One-step generation of multipartite entanglement among nitrogen-vacancy center ensembles

Wan-lu Song1,2, Zhang-qi Yin3, Wan-li Yang1, Xiao-bo Zhu4,5, Fei Zhou1 & Mang Feng1

1State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071, China, 2University of the Chinese Academy of Sciences, Beijing 100049, China, 3Center for Quantum Information, Institute for Interdisciplinary Information Sciences, Tsinghua University, Beijing 100084, China, 4The Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China, 5Collaborative Innovation Center of Quantum Matter, Beijing 100190, China.

We describe a one-step, deterministic and scalable scheme for creating macroscopic arbitrary entangled coherent states (ECSs) of separate nitrogen-vacancy center ensembles (NVEs) that couple to a superconducting flux qubit. We discuss how to generate the entangled states between the flux qubit and two NVEs by the resonant driving. Then the ECSs of the NVEs can be obtained by projecting the flux qubit, and the entanglement detection can be realized by transferring the quantum state from the NVEs to the flux qubit. Our numerical simulation shows that even under current experimental parameters the concurrence of the ECSs can approach unity. We emphasize that this method is straightforwardly extendable to the case of many NVEs.

As one of the promising building blocks for solid-state quantum information processing (QIP)4–6, the nitrogen-vacancy (NV) center consisting of a substitutional nitrogen atom and an adjacent vacancy in diamond can be optically controlled and fast microwave manipulated with long coherence time even at room temperature7,8. Additionally, benefiting from the enhancement of the magnetic-dipole coupling by a factor $\sqrt{N}$ through the collective excitation, the strong coupling regime has been reached in the hybrid systems consisting of NV center ensemble (NVE) and superconducting circuit9–17, which have recently attracted much attention. Several potential applications based on the collective excitation of the NVEs are investigated both theoretically and experimentally, such as quantum memories18–21, continuous variable entanglement22,23, quantum state transfer24 and quantum simulation25.

The practical quantum information processing requires generation of multipartite entanglement among NVEs, which is usually quite complicated and time-consuming. For example, to generate entanglement between two NVEs, we may employ a sequence of single- and two-qubit operations, such as SWAP gates. In this way, either the gate number or the operation time increases much more quickly as the number of the NVEs increases. Therefore, to efficiently generate multipartite entanglement among NVEs, we need to find a faster method with less operation time and gates. To this end, we present in this article a scalable and tunable framework for generating macroscopic arbitrary entangled coherent state (ECS)26–32 among many separate NVEs in a deterministic way. The key point of our proposal is that the flux qubit plays the role of data bus and the collective excitations of the NVEs behave as bosonic modes or harmonic oscillators in the low-excitation limit. Through the collective magnetic coupling and the in situ tunability of the flux qubit, we show that a macroscopic ECS of NVEs can be generated with high success possibilities even under the decoherence of the flux qubit and NVEs. More importantly, we consider the magnetic field applied along a direction [100] of the NV centers18 to suppress the broadening of the ODMR spectral lines due to excitations in four crystalline orientations. This can largely reduce the overheads in gate operations with the increment of the number of NVEs. So our scheme not only allows an efficient way to accessing the large-scale continuous variable quantum computing in many-body systems, but also serves as a critical building block towards scalable architectures based on solid-state QIP. It should be noted that the ECS owns unique quantum characteristics33–35 (e.g., robustness to single-particle decoherence) and can be used for various applications beside quantum information processing36, ranging from the fundamental quantum information theory37 and the implementation of quantum communication38,39 to quantum metrology40. The quantum error correction may also be easily implemented with the logical qubit defined by the superposition of the coherent states41,42.
The remainder of this paper is organized as follows. We present our system architecture in details in Sec. II, and the preparation and readout of the ECS of NVEs in Sec. III. Sec. IV focuses on the influence from the decoherence effects of the system. Discussion is presented in Sec. V. Finally, we conclude our findings in Sec. VI.

