Quantum tomography of entangled spin-multi-photon states.

Dan Cogan, Giora Peniakov, Oded Kenneth, Yaroslav Don, and David Gershoni

The Physics Department and the Solid State Institute, Technion–Israel Institute of Technology, 3200003 Haifa, Israel

We present a novel method for quantum tomography of multi-qubit states. We apply the method to spin-multi-photon states, which we produce by periodic excitation of a semiconductor quantum-dot-confined spin every 1/4 of its coherent precession period. These timed excitations lead to the deterministic generation of strings of entangled photons in a cluster state. We show that our method can be used for characterizing the periodic process map, which produces the photonic cluster. From the measured process map, we quantify the robustness of the entanglement in the cluster. The 3-fold enhanced generation rate over previous demonstrations reduces the spin decoherence between the pulses and thereby increases the entanglement.

I. INTRODUCTION

Measurement-based quantum protocols are very promising for quantum computation in general [1–3] and for quantum communication in particular [4–7]. The use of multi-partite entangled state named graph-states [8, 9], enables quantum computation by single qubit measurements and rapid classical feedforward, depending on the measurement outcome [2]. For quantum communication, graph-states of photons are particularly attractive [10–13], since they provide redundancy against photon loss, and compensation for the finite efficiency of quantum gates. Moreover, since the quantum information is contained in the graph state they eliminate the need to communicate within the coherence time of the local nodes [7]. Graph-states are therefore considered for efficient distribution of entanglement between remote nodes [14] as well as for quantum repeaters [5, 6]. Developing devices capable of deterministically producing high-quality photonic graph states at a fast rate is, therefore, a scientific and technological challenge of utmost importance [15].

The technological quest for generating photonic graph states which are required for building scalable quantum network architectures, led to new schemes. Of particular importance and relevance to this work is the Lindner and Rudolph proposal [16] for generating one-dimensional cluster state of entangled photons using semiconductor quantum dots (QDs). The scheme uses a single confined electronic spin in a coherent superposition of its two eigenstates. The spin precesses in a magnetic field while driven by a temporal sequence of resonant laser pulses. Upon excitation of the QD spin, a single photon is deterministically emitted and the photon polarization is entangled with the polarization of the QD spin. This timed excitation repeats itself indefinitely, thus generating a long 1D-cluster of entangled photons.

Schwartz and coworkers demonstrated the first proof-of-concept realization of this proposal in 2016 [17]. They showed that the entanglement robustness of the 1D-photonic string is mainly determined by the ratio between the photon radiative time and the spin-precession time and to a lesser extend also by the ratio between the later and the confined spin coherence time [17].

In Ref [17], the entangler was the spin of the dark exciton (DE). The short-range electron-hole interaction removes the degeneracy of the DE even in the absence of external field, therefore a coherent superposition of the DE eigenstates naturally precesses. Due to the limited temporal resolution of the silicon avalanche photodetectors which Schwartz et al used, the spin was re-excited every 3/4 of its precession period. In this work, we use instead superconducting single photon detectors with an order of magnitude better temporal resolution. Therefore we are able to drive the system every 1/4 of the DE precession period. This leads to photon generation rate which is three fold faster than previously demonstrated.

We develop a novel experimental and theoretical method for characterizing the improved cluster state and the spin - multi-photon quantum states that we generate. Our tomographic method differs from the traditional method [18] in the sense that it enables to measure the spin that remains in the QD after projecting all the emitted photons. The method uses time resolved spin-multi-photon correlations for measuring the quantum state, and for characterizing the periodically used process map which generates the photonic cluster.

We use a novel gradient descent method to find the process map which best fits the data in the sense of having maximum likelihood. Our gradient descent method differs from the standard one in the fact that we define the gradient relative to a specific non-euclidean metric which is adapted to the geometry of the set of physical (completely positive) process maps. This approach is very different from known algorithms such as projected gradient descent [19, 20].

In the following, we demonstrate our tomographic technique by characterizing the enhanced gigahertz rate generated cluster state. We show that as a result of the time reduction between the sequential excitations, the effect of the DE spin decoherence [21] is reduced, and the robustness of the entanglement in the cluster state increases, persisting for 6 consecutive photons.
The tomographic method and our experimental results are described below.

