi \rangle = \frac{4(1 + \lambda_1^2)}{\left[ \eta_2 (2 - \eta_2) (1 + \sum_{\alpha} \lambda_{\alpha}^2) + 4 \right]^2}.
\]

For \( \eta_2 \ll 1 \), \( P_{\text{PME}} \) reaches the maximum of 1/2 when a pure source \( (\lambda_i = 1) \) is used. Compared with the original DLCZ proposal \( \hat{\Psi} \), the success probability.
The fidelity and heralding probabilities conditioned on the other three pairs of clicks are the same as $F_1$ and $P_1$, respectively, so the success probability is

$$P_{SQT} = \sum_i^4 P_i F_i = 4P_1 F_1,$$

$$= \frac{F^2}{(1 + \lambda_1)^2} [1 + (2\lambda_1^2 - 2)|d_0|^2|d_1|^2],$$

(43)

where $F$ is the fidelity of entanglement swapping for NRPD, equation (36). For PNRD, the success probability for quantum teleportation is unchanged.

The success probability for quantum teleportation depends on the probability amplitude of the quantum state and the fidelity $F$ of the entanglement swapping. In figure 9, for $\eta_s = 0.5$ and $\eta_t = 1$, we can see in the region $|d_0| \approx 0.3-0.9$, a higher success probability requires a less entangled cascade-emission source. Outside this region, it prefers a more entangled source. When a pure source is used ($\lambda_1 = 1$) and when we let $\eta_t \ll 1$, $\eta_t = 1$, we can achieve the maximum of the success probability $P_{SQT} = \frac{1}{3}$ when $F = 1$, which is also achieved in the traditional DLCZ scheme with perfect quantum efficiencies [33].

5. Discussions and conclusions

We have described probabilistic protocols for the DLCZ scheme implementing the cascade-emission source. We characterize the spectral properties of the cascade emission by the Schmidt mode analysis and investigate the fidelity and success probability of the protocols using photon resolving and non-resolving photon detectors. The success probability is independent of the detector type, but photon-number resolving detection improves the fidelity.

The performance of the protocol also depends on the ratio of efficiencies in generating the cascade and Raman photons. The success probability is optimized for equal efficiencies, while the fidelity is higher when the ratio is smaller than 1 for NRPD.

The frequency space entanglement of telecom photons produced in cascade emission deteriorates the performance of DLCZ protocols. The harmful effect can be diminished by using shorter pump pulses to generate the cascade emission. A state-dependent success probability of quantum teleportation was calculated, and in some cases, a more highly frequency-entangled cascade-emission source teleports more successfully. An improved performance could be achieved if the error source (vacuum part) were removed. This could be done by entanglement purification [26] at the stage of entanglement swapping and then using the purified source to teleport the quantum state.

The quantum efficiency of detectors has improved to above 60% in the infrared wavelength for avalanche photodiodes (APDs) and a maximum of 95% in the telecom wavelength for superconducting devices at a very low temperature (100 mK) [34]. We expect that our optimal performance in the modified DLCZ scheme can be achieved as shown in figure 7 where a fidelity $F \approx 0.9$. Our cascade-emission-based quantum communication scheme utilizes a telecom wavelength photon that has a minimal loss 0.2 dB km$^{-1}$ through fibre transmission. Compared with 2 dB km$^{-1}$ loss for the infrared bandwidth, the telecom photon has an attenuation length ten times longer which is about 22 km. In terms of the rate of direct single-photon transmission over continental distances (several hundreds of kilometres), it is overwhelmingly desirable to use telecom over the infrared bandwidth [34].

We note that an alternative method to generate telecom photons in atomic ensembles is frequency down-conversion [10]. Two cold and non-degenerate rubidium gas samples are used to correlate a stored atomic excitation and a telecom photon. The stored excitation is correlated with an infrared photon (idler) in one sample, and the idler is converted to a telecom wavelength photon in the other ensemble. Thus, a matter–light entanglement is created to serve as a basic element in entanglement connection of the DLCZ scheme with an advantageous telecommunication bandwidth. Similar to our cascade-emission scheme, frequency conversion also requires a phase-matching of the FWM condition in a diamond configuration of atomic levels [9, 35]. To implement it into our modified DLCZ scheme, an extra conversion efficiency needs to be taken into account. The efficiency has reached a maximum of 0.54 [10] and can be close to 1 if we use atoms with a larger optical depth (odp > 200) [11]. Therefore, this alternative method serves as well as our cascade-emission scheme but demands one more cold atomic ensemble which might cause difficulties when a large scale quantum repeater is considered.