Results

System and Model. As schematically shown in Fig. 1, we consider two diamond NVEs glued near the boundary of a big superconducting gap-tunable flux qubit, which consists of four Josephson junctions characterized by Josephson energies $E_J$ and $\alpha E_p$, respectively. Near each of the NVEs, there are small flux qubits employed for detection, which are initially tuned to be uncoupled with both the NVEs and the big flux qubit. When the biasing of the main loop is close to one-half of a flux quantum $\Phi_0 = h/2e$, the gap-tunable flux qubit can be treated as an effective two-level system described by the Hamiltonian (in units of $\hbar = 1$ throughout the paper) $H_f = -\frac{1}{2} [ \sigma_x (\Phi_0 + \Delta(\Phi_0)/2) + \sigma_z ]$, where $\sigma_x$ and $\sigma_z$ are usual Pauli operators in the basis of flux eigenstates $|0\rangle$ (clockwise persistent current) and $|1\rangle$ (counterclockwise persistent current). Here, the energy splitting of the qubit is given by $\Delta(\Phi_0) = 2I_p (\Phi_0 - \Phi_0/2)$ with $I_p$ the persistent current in the flux qubit. $h \Delta(\Phi_0)/2$ is the energy of the tunnel splitting, which induces flip of the qubit states. Note that $\sigma_x$ and $\sigma_z$ can be controlled independently by the external magnetic flux threading the main loop and $x$ loop, respectively, through two extra microwave lines for a finite time period, e.g., of the order of nanosecond. This enables us to couple the flux qubit to the NVE at the optimal point where the flux qubit has its longest coherence time. In our case we tune $\sigma_x$ to be zero, and tune $\sigma_z$ to be equal to NVEs’ transition frequency. In this way, the flux qubit basis $|0\rangle, |1\rangle$ is defined in the eigenstates of the Pauli operator $\sigma_z$. Additionally, a resonantly driven microwave on the flux qubit with Rabi frequency $\Omega_d$ is also required to control the dynamics of the system.

For the spin-triplet ground state of a NV center with a zero-field splitting $D_\omega = 2.87$ GHz between $m_s = 0$ and the nearly degenerate sublevels $|m_s = \pm 1\rangle$, we apply an external magnetic field $B_{ext}$ along the crystalline direction [100] of the NV centers to split the degenerate sublevels $|m_s = \pm 1\rangle$, which results in a two-level system denoted by $|g\rangle = |\uparrow\rangle, m_s = 0$ and $|e\rangle = |\downarrow\rangle, m_s = -1$, respectively. So the Hamiltonian of the NVEs is given by

$$H_{NV E} = \sum_{j=1}^{N_v} \sum_{i=1}^{N_f} \left\{ D_{gs} \left( S_{j,i}^z \right)^2 + E \left[ \left( S_{j,i}^x \right)^2 - \left( S_{j,i}^y \right)^2 \right] + g_\mu B_{ext} S_{j,i}^z \right\},$$

where the last term represents the Zeeman splitting under the magnetic field $B_{ext}$ (i.e., the part of the external magnetic field along the crystalline directions). $E$ is the strain-induced splitting, $g_\mu$ is the ground-state Lande factor, $\mu_B$ is the Bohr magneton, and $N_v$ is the number of the NV centers involved in the $j$-th spin ensemble. To have a good two-level approximation for the NV centers, the diamond crystal is assumed to be bonded on top of the flux qubit chip with its [001] surface facing the chip. $S_{j,i}^x$, $S_{j,i}^y$, and $S_{j,i}^z$ are spin-1 Pauli operators for the $i$-th NVE in the $j$-th spin ensemble. Furthermore, we consider the case where the strain-induced fine-structure splitting is negligible compared to the Zeeman splitting, i.e., $E \left[ \left( S_{j,i}^x \right)^2 - \left( S_{j,i}^y \right)^2 \right] \ll g_\mu B_{ext} S_{j,i}^z$, and thereby the second term in $H_{NV E}$ can be neglected. Considering the subspace spanned by the states $|m_s = 0\rangle$ and $|m_s = -1\rangle$, we have the total Hamiltonian for the whole system as

$$H = H_f + H_{NV E}.$$
\[ H_1 = -\frac{1}{2} \Delta(\Phi_{\text{ext}}) \sigma_z + \Omega_d \cos(\omega_t t) \sigma_z + \sum_{j=1}^{N} \left( \frac{1}{2} \omega_j \tau_{x,j} + \frac{1}{\sqrt{2}} \epsilon_j \mu_B B_x \tau_{y,j} \sigma_z \left( 1 - \frac{i}{\hbar} \gamma_{j} \sigma_z \right) \right), \]  