II. CLUSTER-STATE GENERATION - METHOD

In the heart of our device is a semiconductor QD. The QD contains a confined electronic-spin, which serves as the entangler qubit [16, 23, 24]. We define the sample growth direction, which is also the QD’s shortest dimension (about 3nm) as the quantization z-axis. The QD is embedded in a planar microcavity, formed by 2 Bragg-reflecting mirrors (Fig. 1a), facilitating efficient light-harvesting by an objective placed above the QD.

The DE is an electron-hole pair with parallel spins [25–27], having two total angular momentum states of ±2 as projected on the QD z-axis. Since the DE optical activity is weak [28], it has long life and coherence time [21, 26]. Upon optical excitation, the DE (|⇑⇑⟩) is excited to form a biexciton (BIE) (|↑↑↓↓⟩). The BIE is formed by pairs of electrons in their first conduction-subband level and 2 heavy holes with parallel spins one in the first and one in the second valence-subband levels. Fig. 1b and Fig. 1c schematically describe the DE-BIE energy level structure, and the selection rules for optical transitions between these levels, respectively [25, 29, 30].

Each laser pulse (in red) excites the QD confined DE to its corresponding BIE state. The BIE decays to the DE* level within about 370ps by radiative recombination in which a single photon is emitted (marked in pink). The DE* then decays to the ground DE state by about 100µeV spin-preserving acoustical-phonon relaxation. The DE* then decays to the ground DE state by about 6µeV spin-preserving acoustical-phonon relaxation. The DE and BIE act as spin qubits. Angular momentum conservation during the optical transitions between these eigenstates precesses with a period much smaller than the spectral width of our laser radiative width of the BIE optical transition (between these eigenstates are about 100µeV). Therefore the excitation and photon emission act as a 2-qubit entangling (CNOT) gate between the spin and the photon.

The method for generating the cluster state is described in Fig. 1d. The confined DE is resonantly excited repeatedly by a laser pulse to its corresponding BIE. The BIE decays radiatively by emitting a photon. The excitation and photon emission act as a two-qubit CNOT gate which entangles the emitted photon polarization qubit and the spin qubit, thus adding a photon to the growing photonic cluster. The excitation pulses are timed such that between the pulses the DE-spin precesses quarter of its precession period. This temporal precession can be ideally described as a unitary Hadamard gate acting on the spin qubit only. The combination of the CNOT 2-qubit gate and the Hadamard 1-qubit gate forms the basic cycle of the protocol which when repeated periodically generates the entangled spin + photons cluster state [16].

For the experimental realization of the cluster protocol and the characterization of the generated state, we use the experimental setup described in Fig. 1e. The QD is first optically-emptied from carriers, making it ready for initialization. The first 7-ns-long optical pulse (Blue downward arrow) depletes the QD from charges and the remaining DE [22]. We then write the DE spin state using horizontally polarized 12-ps optical π-pulse to the DE* state. This is possible due to small mixing between the bright exciton (BE) and the DE [27]. The pulsed polarization defines the DE* initial spin state. The DE* then relaxes to its ground DE state, making the QD ready for implementing the cluster protocol. A sequence of resonantly tuned linearly polarized π-area laser pulses is then applied to the QD. Each pulse results in the emission of a photon from the QD’s BIE-DE optical transition. During the last emission, the BIE spin evolution can be conveniently used as a resource for the DE spin tomography.

To characterize the generated multi-qubit quantum state, we project the polarization of the detected photons on 6 different polarization states using liquid-crystal-variable-retarders (LCVRs) and polarizing-beam-splitters (PBSs). We then use highly efficient transmission gratings to spectrally filter the emitted photons from the laser light. The photons are eventually detected by 6 efficient (>80%) fast single-photon superconducting detectors with temporal resolution of about 30ps.