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Appendix A. Hamiltonian and Schrödinger equation

In this appendix, we derive the Hamiltonian for the cascade emission (signal–idler) from a four-level atomic ensemble.

\[ H = ... \]
We use Schrödinger’s equation to study the correlated two-photon state from a two-photon laser excitation. Consider an ensemble of $N$ four-level atoms interacting with two classical fields and spontaneously emitted signal and idler photons as shown in figure 1. These identical atoms distribute randomly with a uniform density. Using dipole approximation of light–matter interactions, $-\hat{H} \hat{E}$, where $\hat{E}$ is the classical or quantum electric field, and rotating wave approximation (RWA) [29], the Hamiltonian in the interaction picture is

$$V_I(t) = -\hbar \Delta_1 \sum_{\mu=1}^N \langle 1 \rangle_\mu \langle 1 \rangle - \hbar \Delta_2 \sum_{\mu=1}^N \langle 2 \rangle_\mu \langle 2 \rangle$$

$$-\hbar \{ \Omega_\mu \hat{P}_{\mu}^\dagger \hat{I}_{\mu} + \Omega_\mu \hat{P}_{2\mu} \hat{I}_{2\mu} + \text{h.c.} \}$$

$$-\hbar \left\{ \sum_{k_i \lambda_i} g_{k_i}(\epsilon_{k_i, \lambda_i} \cdot \hat{d}_i^\dagger) \hat{a}_{k_i, \lambda_i} \hat{b}_i e^{-i(\omega_{k_i} - \omega_{k_i})t} + \text{c.c.} \right\}$$

$$+ \sum_{k_i \lambda_i} g_{k_i}(\epsilon_{k_i, \lambda_i} \cdot \hat{d}_i^\dagger) \hat{a}_{k_i, \lambda_i} \hat{b}_i e^{-i(\omega_{k_i} - \omega_{k_i})t} - \text{h.c.} \right\},$$

where the collective dipole operators and positive frequency parts of the electric fields are defined as

$$\hat{P}_{\mu}^\dagger \equiv \sum_{\mu=1}^N \langle 1 \rangle_\mu \langle 1 \rangle e^{\hat{P}_{\mu}^\dagger} \hat{r}_\mu,$$

$$\hat{S}_{\mu}^\dagger \equiv \sum_{\mu=1}^N \langle 2 \rangle_\mu \langle 0 \rangle e^{\hat{S}_{\mu}^\dagger} \hat{r}_\mu,$$

$$\hat{E}_{\mu}^+ (\vec{r}_1, t_1) = \sum_{k_i \lambda_i} \sqrt{\frac{\hbar \omega_{k_i}}{2eV}} \hat{a}_{k_i, \lambda_i} \epsilon_{k_i, \lambda_i} e^{i\vec{k}_i \cdot \vec{r}_1 - i\omega_{k_i} t_1},$$

$$\hat{E}_{\mu}^- (\vec{r}_2, t_2) = \sum_{k_i \lambda_i} \sqrt{\frac{\hbar \omega_{k_i}}{2eV}} \hat{a}_{k_i, \lambda_i} \epsilon_{k_i, \lambda_i} e^{i\vec{k}_i \cdot \vec{r}_2 - i\omega_{k_i} t_2}.$$
\[
\sum_{\mu,\nu} e^{i(k_i-\hat{q}_i-\hat{q}_s)} r_{\mu} \text{ or } \sum_{\nu} e^{-i(k_i-\hat{q}_i-\hat{q}_s)} r_{\nu}, \text{ the coupling from the modes } \hat{q}_i \text{ and } \hat{q}_s \text{ is significant only when } |\hat{q}_i| = |\hat{q}_s|, \text{ so we finally have}
\]
\[
\bar{\mathcal{C}}_{s,q_i} = g_s^2 (e^* \cdot \hat{d}_s) \sum_{\mu} e^{-i(k_i+\hat{q}_i) \cdot \hat{r}_\mu} e^{i(\omega_\mu-\omega_{a2}+\Delta_2)\gamma B_\mu}
\]
\[
- \frac{\Gamma_3}{2} (N\mu + 1) C_{s,q_i} + i\delta_{\alpha} C_{s,q_i}.
\]
(A.13)