where \( \omega_j = D_{\text{sp}} - \frac{\mu_B B_x}{2} \), the last term is the interaction between the NVE and the flux qubit, and \( B_x \) is the magnetic field (along the direction [001] of the NV centers) produced by the flux qubit. \( \tau_{x,j}, \tau_{y,j} \) are the spin \( \frac{1}{2} \) Pauli operators for the \( i \)-th NV center in the \( j \)-th spin ensemble. Actually, the additional magnetic field \( B_x \) is attributed to the superposition state of the \emph{clockwise} and \emph{counterclockwise} persistent currents. \( \Omega_d(\omega) \) is the Rabi frequency (frequency) of the additional microwave resonantly driving on the flux qubit.

Using the Holstein-Primakoff (HP) transformation, we map the spin operators to the bosonic eigenstates as follows:  
\[ \sum_{i=1}^{N_j} \frac{1}{\sqrt{N}} b_i^+ \cdot \sum_{i=1}^{N_j} \frac{1}{\sqrt{N}} b_i = \sqrt{N} b_j + \frac{N_j}{1} b_j + \frac{N_j}{N} b_j \]  
where the operators \( b_j \) and \( b_j^+ \) obey the standard bosonic commutator \( [b_j, b_j^+] = 1 \) in the weak excitation limit \( \langle b_j^+ b_j \rangle \ll N_j \). Here we denote the Fock state basis of the bosonic mode \( b_j \) and \( n_j \) \( b_j \equiv b_j^1 b_j^2 \ldots b_j^n \), for \( j = 1, 2 \). The coherent state of the mode \( b_j \) is defined as \( \alpha_j \equiv b_j^0 \). Then effective Hamiltonian on the basis of the HP transformations above becomes

\[ H_2 = -\frac{\Delta(\Phi_{\text{ext}})}{2} \sigma_z + \frac{\Omega_d}{2} \cos(\omega_t t) \sigma_z + \sum_{j=1}^{2} \left[ \alpha_j b_j^+ + G_j \left( b_j^+ + b_j \right) \right] \sigma_z, \]  

where \( G_j = \sqrt{N_j/2 \epsilon_j} \) is the effective coupling strength between the NVE and the flux qubit. In the case of \( G_j \ll \omega_j \) and \( \Omega_d \ll \omega_j \), the Hamiltonian in the basis of the eigenstates of the flux qubit takes the form  
\[ H_3 = \frac{\Delta(\Phi_{\text{ext}})}{2} \sigma_z + \frac{\Omega_d}{2} \left( e^{i\omega_t t} \sigma_z + e^{-i\omega_t t} \sigma_z \right) + \sum_{j=1}^{2} \left[ \alpha_j b_j^+ + G_j \left( b_j^+ \sigma_- + b_j \sigma_+ \right) \right] \sigma_z, \]  

where the rotating-wave approximation (RWA) is adopted to neglect fast oscillating terms. The ladder operators \( \sigma_\pm \) are defined as \( \sigma_\pm = (\sigma_x \pm i \sigma_y)/2 \).

Preparation and readout of the ECS. In what follows, we describe how to generate deterministically the ECS of two spin ensembles in our setup by following three steps:

(i) The system is initially prepared in the state \( |\psi(0)\rangle = |0\rangle_1 |0\rangle_2 \). Assuming that the splitting frequency \( \Delta(\Phi_{\text{ext}}) \) of the flux qubit and the frequency \( \omega_j \) of the driving field are set to be equal to the NVE collective excitation frequency \( \omega_j \), the effective Hamiltonian in the rotating frame becomes  
\[ H_4 = \sum_{j=1}^{2} \left[ \alpha_j b_j^+ + G_j \left( b_j^+ \sigma_- + b_j \sigma_+ \right) \right] \sigma_z. \]  
At this step, \( \Omega_d(\omega) \) in Eq. (3) is set to zero.