III. CLUSTER-STATE CHARACTERIZATION

The cluster state entanglement robustness is characterized using three cycles of the repeated protocol. The characterization is done by correlating one, two, and three detected photon events. In all cases the last de-
Figure 1.  a) Schematic description of the QD-based device. DBR- distributed Bragg reflector. b) The energy levels of the DE - DE* - BIE system and the transitions between these levels. Each laser pulse (red upward arrow) excites the DE and photogenerates a BIE. The BIE then decays to an excited DE* by emitting a single-photon (downward pink arrow). The DE* then decays to its ground level by emitting acoustical phonon (curly downward arrow). c) The BIE-DE*-DE energy levels and the polarization-selection-rules for the optical transitions. Here +X (-X) describes horizontally (vertically) rectilinearly polarized optical transition. The short-range electron-hole exchange interaction removes the degeneracy between both the DE and the BIE two eigenstates. A coherent superposition of the DE (BIE) eigenstates precesses with a period marked by $T_{DE}$ ($T_{BIE}$). d) The cluster state repeating protocol containing Hadamard gates acting on the QD confined spin followed by a CNOT-two qubit gates, which entangles the spin polarization with the polarization of an emitted photons. e) The experimental setup, pulse sequence, photon emissions, and detection for realizing and characterizing the cluster state. Blue downward wide-arrow is an optical-depletion 7ns long pulse [22], which depletes the QD from carriers. Upward arrows are 12 ps long π-area laser pulses, while the downward pink exponential decays represent the emitted photons. We project the photons' polarization using liquid-crystal-variable-retarders (LCVRs) and polarizing-beam-splitters (PBS) while looking for three consecutive photon detections.

tected photon is used for the tomography of the DE-spin. We use two different orthogonal, +X and +Y linearly polarized excitation pulses respectively [31]. The +X-polarized laser pulse promotes the DE state to a similar superposition of BIE states, while +Y excitation introduces a $\pi/2$ phase shift to the superposition [31]. In addition, we utilize the BIE state evolution during its radiative decay back to the DE* to measure the degree-of-circular-polarization ($D_{cp}$) of the emission as a function of time:

$$D_{cp}(t) = \frac{P_{+Z}(t) - P_{-Z}(t)}{P_{+Z}(t) + P_{-Z}(t)},$$

(2)

where $P_j$ represents the detected photon polarization-projection on the $j$ basis. By fitting the measured $D_{cp}(t)$ to a central-spin-evolution-model that we recently developed for QD confined charge carriers [31], we accurately extract the DE spin state in the time of its excitation.

As we implement the protocol, we perform full tomographic measurements of the growing quantum state. First, we measure the initialized DE state. Then we apply one cycle of the protocol and measure the resulting spin+1photon state. Finally, we apply a second cycle of the protocol and measure the spin+2photons state.

In Fig 2 we present a small set of the measurements used to deduce the spin, spin+1photon, spin+2photon quantum states and the process map of the periodic cycle of the cluster protocol. The first row shows $D_{cp}$ measurements characterizing the initialized spin, the second row shows the $D_{cp}$ of the correlated spin+1photon state after the photon was projected on a +Y polarization basis, and the third row shows the $D_{cp}$ of the correlated spin+2photons state after both photons were projected on +Y polarization basis. Each row differs from the row above it by applying one additional cycle of the protocol. In all measurements shown in the Fig. 2, the DE was initialized to -X state. In each row, the left-panel displays time resolved PL, the center-panel (right-panel) displays time resolved $D_{cp}$ measured after +X (+Y) polarized excitation of the final spin. By fitting the $D_{cp}$
correlation measurements, we extract the following spin, spin+1 photon, and spin+2 photon polarization density matrix elements, respectively:

\[
[S_X, S_Y, S_Z] = [-0.73, 0.05, 0.06]
\]

\[
[P_Y(1)^{T} S_X, P_Y(1)^{T} S_Y, P_Y(1)^{T} S_Z] = [0.01, 0.16, -0.59]
\]

\[
[P_Y(1)^{T} P_Y(2)^{T} S_X, P_Y(1)^{T} P_Y(2)^{T} S_Y, P_Y(1)^{T} P_Y(2)^{T} S_Z] = [0.03, -0.49, -0.08]
\]

where \(P_Y^{(i)}\) represents the polarization projection of the \(i\)th-photon in the string, on the \(j\) polarization base, and \(S_j\) is the DE-spin polarization, projected on the \(j\) base. The typical measurement uncertainties are about 0.01, 0.02, and 0.04 for the spin, spin+1 photon, and spin+2 photon polarization density matrix elements, respectively.