The collective decay rate is [27, 36]
\[
\frac{\Gamma_3}{2} (N\mu + 1) \equiv \frac{\Gamma_3}{2(2\pi)} \int d\Omega [1 - (\hat{k}_i \cdot \hat{d}_i)^2]
\]
\[
\times \frac{1}{N} \sum_{\mu,\nu} e^{i(k_i-\hat{q}_s) \cdot (\hat{r}_\mu-\hat{r}_\nu)},
\]
(A.14)

and the collective frequency shift expressed in terms of the continuous integral over a frequency space is [27, 37, 38, 28]
\[
\delta \omega_i \parallel = \int_0^\infty \frac{\Gamma_3}{2\pi} \cdot \left[ P.V. (\omega_i - \omega_s)^{-1} + P.V. (\omega_i + \omega_s)^{-1} \right] N\mu(k_i),
\]
(A.15)

which is derived after we renormalize the Lamb shift and consider the non-RWA terms in the original Hamiltonian. Non-RWA terms contribute to the term proportional to P.V. \((\omega_i + \omega_s)^{-1}\).

The geometrical constant \(\bar{\mu}\) for a cylindrical ensemble (of height \(h\) and radius \(a\)) is
\[
\bar{\mu}(k_i) = \frac{6(N-1)}{NA^2H^2} \int_{-1}^1 dx (1+x^2)
\]
\[
\times \sin^2 \left[ \frac{1}{2} H(1-x) \right] J_1^2 (A(1-x^2)^{1/2}),
\]
(A.17)

where \(H = k_s h\) and \(A = k_s a\) are the dimensionless length scales, and circular polarizations are considered [36]. \(J_1\) is the Bessel function of the first kind.

### Appendix B. Multimode description of the correlated two-photon state

In this appendix, we review a general model for quantum detection efficiency [39] for multimode analysis in various quantum communication schemes. Based on this detection model with the spectral description of the correlated two-photon state, we derive the effective density matrix conditioning on the detection events of entanglement swapping, PME state projection and quantum teleportation.

#### B.1. Quantum efficiency of the detector

To account for the quantum efficiency of the detector and the effect of its own spectrum filtering, we introduce an extra B.S. with a transmissivity \(\eta(\omega, \omega_0)\) [39] before the detection event. \(\eta\) models the quantum efficiency of the detectors in the microscopic level (response at the frequency \(\omega_0\)) and the macroscopic level (time-integrated detection). Considering one example of conditioning on the single click of the detector, the output density operator becomes

\[
\rho_{\text{out}} = \int_{-\infty}^{\infty} d\omega_0 \hat{\Pi}_1 \text{Tr}_{\text{ref}} \left[ \hat{U}_{\text{BS}} \rho_{\text{in}} \hat{U}_{\text{BS}}^\dagger \right] \hat{\Pi}_1,
\]
(B.1)

\[
\hat{\Pi}_1 \equiv \int_{-\infty}^{\infty} d\omega |\omega\rangle \langle \omega|,
\]
(B.2)

\[
\hat{U}_{\text{BS}} = \left( \begin{array}{cc} \sqrt{1 - \eta} & \sqrt{\eta} \\ \sqrt{\eta} & -\sqrt{1 - \eta} \end{array} \right),
\]
(B.3)

where \(\text{Tr}_{\text{ref}}\) is the trace over the reflected modes \(m_3^+\), and the flat spectrum projection operator \(\hat{\Pi}_1\) (only the photon number is projected and no frequency resolution) is considered in the measurement process [23]. In figure B1, \(m_1^+\) is the incoming photon operator before the detection, \(m_3^+\) is the reflected mode and \(m_4^+\) is now the detection mode with a modelling of spectral quantum efficiency and an effective quantum efficiency is defined as

\[
\int_{-\infty}^{\infty} \eta(\omega, \omega_0) d\omega_0 = \eta_{\text{eff}}(\omega).
\]
(B.4)