(ii) Turning on the drive \( \Omega_d(t) \) from zero to \( \Omega_d(\epsilon_j < \Omega_d \ll \omega_j) \) on the time scale of nanoseconds, we can switch from the flux qubit basis \( \{|0\rangle, |1\rangle\} \) to the dressed state basis \( \{|+\rangle, |\rangle\} \). The evolution operator takes the form  
\[ U(t) = U_0 \exp \left( -it \sum_{j=1}^{2} \frac{G_j}{2} \left( b_j^+ + b_j \right) \sigma_z \right), \]  

where \( U_0 = \exp \left( -i\frac{\Omega_d}{\hbar} \sigma_z /2 \right) \) is the rotating unitary mode of NVE by the amount of \( \pm i \Omega_d(t) \) conditional on the flux qubit states \( |0\rangle \) or \( |1\rangle \), and this complex amplitude \( i \Omega_d(t) \) is proportional to the evolution time. As a result, the system state \( |\psi(t)\rangle \) evolves into  
\[ |\psi(t)\rangle = |+\rangle_{1} |1\rangle_{2} + e^{i\Omega_d(t)/2} |\rangle_{1} |+\rangle_{2} \]  
with \( \zeta_1 = -i \Omega_d(t) \) and \( \zeta_2 = -i \Omega_d(t/2) \). For simplicity we set the phase factor \( \Omega_d(t) = \pi \sigma_n \) \((n = 1, 2, 3 \ldots)\). Then effective Hamiltonian in the form of \( \sigma_z \) is derived by precisely controlling the operation time. One can see the generated entanglement among the flux qubit and the two NVs.

(iii) To generate the ECS of NVs, we need to measure the state of the flux qubit on \(|0\rangle \) and \(|1\rangle \). This task can be performed by applying a pulse sequence on a dc SQUID attached to the flux qubit (not shown in Fig. 1), where the voltage state of the SQUID is very sensitive to the faint change of the flux, and depends on the switching probability of the energy eigenstates of the flux qubit. So the two NVs will be projected into the ECS \( |\psi\rangle = |\psi_1\rangle |\psi_2\rangle \) \(| \psi_1 \rangle = |\zeta_{11}\rangle |\zeta_{12}\rangle \) \(| \psi_2 \rangle = |\zeta_{21}\rangle |\zeta_{22}\rangle \). It is easy to verify that \(|0\rangle \) and \(|1\rangle \) are orthogonal to each other once \( |\zeta_{12}| \) > 0. In the limit of \( |\zeta_{12}| < 1 \), the coherent qubit states can be approximated as a superposition of Fock states \( |0\rangle = \sqrt{2} \exp \left( -|\zeta_{12}|^2 / 2 \right) |1\rangle_0 + O \left( |\zeta_{12}|^2 \right) \) and \( |1\rangle = \sqrt{2} \exp \left( -|\zeta_{12}|^2 / 2 \right) |0\rangle_1 + O \left( |\zeta_{12}|^2 \right) \). Therefore, in the case of \( |\zeta_{12}| < 1 \), we can transfer the state from NVs to flux qubits in the Fock state basis using the SWAP gate between the j-th NVE and the j-th flux qubit.

For a more general case of \(|\zeta_{12}| > 1 \), the usual way to the state transfer from the j-th NVE to the j-th flux qubit,  
\[ \left( \cos \theta / |\zeta_{12}| + \sin \theta |\zeta_{12}| \right) |0\rangle_0 \otimes |1\rangle_1 \otimes \cos \theta / |\zeta_{12}| + \sin \theta |\zeta_{12}| \right) \]  
by performing a sequence of single-qubit gates and the operation  
\[ U_1 = \exp \left( i \left( \frac{b_j^+ + b_j} {\sqrt{2}} \right) \right) \exp \left( \frac{\pi i}{2} b_j^+ b_j \right) \]  
where the operator \( \exp \left( -i \pi b_j^+ b_j \right) \) plays the role of flipping the flux qubit, achievable by first applying a single-qubit rotation and then by performing the operation \( \exp \left( -i \pi b_j^+ b_j \right) \) on the flux qubit. Alternatively, the operations above can be accomplished by the following method. Suppose that the effective Hamiltonian between the NVs and the small flux qubit is similar to Eq. (3) but without the microwave drive \( \Omega_d \). So the corresponding operation is \( \exp \left( -i \pi \alpha_j \sigma_z / 2 \right) \), where \( \alpha_j = G_j / \Lambda_j \), and \( \Lambda_j \) is the detuning between the transition frequencies of the j-th small flux qubit and the j-th NVE. Additionally, the operation \( \exp \left( i \epsilon_j \left( b_j^+ + b_j \right) \right) \) with \( \epsilon_j = \pi / (2 \zeta_{1j}) \) is actually the conditional displacement of the bosonic mode if only if the flux qubit is in the excited state \( |1\rangle \). Such conditional displacements have been achieved by Eq. (3) with a strong microwave drive. Hence the operation \( U_1 \) should be achievable with currently experimental technology, and then the
entanglement between NVEs can be detected by measuring the states of small flux qubits.