For a perfect initialization and application of the process we expect these polarization elements to be \([-1,0,0]\), \([0,0,-1]\), \([0,-1,0]\) respectively. Here, the DE spin is initialized to the \(-X\) state with polarization degree of 0.73, due to the limited efficiency of the depleting pulse [22].

After each cycle of the protocol, the measured \(D_{cp}(t)\) is reduced by approximately 20%, indicating an exponential decay as expected [17, 32].

The measured correlations are used to infer directly the spin+1 photon 2qubit state, obtained by applying one cycle of the protocol, the spin+2 photon state obtained by applying two cycles of the protocol, and a full process tomography of the periodic cycle of the cluster protocol.

To measure the spin+1 photon density matrix (displayed in Fig. 3a), we use a set of 12 DE-photon \(D_{cp}(t)\) correlation measurements. Two of those measurements are displayed in Fig. 2e and Fig. 2h. The measured density matrix has fidelity of \(F = 0.77 \pm 0.04\) with the maximally entangled Bell state. To measure the spin+2 photons 3-qubit density matrix (displayed in Fig. 3b) we use a set of 72 \(D_{cp}(t)\) DE-2-photon correlation measurements. Two of those measurements are displayed in Fig. 2f and 2i. The measured spin+2 photons density matrix has fidelity of 0.68 \(\pm 0.07\) with the maximally entangled 3-qubit state.

We produce the cluster state by repeatedly applying the same process, as shown in the protocol of Fig. 1d. As a result, one can fully characterize the cluster state for any number of qubits if the single-cycle process-map is known [17]. Ideally, the process-map contains a CNOT and a Hadamard gate. It maps the 2×2 spin qubit density matrix into a 4×4 density matrix representing the entangled spin-photon state. The process map \(\Phi\) can be fully described by a 4×16 positive and trace-preserving map with 64 real matrix elements.

We use the convention \(\Phi(\hat{\rho}_{DE}) = \sum_{\alpha,\beta,\gamma} \Phi_{\alpha,\beta,\gamma} \hat{\rho}_{DE} \hat{\sigma}_{\alpha,\beta,\gamma}\),

where \(\hat{\rho}_{DE} = \sum_{\gamma} \rho_{\gamma}^{DE} \hat{\sigma}_{\gamma}\), is the density matrix that describes the input DE state and \(\Phi(\hat{\rho}_{DE})\) describes the DE+1 photon state after one application of the process to the input DE state. The sums are taken over \(\alpha,\beta,\gamma = X, Y, Z\), where \(\hat{\sigma}_0\) is the identity matrix and \(\hat{\sigma}_X, \hat{\sigma}_Y, \hat{\sigma}_Z\) are the corresponding Pauli matrices. The 64 real parameters \(\Phi_{\alpha,\beta,\gamma}\) thus fully specify \(\Phi\).

Fig. 3c shows the results of the full-tomographic measurements of the process map. For acquiring these measurements, we initialize the DE-spin-state to six different states from three orthogonal bases [27]. For each of those 6 states we use 2 \(D_{cp}(t)\) single-photon measurements like the measurements displayed in Fig. 2d and Fig. 2g for tomography. Then we apply one cycle of the protocol for each initialization and measure the resulting spin+1 photon states by projecting the first photon on different orthogonal polarization bases [18] and correlating it with the \(D_{cp}(t)\) of the second photon. For characterizing each of those 6 spin+1 photon states, we use 12 \(D_{cp}(t)\) 2-photon correlations measurements like the ones presented in Fig. 2e and Fig. 2h.