#### B.2. Multimode description of entanglement swapping

From equation (25), we use the single mode \(\Phi(\omega)\) for the Raman photon and a multimode description \(f(\omega_1, \omega_2)\) for cascade photons and rewrite the effective state. Note that a symmetric set-up is considered, so the mode description is the same for both sides \(A\) and \(B\) in the scheme of entanglement swapping:

\[
|\Psi\rangle_{\text{eff}} = \eta_1 (1 - \eta_2) \times \int f(\omega_1, \omega_2) \hat{a}_1^{A\dagger} (\omega_1) \hat{a}_2^{A\dagger} (\omega_2) d\omega_1 d\omega_2
\]
\[
\times \int f(\omega'_1, \omega'_2) \hat{a}_1^{B\dagger} (\omega'_1) \hat{a}_2^{B\dagger} (\omega'_2) d\omega'_1 d\omega'_2,
\]
\[
+ \eta_2 (1 - \eta_1) \int \Phi(\omega) d\omega_1 \hat{a}_1^{A\dagger} (\omega_1) \hat{a}_2^{A\dagger} (\omega_2)
\]
\[
\times \int \Phi(\omega') d\omega_2 \hat{a}_1^{B\dagger} (\omega') \hat{a}_2^{B\dagger} (\omega') + \sqrt{\eta_1 (1 - \eta_1)}
\]
\[
\times \sqrt{\eta_2 (1 - \eta_2)} \int f(\omega_1, \omega_2) d\omega_1 d\omega_2
\]
\[ \hat{\rho}_{\text{out}} = \int_{-\infty}^{\infty} \rho_{\text{in}}(\omega) d\omega \sum_{\text{channels}} \left( \hat{\rho}_{\text{out}} \right)_{\text{channel}} \]

The normalization factor is derived by tracing over the atomic degree of freedom:

\[ \text{Tr}(\hat{\rho}_{\text{out}}) = \frac{\eta_1^2(1 - \eta_2)^2}{4} \int d\omega_1 d\omega_1' \eta_{\text{eff}}(\omega_1) \eta_{\text{eff}}(\omega_1') \]

where \( \hat{M}_{4} \equiv \hat{U}_{\text{out}}^{-1} \hat{U}_{\text{in}} \hat{M}_{4} \hat{U}_{\text{out}}^{-1} \).

Next we interfere telecom photons with B.S. that \( \hat{c}_1 = \hat{c}_1^+ + \hat{c}_2^+ + \hat{c}_3^+ + \hat{c}_4^+ \), and again a quantum efficiency \( \eta(\omega, \omega_0) \) for the telecom photon is introduced. Use \( \hat{c}_1^+ = \sqrt{1 - \eta_1^2} + \sqrt{\eta_1^2} \) and trace over the reflected mode \( \hat{c}_1^+ \) conditioning on the click of \( \hat{c}_1^+ \) from NRPD [33]. The effective density matrix becomes

\[ \hat{\rho}_{\text{out}}^{(2)}(\omega_0) = \int_{-\infty}^{\infty} \rho_{\text{in}}(\omega) \left\{ \sum_{\text{channels}} \left( \hat{\rho}_{\text{out}}^{(2)} \right)_{\text{channel}} (\omega_0) \right\} \]

where \( \eta_{\text{eff}}(\omega) \) is introduced after the integration of \( \omega_0 \), and we denote it as an effective quantum efficiency for the idler field \( \omega_s \) or the Raman photon at the frequency \( \omega \) (wavelength 780 nm for the D2 line of the Rb atom). \( \hat{\rho}_{\text{out}}^{(2)} \) includes the terms that would not survive after the interference of telecom photons in the middle B.S. (conditioning on a single click of the detector). They involve operators like \( \hat{a}_i^{+A} \hat{c}_1^+ (0) \hat{a}_i^{+B} \hat{S}_B \), \( \hat{a}^{+A} \hat{c}_4^+ (0) \hat{S}_B \), and \( \hat{a}_3^{+A} \hat{c}_1^+ (0) \hat{S}_B \).