We should note that the detection in our scheme requires the coupling strength of each NV center to the large flux qubit to be proportional to that to the small one, which guarantees the collective mode detected by the small flux qubit to be identical to that already prepared in the large flux qubit. Although it looks like an additional technical difficulty, the operations are experimentally doable. For example, the NVEs can be placed to the positions where the magnetic fields generated by both the small and the large flux qubits are nearly homogenous due to careful design of the qubits’ circuit patterns.51.

Discussion

To visualize the decoherence effect on the evolution of concurrence between the NVEs, we have plotted the time-dependent $C_{12}(t)$ in Fig. 2. Due to the presence of decoherence, the concurrence increases first and then decreases gradually to zero. So to carry out our scheme more efficiently, we have to suppress these imperfect factors as much as we can. On the other hand, the concurrence keeps to be of very high values ($\approx 0.95$) under the decoherence as long as the time is evaluated within the domain $[2/G, 2.5/G]$, and the maximal entanglement ($\approx 0.97$) can be obtained if the operation time $t'_{op} \approx 2.3/G$. We emphasize that a stronger decoherence only slightly reduces the maximal value of the concurrence, as shown in the bottom panel of Fig. 2.

In Fig. 3, we have exactly calculated the evolution of the system, where the fast oscillating terms are involved. We may compare the generated ECS $|\psi'(t)\rangle$ in the non-RWA case with the ECS $|\psi(t)\rangle$ generated by the effective unitary operator $U(t) = U_0 \exp \left\{ -i \left[ \sum_{j=1}^{5} C_{j} \left( b_j^+ + b_j \right) \hat{a}_j\right] \right\}$. Here, the fidelity is defined as $F(t) = |\langle \psi'(t)|\psi(t)\rangle|$. As shown in Fig. 3, the fidelity $F(t)$ decreases in time evolution, which is larger than 0.98 only for $t_0 \approx 2/G$ and $\Omega/G = 30$. Therefore, effective unitary operator $U(t)$ is valid within this regime, and the generated ECS has the amplitude $|x| = |\beta| = |ut_0/G| = 1$ at time $t_0$, which is large enough to define the orthogonal qubit. In practice, if we choose $G = 25$ MHz, the microwave drive is $\Omega = 750$ MHz, which is within the reach of experimental feasibility.56

The condition on HP transformation for the NVEs requires that the total number of the excitations be much smaller than the number of the NV centers in each NVE. In our case, the average photon number in the two NVEs can be calculated as

$$N_1 = |x|^2 \xi / S, \quad N_2 = |\beta|^2 \xi / S,$$

where $\xi = e^{-\gamma t} + e^{\gamma t} = 2qP_1P_2 \cos(\Omega_0 \delta)$. Considering the case of $G_1 \neq G_2$, we set $G_2 = (1 + \Delta)G_1$. As shown in Fig. 4, we plot the total photon number $N = N_1 + N_2$ as a function of the parameter $\Delta$ and time. One can find that the value of $N$ is smaller than 20. So the small excitation number ensures a reasonable HP transformation in our scheme.