To obtain the physical process map that best fits our measured results, we use a specifically developed edge-
Figure 3.  a) The measured DE+1photon density matrix. b) The measured DE+2photons density matrix. In a) and b), the colored bars represent measured density matrix elements, while the empty bars represent the ideal maximally entangled 2- and 3-qubit states. The fidelity of the measured density matrix to the ideal one is $F = 0.77 \pm 0.04$ in a) and $F = 0.68 \pm 0.07$ in b). In both cases, the DE spin is initialized to the $|\text{DE}^{-}\rangle$ state with measured fidelity of $F = 0.86 \pm 0.02$ and then applying once or twice the process. The process map $\Phi$, which describes the system’s evolution in each cycle of the protocol is given by $\Phi(\hat{\rho}_\text{DE}) = \sum_{\alpha,\beta,\gamma} \Phi_{\gamma}^{\alpha\beta} \hat{\sigma}_\alpha \hat{\sigma}_\beta \rho_{\text{DE}}$, where $\hat{\sigma}_0$ is the identity matrix and $\hat{\sigma}_X$, $\hat{\sigma}_Y$, and $\hat{\sigma}_Z$ are the corresponding Pauli matrices. The 64 real parameters $\Phi_{\gamma}^{\alpha\beta}$, thus fully specify $\Phi$. c) The process map $\Phi_{\gamma}^{\alpha\beta}$ as measured by the quantum process tomography. The matrix elements of $\Phi_{\gamma}^{\alpha\beta}$ are presented such that the rows correspond to the indices $\alpha, \beta$ of the DE+1photon output state, and the columns correspond to the index $\gamma$ of the input DE state. d) The process map $\Phi_{\gamma}^{\alpha\beta}$, calculated assuming the ideal protocol (CNOT and Hadamard gates), as in Fig. 1d.

sensitive twisted-gradient-descent minimization method (See Appendix). It is well known that the space of physically completely-positive (CP) maps can be identified with a cone-like space where any unitary process sits on an extremal ray of the cone. To find the best CP fit of $\Phi$ therefore requires minimizing a known function $F$ (representing minus log likelihood) over this cone. Gradient descent tries to find the minimum of a function $F(x)$ by going roughly along gradient lines of $F$. Our newly developed approach uses an edge-sensitive twisted-gradient-descent in order to prevent our gradient descent solution from getting stuck in the boundary of the physically allowed cone-like region.

Fig. 3c-d presents the physical CP-process-map obtained using this method. We compare the acquired physical process with the ideal unitary process of the cluster protocol. The fidelity [17, 33] between the two processes is 0.83. The obtained fidelity is higher than in the previous demonstration [17]. The higher fidelity is attributed to the 3-fold shorter time between the excitations, which reduces the influence of the DE decoherence during its precession. The relatively high fidelity to the ideal protocol indicates that our device can deterministically generate photonic cluster states of high quality, thereby providing a better resource for quantum information processing.

IV. DISCUSSION

We characterize the robustness of the entanglement in the 1D cluster-state using the notion of localizable entanglement (LE) [35]. The LE is the negativity [34] between two qubits in the cluster after all the other qubits are projected onto a suitable polarization-basis. The LE decays exponentially with the distance between the qubits [17, 32]:

$$N(d) = N_{nn} \exp\left(- (d-1)/\zeta_{LE}\right),$$

(3)

where $N_{nn}$ is the negativity between nearest-neighbor qubits, $d$ is the distance between the qubits, and $\zeta_{LE}$ is the characteristic decay-length of the LE. In Fig. 4, we plot using pink circles the LE in the state of a spin+Nphotons, obtained from the measured process map, as a function of the distance between two qubits in the string. As expected, the LE in the 1D cluster state decays exponentially with the distance between the two qubits [32]. Fig. 4 shows that the entanglement in the cluster persists up to six photons. This presents an improvement over Ref. [17], resulting from the reduction in the DE spin decoherence between the optical pulses.

The negativity between nearest neighbor and next nearest neighbor 2 qubits can be directly obtained from our quantum state tomography. The entanglement...
Figure 4. The localizable entanglement (LE - pink circles) in the generated state vs. the distance between two qubits in the string. The pink circles correspond to the LE between qubits $m$ and $m + d$ in the state of DE+$N$photons obtained using the measured process map. The dashed lines represent the best fit to an exponential decay law $N(d) = N_m \exp(-(d - 1)/\zeta_{LE})$, where $N_m$ is the negativity [34] between nearest-neighbor qubits, $d$ is the distance between the qubits, and $\zeta_{LE}$ is the characteristic decay-length of the LE. The light-blue shaded region represents one standard deviation of the uncertainty in the measurements determining the process map. The purple and yellow data points represent the directly measured LE of the DE and the first emitted photon in a two- and a three-qubit string, respectively, acquired from the complete tomographic measurements of those states presented in Fig. 3c and d.

between the DE and the photon emitted after one cycle of the protocol is obtained from the density matrix of the DE+1photon in Fig. 3a which has negativity of $N = 0.27 \pm 0.03$. This negativity is marked as a purple data point in Fig. 4. Similarly, the spin+2photons 3-qubit state resulted from application of two cycles of the protocol is represented by the 3-qubit density matrix, displayed in Fig. 3b. The negativity of the density matrix of the two external qubits, after projecting the central qubit on the X polarization basis is $N = 0.18 \pm 0.05$, marked by the yellow point in Fig. 4.