The normalization factor is derived by tracing over the atomic degree of freedom:

\[ \text{Tr}(\hat{\rho}_{\text{out}}^{(2)}) = \frac{\eta_1^2(1 - \eta_2)^2}{4} \int d\omega_1 d\omega_1' \eta_{\text{eff}}(\omega_1) \eta_{\text{eff}}(\omega_1') \]

which will be put back when we calculate the heralding and success probabilities.
the interchange of variables in integration: 

\[ \hat{\sigma}(\omega) \hat{\sigma}^\dagger(\omega) \sigma(\omega, \omega) \sigma(\omega, \omega) \]

where the trace over the two-photon states requires the commutation relation of photon operators:

\[ \text{Tr}[\hat{m}_4(\omega_s)\hat{m}_4(\omega_s)] = \langle 0|\hat{m}_4(\omega_s)\hat{m}_4(\omega_s)|0\rangle \]

The above equation is the general formulation for the unnormalized density matrix conditioning on three clicks of NRPDs. We have included the spectral quantum efficiency of the detector either for the near-infrared (\( \eta_{\text{eff}} \)) or telecom wavelength (\( \eta_t \equiv \int_{-\infty}^{\infty} \eta(\omega, \omega) \, d\omega \)).

To proceed, we assume a flat and finite spectrum response of the detector for the range \( \Omega - \Delta, \Omega + \Delta \) centred at \( \Omega \) (near-infrared or telecom) and \( \omega \in [\omega_0 - \delta, \omega_0 + \delta] \) [39]. The widths 2\( \Delta \) and 2\( \delta \) are large enough compared to our source bandwidth, so these detection events do not give us any information of the spectrum for our source. A perfect efficiency also means no photon loss during detection. Note that the integral involves the multiplication of two telecom photon efficiencies \( \int_{-\infty}^{\infty} \eta(\omega, \omega_0) \eta(\omega', \omega_0) \, d\omega = \eta_t^2(\omega) \) that is valid if the source bandwidth is smaller than the detector's.

After the integration of \( \omega_0 \), we have

\[ \hat{\rho}_{\text{out}}^{(2)} = \frac{\eta_t^2(1 - \eta_2)^2}{8} \eta_{\text{eff}}^2 \int \! d\omega_0 \, d\omega_i \int \! d\omega_0 \, d\omega_i \]

with a brief notation for the spectrum \( f(s, i) \equiv f(\omega_s, \omega_i) \) and quantum efficiency \( \eta(\omega) \equiv \eta(\omega, \omega_0) \). This quantum efficiency refers to the telecom photon. We proceed to trace over the detected modes and the density matrix can be simplified by the interchange of variables in integration:
\[ \times \int \frac{d\omega}{2 \pi} \Phi(\omega)^2 \langle \hat{S}_B^\dagger(0)|0\rangle \langle 0|\hat{S}_A + \hat{S}_D^\dagger(0)|0\rangle \langle 0|\hat{S}_B \rangle \\
+ \iint f(\omega_1, \omega_2) d\omega_1 d\omega_2 \int f^*(\omega_1, \omega_2) d\omega_1 d\omega_2 \cdot (\hat{S}_B^\dagger(0)|0\rangle \langle 0|\hat{S}_A + \hat{S}_D^\dagger(0)|0\rangle \langle 0|\hat{S}_B \rangle) \right) \right] \quad (B.16) \]