We address some remarks for experimental implementation of our scheme. In our proposal, the flux qubit is strongly driven and

![Figure 2](https://example.com/figure2.png)

**Figure 2** | Top: Density plot of the concurrence of the ECS versus the dimensionless parameters $\Gamma$ and $t$, where $\gamma = 0.01$. Middle: Density plot of the concurrence of the ECS versus the dimensionless parameters $\gamma$ and $t$, where $\Gamma = 0.01$. Bottom: Concurrence of the ECS versus the dimensionless time $t$, where the solid, dashed, and dotted lines denote $\Gamma = \gamma = 0$, $\Gamma = \gamma/10 = 0.001$, and $\Gamma = \gamma = 0.01$, respectively. Here we have used the method in$^\text{\textnormal{54}}$ to calculate the concurrence in the case of $\Gamma = \gamma = 0$, $\Omega_0 = 15$ and $G_1 = G_2 = 1$ are assumed in all panels.
the qubit basis is changed to dressed states $|+\rangle$ and $|\rangle$. This is equivalent to the continuous dynamical decoupling. Based on recent experiments, the NVE-flux qubit coupling strength has reached up to 70 MHz. In our proposal, if we set $G_1 = G_2 = 50$ MHz and $\tau_{\text{EE}} = 2.3/G$, the operation time for obtaining the ECS of NVEs with high values of concurrence is about 40 ns, which is much shorter than the coherence times of the NV centers and the flux qubit. In addition, the decoherence of the NVE increases with the density of the NV centers. Thus, it is required to suppress the decoherence of the NVE by the spin-echo pulses and/or by improving the conversion rate from nitrogen to NV which decrease the redundant nitrogen spins and thereby reduce the linewidth of the NVEs.

As a final remark, we emphasize that the setup plotted in Fig. 1 is straightforwardly extendable to the case with many NVEs involved, which would be essential to scalable QIP. For a more specific description, we have designed in Fig. 5 a novel tunable architecture that allows superconducting flux qubits to provide flexibility in arrangement of NVEs. In such a way, all spin ensembles are arranged in two or three spatial dimensions, where the flux qubit serves as a quantum bus for the non-interacting spin ensembles. The total Hamiltonian is given by

$$H_N = -\frac{\Delta(\Phi^\text{ext})}{2}\sigma_z + \Omega_d \cos(\omega_d t)\sigma_z + \sum_{j=1}^{N} [(\omega_j b_j^+ b_j + G_j(b_j^+ + b_j)\sigma_z],$$

with $N$ the total number of NVEs. Eq. (6) can take an effective form

$$\sum_{j=1}^{N} G_j \left( b_j^+ + b_j \right) \sigma_z + \frac{1}{2} \Omega_d \sigma_z$$

in the dressed state basis by performing the similar transformations to the two-NVE case. As a result, all the NVEs will be projected into the ECS

$$|\psi^N_\pm\rangle = \frac{1}{\sqrt{2}} (|\alpha_1\rangle|\alpha_2\rangle \cdots |\alpha_N\rangle_{N} \pm |\alpha_1\rangle |\alpha_2\rangle \cdots |\alpha_N\rangle_{N})$$

with the normalization factor $Y$, and $\sigma_z = -iG_d/2$ corresponding to the readout from the states $|0\rangle$ and $|1\rangle$ with respect to the flux qubit, respectively. We note that optical pulses are required to individually address the NVEs in the spatial scale. The present method would be very efficient for generating a continuous-variable entanglement among separate spin ensembles, which is a necessary step on the path towards a scalable solid-state quantum computing.

In summary, we have proposed a scheme of one-step creation of arbitrary macroscopic ECSs among many separate NVEs through the collective magnetic coupling to the flux qubit. We have shown that the ECSs of the spin ensembles can be achieved with high success possibilities even under the influence of decoherence from the flux qubit and NVEs. We have realized the worry about the large loop of the flux qubit which may cause more noise. However, we consider that the gap-tunable flux qubit employed in our scheme can work at the optimal point, strongly suppressing the dephasing time. On the other hand, we have recently fabricated some large loop gap-tunable flux qubits experimentally with the coherent time on the order of one microsecond, in which no significant drop was observed when the flux qubit loop size is increased from $4 \times 5$ $\mu$m$^2$ to $20 \times 20$ $\mu$m$^2$. Considering the fact that we can use small pieces of NV sample with the size about $1 \times 1$ $\mu$m$^2$, we are confident of the feasibility of a multi-qubit entanglement using a flux qubit with the loop size of $400$ $\mu$m$^2$. Therefore we argue that the proposal is practical under the present experimental parameters, and should be helpful for large-scale QIP in solid-state quantum systems, particularly for continuous-variable quantum computing and quantum communication.
Method