In summary, we demonstrate a gigahertz-rate deterministic generation of entangled photons in a cluster state, which is 3-times faster than previously demonstrated. We developed a novel method for spin - multi photon quantum state tomography and for characterizing the periodic process map which generates the photonic cluster state. Using this method we show that the enhanced cluster generation rate also improves the robustness of the entanglement in the generated multi -photon state. The measured process map has fidelity of 0.83 to the ideal one, and the entanglement in the cluster state persists up to 6 consecutive qubits. Our studies combined with further feasible optimizations of the device may lead to implementations of quantum communication and efficient distribution of quantum entanglement between remote nodes.

ACKNOWLEDGMENTS

The support of the Israeli Science Foundation (ISF), and that of the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme (Grant Agreement No. 695188) are gratefully acknowledged.
The need for Lagrange multiplier is a cost we pay for E. R. Schmidgall, I. Schwartz, D. Cogan, L. Gantz, T. Heindel, S. Reitenstein, and D. Gershoni, Applied Physics Letters 106, 193101 (2015).

D. Loss and D. P. DiVincenzo, Physical Review A 57, 120 (1998).

D. P. DiVincenzo, Fortschritte der Physik 48, 771 (2000).

E. Poem, Y. Kodriano, C. Tradonsky, N. H. Lindner, B. D. Gerardot, P. M. Petroff, and D. Gershoni, Nature Physics 6, 993 (2010).

I. Schwartz, E. R. Schmidgall, L. Gantz, D. Cogan, E. Bordo, Y. Don, M. Zielinski, and D. Gershoni, Physical Review X 5, (2015).

I. Schwartz, D. Cogan, E. R. Schmidgall, L. Gantz, Y. Don, M. Zielinski, and D. Gershoni, Physical Review B 92 (2015).

M. Zielinski, Y. Don, and D. Gershoni, Physical Review B 91, (2015).

M. Bayer, G. Ortner, O. Stern, A. Kuther, A. A. Gorbunov, A. Forchel, P. Hawrylak, S. Fafard, K. Hinzer, T. L. Reinecke, S. N. Walck, J. P. Reithmaier, F. Klopff, and F. Schäfer, Physical Review B 65, (2002).

E. L. Ivchenko, Optical Spectroscopy of Semiconductor Nanostructures (Alpha Science, 2001).

D. Cogan, G. Peniakov, Z.-E. Su, and D. Gershoni, Physical Review B 100, 101.055424.

M. Popp, F. Verstraete, M. A. Martin-Delgado, and J. I. Cirac, Physical Review A 71 (2005), 10.1103/PhysRevA.71.042306.

R. Jozsa, Journal of Modern Optics 41, 2315 (1994).

A. Peres, Physical Review Letters 77, 1413 (1996).

F. Verstraete, M. Popp, and J. I. Cirac, Physical Review Letters 92 (2004), 10.1103/physrevlett.92.027901.

We identify H,B,R polarizations with \( \hat{x}, \hat{y}, \hat{z} \) respectively.

The error estimate \( \Delta_{\text{tot}} \) appearing in \( \tilde{F}_0 \) is constructed in the standard way from the error estimates \( \Delta R \) and \( \Delta S \). The error \( \Delta S \) is now taken care of by the second term (and \( \Delta R \) is negligible anyway).

It is convenient to extend \( F \) to arbitrary matrices by defining \( F(A) = F \left( \frac{1}{2}(A + A^\dagger) \right) \).

Euclidean gradient descent fails even if one includes similar improvement in it.

The need for Lagrange multiplier is a cost we pay for using non flat metric. In flat metric, the constraints may be solved trivially.