B.3. Density matrix of PME projection and quantum teleportation

In section 4.2, we have the normalized density operator \( \hat{\rho}_{\text{out},n} \) of the DLCZ entangled state through entanglement swapping. With another pair of DLCZ entangled state, \( \hat{\rho}_{\text{out},CD} \), the joint density operator for these two pairs constructs the PME state projection and is interpreted as

\[ \hat{\rho}_{\text{out},n(AB)} \otimes \hat{\rho}_{\text{out},n(CD)} = \frac{1}{(a+b)^2} \times \left[ \frac{1}{a^2}|0\rangle \langle 0| + \frac{ab}{2} |0\rangle \langle 0| (\hat{S}_C^\dagger(0)|0\rangle \langle 0|\hat{S}_C + \hat{S}_D^\dagger(0)|0\rangle \langle 0|\hat{S}_D \rangle \\
+ \frac{b^2}{4} (\hat{S}_C^\dagger(0)|0\rangle \langle 0|\hat{S}_C + \hat{S}_D^\dagger(0)|0\rangle \langle 0|\hat{S}_D \rangle \\
+ \lambda_1 (\hat{S}_C^\dagger(0)|0\rangle \langle 0|\hat{S}_C + \hat{S}_D^\dagger(0)|0\rangle \langle 0|\hat{S}_D \rangle) \\
\otimes (\hat{S}_B^\dagger(0)|0\rangle \langle 0|\hat{S}_B + \hat{S}_A^\dagger(0)|0\rangle \langle 0|\hat{S}_A \rangle \\
+ \lambda_1 (\hat{S}_B^\dagger(0)|0\rangle \langle 0|\hat{S}_B + \hat{S}_A^\dagger(0)|0\rangle \langle 0|\hat{S}_A \rangle) \right), \quad (B.17) \]

which is used to calculate the success probability post-measurement (a click from each side, the side of \( A \) or \( C \) and \( B \) or \( D \)). \( a = \eta_2(2-\eta_1)(1 + \sum_{i} \lambda_i^2), b = 4, \) where \( \eta_1 = \eta_{12}/\eta_2, \eta = \eta_1 + \lambda_j \) is the Schmidt number that is used to decompose the two-photon source from the cascade transition.

In the DLCZ protocol, quantum teleportation uses the similar set-up in PME projection and combines with the desired teleported state, \( \ket{\Phi} = (d_0 \hat{S}_B^\dagger + d_1 \hat{S}_C^\dagger)|0\rangle \), which is represented by two other atomic ensembles \( I_1 \) and \( I_2 \). The requirement of normalization of the state is \( |d_0|^2 + |d_1|^2 = 1 \), and the density operator of quantum teleportation is \( \hat{\rho}_{\text{QT}} = \langle \Phi \rangle \otimes \hat{\rho}_{\text{out},n(AB)} \otimes \hat{\rho}_{\text{out},n(CD)}. \) Conditioning on the clicks of \( \hat{D}_B \) and \( \hat{D}_C \), the effective density matrix for quantum teleportation is (using \( \hat{S}_1 = (\hat{D}_B + \hat{D}_C)/\sqrt{2}, \hat{S}_2 = (\hat{D}_B + \hat{D}_C)/\sqrt{2} \) for the effect of B.S.)

\[ \hat{\rho}_{\text{QT,eff}} = \left[ \frac{|d_0|^2}{2} (\hat{D}_B^\dagger(0)|0\rangle \langle 0|\hat{D}_B + d_1^2 (\hat{D}_C^\dagger(0)|0\rangle \langle 0|\hat{D}_C) \right] \otimes \frac{1}{(a+b)^2} \times \left[ a^2|0\rangle \langle 0| + \frac{ab}{2} |0\rangle \langle 0| (\hat{D}_C^\dagger(0)|0\rangle \langle 0|\hat{D}_C + \hat{S}_D^\dagger(0)|0\rangle \langle 0|\hat{S}_D \rangle \\
+ \lambda_1 (\hat{S}_B^\dagger(0)|0\rangle \langle 0|\hat{S}_B + \hat{S}_A^\dagger(0)|0\rangle \langle 0|\hat{S}_A \rangle) \right] \left[ \hat{D}_B^\dagger(0)|0\rangle \langle 0|\hat{D}_B + \hat{S}_D^\dagger(0)|0\rangle \langle 0|\hat{S}_D \rangle + \lambda_1 (\hat{S}_B^\dagger(0)|0\rangle \langle 0|\hat{S}_B + \hat{S}_A^\dagger(0)|0\rangle \langle 0|\hat{S}_A \rangle) \right] \right) \quad (B.18) \]

which is used to calculate the success probability for the teleported state.

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