Modeling decoherence effects. From now on we analyze the influence from decoherence of the flux qubit and the NVs. Below we may focus on the decoherence effect in the step (ii) using the master equation

\[
\dot{\rho}(t) = -i \sum_{j=1}^{G} \left( \frac{G_j}{2} \left( b_j^+ b_j + b_j^+ b_j^\dagger - 2 \Omega_j \sigma_x \sigma_y \rho(t) \right) + \frac{G_j}{2} \Omega_j \sigma_x \rho(t) \right) + \mathcal{L}(\rho(t)),
\]

\[
\mathcal{L}(\rho(t)) = \sum_{j=1}^{G} \left( 2 (b_j^+ b_j - b_j b_j^+) \rho(t) - \rho(t) (b_j^+ b_j^+ - b_j^+ b_j) \right)
\]

where \( \gamma(\Gamma) \) is the decay rate of the flux qubit (NVE). For simplicity, we have assumed \( \Gamma_1 = \Gamma_2 = \Gamma \). Using the superoperator technique and Hausdorff similarity transformation, we deduce the following differential equations with respect to the density matrix of the flux qubit,

\[
\dot{\rho}_{+}(t) = \frac{G_1}{2} \left( b_1^+ b_1^+ + b_1^+ b_1^\dagger + \frac{1}{2} \Omega_1 \rho_{+}(t) \right) + \mathcal{L}(\rho_{+}(t)),
\]

\[
\dot{\rho}_{-}(t) = \frac{G_1}{2} \left( b_1^+ b_1^+ + b_1^+ b_1^\dagger + \frac{1}{2} \Omega_1 \rho_{-}(t) \right) + \mathcal{L}(\rho_{-}(t)),
\]

So we have the concurrence for the ECS in the form

\[
C_E(t) = \max \{0, \Omega_1 M_2 \rho(t) M_2^\dagger \}.
\]