**Appendix A: the process likelihood function**

The basic (single cycle) process taking the QD-spin into a spin plus emitted photon is described by a process map \( \Phi \) taking one-qubit state into a two-qubit state. Expanding these quantum states in terms of Pauli matrices allows writing the map as

\[
\rho = \sum r_{\mu} \sigma^\mu \mapsto \Phi(\rho) = \sum r_{\mu} \phi_{\mu\lambda} \sigma^\nu \otimes \sigma^\lambda.
\]

Here \( \phi_{\mu\lambda} \) with \( \mu, \lambda \in \{0, x, y, z\} \) are 64 real coefficient which define the process map \( \Phi \).

The trace preserving condition \( Tr\Phi(\rho) = Tr\rho \) fixes 4 out of the 64 coefficient \( \phi_{0\lambda} \) namely it requires that \( \phi_{00} = \frac{1}{2} \phi_{00} \). We wish to estimate the other 60 parameter by finding the best fit to the experimental data.

In an experiment where the initial spin was \( \vec{s} \) and the emitted photon was projected on state of spin \( [30] \vec{p} \) we would ideally expect the measured rate \( R \) of photon emission and final spin \( \vec{S} \) to satisfy \( \phi_{\nu\lambda} \sigma_{\mu} \rho_{p=0} \propto R S_\nu \). Here \( s, p, S \) are four-ectors whose zeroth component equals 1 and their spatial components are \( \vec{s}, \vec{p}, \vec{S} \). Repeating such measurement for six different spin polarizations \( \vec{s} \) and six different photon polarizations \( \vec{p} \) gives 6 \times 6 \times 4 = 144 equations for the 60 unknown components of \( \Phi \).

A straightforward approach to determine \( \Phi \) is then to use a least square method minimizing the expression

\[
F_0 = \sum_{s, p, S} \frac{1}{\Delta_{\text{tot}}^2} \left[ \phi_{\nu\lambda} \sigma_{\mu} \rho_{p=0} - R S_\nu \right]^2. 
\]

(1)

Essentially representing minus log likelihood.) Here \( \Delta_{\text{tot}} \) are the error estimates of the corresponding measurement. A slightly more sophisticated approach would recall that appearance of each of the six initial polarizations \( \vec{s} \) in 24 different sums, can potentially make the errors correlated. One way to take this into consideration is to use a more complicated square sum constructed using a non-diagonal covariance matrix. A completely equivalent method consists of defining new variables \( \vec{s}' \) representing the true initial polarization of the DE and using the new sum of square error function \( F_0 = \tilde{F}_0(\phi, s') \) defined by [37]

\[
\tilde{F}_0(\phi, s') = \sum_{\nu\lambda} \frac{1}{\Delta_{\text{tot}}^2} \left[ \phi_{\nu\lambda} \sigma_{\mu} \rho_{p=0} - R S_\nu \right]^2 + \sum_{\nu\lambda} \left( \frac{\Delta_{\text{tot}}^2}{\Delta_{\text{tot}}^2} - 1 \right) \left( \frac{\Delta_{\text{tot}}^2}{\Delta_{\text{tot}}^2} - 1 \right) \left( \frac{\Delta_{\text{tot}}^2}{\Delta_{\text{tot}}^2} - 1 \right).
\]

Minimizing this expression with respect to \( s' \) yields a standard square sum \( \tilde{F}_0(\phi) \) corresponding to the correct non-diagonal covariance matrix. We looked for a minimum of \( \tilde{F}_0(\phi, s') \) with respect to both the process map \( \phi \) and the unknown initialization polarizations \( s' \).

**Appendix B: Completely positive condition and twisted gradient descent.**

It is well known that to be physically acceptable, a process map \( \Phi \) must be completely positive (CP). A process map is CP iff the associated Choi matrix which may be defined (up to unimportant normalization factor) by

\[
C_\Phi = \sum_{\nu\lambda} \phi_{\nu\lambda} \sigma_\nu \otimes \sigma_\lambda \otimes \sigma_\lambda
\]

is positive (semi-definite) \( C_\Phi \geq 0 \). The bar over \( \sigma_\mu \) denotes complex conjugation and we make no distinction here between lower and upper indices. In general the Choi matrix \( C_\Phi \) is a (complex) hermitian 8 \times 8 matrix which may be used as an alternative description of \( \Phi \).