1. Childress, L. et al. Coherent dynamics of coupled electron and nuclear spin qubits in diamond. Science 314, 281 (2006).
2. Dutt, M. V. G. et al. Quantum register based on individual electronic and nuclear spin qubits in diamond. Science 316, 1312 (2007).
3. Jiang, L. et al. Repetitive readout of a single electronic spin via quantum logic with nuclear spin ancilla. Science 326, 267 (2009).
4. Neumann, P. et al. Single-shot readout of a single nuclear spin. Science 329, 542 (2010).
5. Neumann, P. et al. Quantum register based on coupled electron spins in a room-temperature solid. Nature Phys. 6, 249 (2010).
6. Togan, E. et al. Quantum entanglement between an optical photon and a solid-state spin qubit. Nature 466, 730 (2010).
7. Balasubramanian, G. Ultralong spin coherence time in isotopically engineered diamond. Nat. Materials 8, 383 (2009).
8. Mizuochi, N. Coherence of single spins coupled to a nuclear spin bath of varying density. Phys. Rev. B 80, 041301 (2009).
9. Xiang, Z. L., Ashhab, S., You, J. Q. & Nori, F. Hybrid quantum circuits: Superconducting circuits interacting with other quantum systems. Rev. Mod. Phys. 85, 623 (2013).
10. Xiang, Z. L., Lu, X. Y., Li, T. F., You, J. Q. & Nori, F. Hybrid quantum circuit consisting of a superconducting flux qubit coupled to a spin ensemble and a transmission-line resonator. Phys. Rev. B 87, 144516 (2013).
11. Kubo, Y. et al. Strong coupling of a spin ensemble to a superconducting resonator. Phys. Rev. Lett. 105, 140502 (2010).
12. Schuster, D. I. et al. High-cooperativity coupling of electron-spin ensembles to superconducting cavities. Phys. Rev. Lett. 105, 140501 (2010).
13. Amstutz, R. et al. Quantum monitoring of coherently coupled spin systems. Phys. Rev. Lett. 107, 060502 (2011).
14. Kubo, Y. et al. Hybrid quantum circuit with a superconducting qubit coupled to a spin ensemble. Phys. Rev. Lett. 107, 220501 (2011).
15. Kubo, Y. et al. Storage and retrieval of a microwave field in a spin ensemble. Phys. Rev. A 85, 012333 (2012).
16. Zhu, X. et al. Coherent coupling of a superconducting flux qubit to an electron spin ensemble in diamond. Nature 478, 221 (2011).
17. Marcos, D. et al. Coherent quantum control of nuclear-vacancy centers in diamond to superconducting flux qubits. Phys. Rev. Lett. 105, 210501 (2010).
18. Saito, S. et al. Towards realizing a quantum memory for a superconducting qubit: Storage and retrieval of quantum states. Phys. Rev. Lett. 111, 107008 (2013).
19. Jilsig, E., Grezes, C., Bertet, P. & Mølmer, K. Quantum memory for microwave photons in an inhomogeneously broadened spin ensemble. Phys. Rev. Lett. 110, 250503 (2013).
20. Yang, W. L., Yin, Z. Q., Hu, Y., Feng, M. & Du, J. F. High-fidelity quantum memory using nitrogen-vacancy center ensemble for hybrid quantum computation. Phys. Rev. A 84, 010301(R) (2011).
21. Cai, J., Jelezko, F., Katz, N., Reitze, A. & Plenio, M. B. Long-lived driven solid-state quantum memory. New. J. Phys. 14, 093030 (2012).
22. Yang, W. L., Yin, Z. Q., Chen, Q., Chen, Y. & Feng, M. Two-mode squeezing of distant nitrogen-vacancy-center ensembles by manipulating the reservoir. Phys. Rev. A 85, 022324 (2012).
23. Ma, S. L., Li, P. B., Fang, A. P., Gao, S. Y. & Li, F. L. Dissipation-assisted generation of steady-state single-mode squeezing of collective excitations in a solid-state spin ensemble. Phys. Rev. A 88, 013837 (2013).
24. Chen, Q., Yang, W. L. & Feng, M. Controllable quantum state transfer and entanglement generation between distant nitrogen-vacancy-center ensembles coupled to superconducting flux qubits. Phys. Rev. A 86, 022327 (2012).
25. Yang, W. L. et al. Quantum simulation of an artificial Abelian gauge field using nitrogen-vacancy-center ensembles coupled to superconducting resonators. Phys. Rev. A 86, 012307 (2012).
26. Sanders, B. C. Entangled coherent states. Phys. Rev. A 45, 6811 (1992).
27. Zhang, W. M., Feng, D. H. & Gilmore, R. Coherent states: Theory and some applications. Rev. Mod. Phys. 62, 867 (1990).
28. Chen, M. Y., Tu, M. W. Y. & Zhang, W. M. Entangling two superconducting LC circuits by manipulating the reservoir. Chin. Phys. B 19, 107008 (2010).
29. Kubo, Y. et al. Quantum memory for a microwave photon in a solid-state spin ensemble. Phys. Rev. Lett. 107, 220501 (2011).
30. Jiang, L. et al. Coherent qubit control in a superconducting circuit. Phys. Rev. B 86, 140502 (2012).
31. Liu, J., Yang, W. L. & Feng, M. Controllable quantum state transfer and entanglement generation between distant nitrogen-vacancy-center ensembles coupled to superconducting flux qubits. Phys. Rev. A 86, 045801 (2012).
32. Zhou, L. & Xiong, H. A macroscopically entangled coherent state generator in a V configuration atom system. J. Phys. B: At. Mol. Opt. Phys. 41, 025301 (2008).
33. Liu, J. Y., Wang, Z. B., Wu, H. Z. & Zheng, S. B. Generation of cluster states for cavity QED. Phys. Rev. A 71, 4207 (2008).
34. Munro, W. J., Milburn, G. J. & Sanders, B. C. Entangled coherent-state qubits in an ion trap. Phys. Rev. A 62, 052108 (2000).
35. Lukin, M. D., Yelin, S. F. & Fleischhauer, M. Entanglement of atomic ensembles by trapping correlated photon states. Phys. Rev. Lett. 84, 4232 (2000).