Simple minded naive minimization of \( \tilde{F}_0 \) leads to a process map which is not CP and hence not physically acceptable. To understand why this happens, recall that our process is very close to an idealized process which is...
unitary. Any unitary process is an extreme point of the cone of CP-maps and has a rank-1 Choi matrix. In other words, for a unitary process \( \Phi \) close to unitary has therefore 7 very small Choi matrix eigenvalues. It is thus not surprising that very small experimental errors can lead us to estimate some of these eigenvalues as negative, in contradiction with the CP condition. (Had our \( \Phi \) been equal to the ideal unitary map, a small random error in each eigenvalue would lead to non CP map with probability \( \frac{127}{128} = 0.992 \).) In other words, the encountered difficulty is actually a good sign indicating that our process has quite low decoherence.

To find the best CP fit of \( \Phi \) therefore requires minimizing a known function \( F \) (representing minus log likelihood) over the subset of CP-maps, which as explained above may be identified (through the Choi representation) with the cone of positive matrices. This is closely related to the extensively studied field of convex optimization. We have not found however in the convex optimization literature a method which looks to exactly fit the boundaries e.g. if it has \( \sum |\lambda_i|^2 \neq 1 \), the positivity constraint allows the estimate \( \sum \lambda_{ij} = 1 \) to vanish. Our \( A \) is therefore a process Choi matrix, is already given to us in such a form by Eq. (B1). One can then write \( \nabla F = \sum \Xi_i \partial F_i \).

The gradient we use corresponds to a non-flat riemannian metric and may be expressed as \( \nabla F = \sqrt{\mathcal{A}} (\nabla F) \sqrt{\mathcal{A}} \) (where \( \nabla F \) is as above). We therefore look for a minimum of \( F \) over the set of positive (semi-definite) matrices by using a gradient descent step of the form

\[
A \rightarrow A + \Delta A, \quad \Delta A = -q\sqrt{\mathcal{A}} (\nabla F) \sqrt{\mathcal{A}}
\]

Here \( q > 0 \) is a scalar chosen so that \( F(A + \Delta A) \) is minimal under the constraint \( A + \Delta A \geq 0 \). Note that if \( \nabla F = O(1) \) then the positivity constraint allows \( q \) to remain \( O(1) \) even if \( A \) is very close to the boundary of the cone of positive matrices. All our process map estimations were obtained using this minimization scheme which we implemented using Mathematica

Although the basic method described above works reasonably well, we found that some extra improvement [39] is gained if after each step we push \( A \) slightly away from the boundary of the allowed region by updating it as \( A \rightarrow (1 - \varepsilon)A + \frac{1}{2}\varepsilon I \) with \( \varepsilon \ll 1 \). (We suspect that the need for this step might be related to the finite precision of the numerical calculations.) We increase or decrease \( \varepsilon \) dynamically during the computation, depending on the performance of previous iteration step. In practice \( \varepsilon \) ranged between \( 10^{-4} \) and \( 10^{-10} \) and scaled roughly as 2-3 times the minimal eigenvalue of \( A \).

The modified gradient descent method described here is quite general and can be applied to any \( F(A) \). In practice, our \( F(A) \) was of the form \( F(A) = F_0(A) + \sum_{\mu=0}^3 \lambda_{i\mu} Tr(A \sigma_\mu \otimes I) \) where \( F_0(A) \) is the square sum described in subsection A, and the second term consists of 4 lagrange multipliers required to enforce the normalization condition \( \sum_{\mu=0}^3 \lambda_{i\mu} = 1 \). The values of the multipliers \( \lambda_{i\mu} \) at each iteration step are easily determined numerically by requiring that \( \Delta A = -q\sqrt{\mathcal{A}} (\nabla F) \sqrt{\mathcal{A}} \) does not break the normalization condition. This amount to demanding the partial trace \( Tr_2(A \sqrt{\mathcal{A}} (\nabla F) \sqrt{\mathcal{A}}) \) to vanish, which is just a linear set of equations for \( \lambda_{i\mu} \).