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## Floquet engineering to entanglement protection of distant nitrogen vacancy centers

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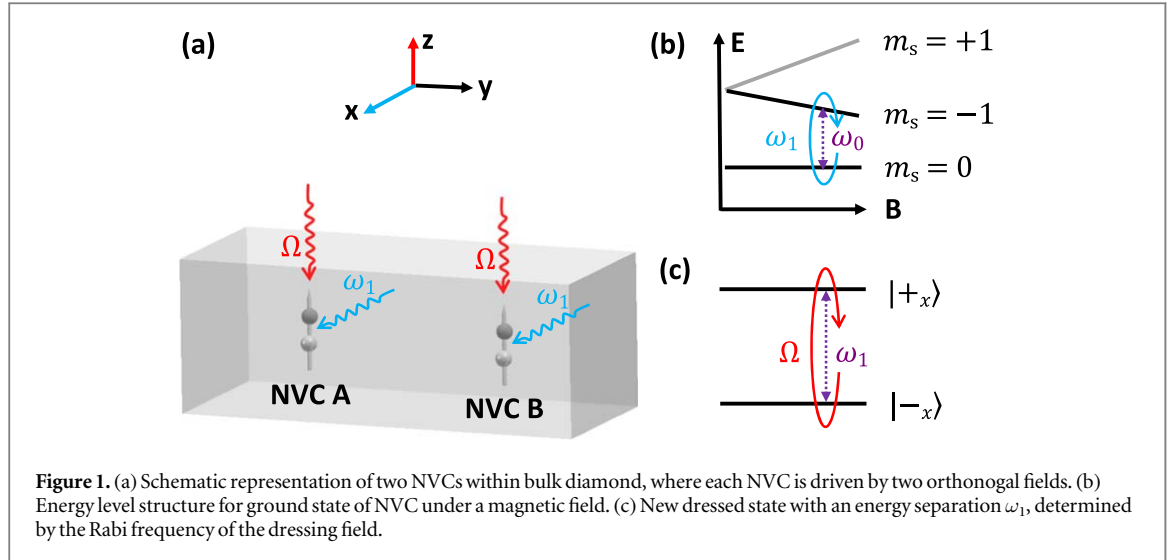
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W L Yang<sup>1</sup>, W L Song<sup>1</sup>, Jun-Hong An<sup>2</sup> , M Feng<sup>1</sup>, D Suter<sup>3</sup> and Jiangfeng Du<sup>4,5,6</sup><sup>1</sup> State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071, People's Republic of China<sup>2</sup> School of Physical Science and Technology, Lanzhou University, Lanzhou 730000, People's Republic of China<sup>3</sup> Fakultät Physik, Technische Universität Dortmund, D-44221 Dortmund, Germany<sup>4</sup> CAS Key Laboratory of Microscale Magnetic Resonance and Department of Modern Physics, University of Science and Technology of China, Hefei 230026, People's Republic of China<sup>5</sup> Hefei National Laboratory for Physical Sciences at the Microscale, University of Science and Technology of China, Hefei 230026, People's Republic of China<sup>6</sup> Synergetic Innovation Center of Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei 230026, People's Republic of China**E-mail:** [djf@ustc.edu.cn](mailto:djf@ustc.edu.cn)**Keywords:** quantum entanglement, strong periodical driving, nitrogen vacancy center**Abstract**

It remains challenging to preserve entanglement between distant solid-state qubits with high-fidelity, such as nitrogen vacancy centers (NVs). We propose a Floquet engineering strategy to protect the maximal entanglement between two weakly interacting NVs separated in long spatial distance by locally applying periodic strong driving on the NVs. It is found that entanglement of the Floquet states of the NVs resonantly reaches its maximum during the whole driving period at certain values of the driving parameters. Our analysis reveals that it is the occurrence of the avoided level crossing in the Floquet quasienergy spectrum which results in such entanglement resonance. The dissipation effect on the generated entanglement has also been analyzed. Our results may be of both theoretical and experimental interests in exploring the long-lasting entanglement between weakly interacting spins to long distances in realistic environments.

**Introduction**

Among the solid-state spins, the diamond nitrogen vacancy center (NV) are particularly attractive, owing to extremely long electronic and nuclear spin lifetimes, well qubit readout, fast initialization as well as robust optical interface [1–6]. Until now, several hallmark demonstrations on the entanglement between NVs have been achieved experimentally using the method of magnetic dipolar coupling [7], strong nonclassical correlations between Raman-scattered photons [8], entanglement swapping [9, 10], and projective measurements [11, 12], respectively. However, the decoherence process, owing to the unavoidable interaction with environment, tends to degrade the entanglement. Therefore, the entanglement protection or preservation has become a long-standing challenge. Along these lines, previous studies have shown that quantum coherence protection [13], bipartite entanglement protection [14], and quantum gate protection [15], have been realized using dynamical decoupling [16]. Motivated by these advances, here we study how to realize a long-time entanglement protection of two distant NVs with weak dipole–dipole interaction using the periodic strong driving during the whole driving period, through controlling the key parameters of local driving fields. As a type of quantum-state engineering, temporal periodic driving has become a highly controllable and versatile tool in quantum coherence control [17–22], quantum computation [23–26], fundamental effects from quantum optics [27, 28], and many-body systems [29–32]. Although the periodically driven systems have no stationary states usually existing in static systems, they have well defined quasi-stationary-state properties described by the Floquet eigenvalues (also called quasienergies). Through controlling the quasienergy spectrum of periodically



driven systems, more colorful quasi-stationary-state behaviors and more non-trivial effects than the original static cases are expected [33–39].

The power of our approach comes from the employment of strong periodical driving fields, where the spin manipulation frequency greatly exceeds the energy splitting of spins in the picture of radiation-dressed states. The existence of the counter-rotating term of the strong periodical driving brings significant effects on the system, and the induced behavior of avoided quasienergy level crossing of NVCs causes the phenomenon of entanglement resonance between NVCs. The entanglement protection mechanism here is that one of the Floquet states of NVCs is strongly entangled over the whole driving period through adjusting the parameters of driving fields. It could mitigate the hardness in adjusting the energy spectrum of the static system with fixed parameters or the interactions are not sufficiently available. Thanks to the resonant coupling contributed from the avoided level crossing in quasienergy spectrum, we could obtain the entanglement amount inaccessible in static system. More importantly, this type of entanglement could be preserved in the absence of decoherence if the system is initially prepared in this Floquet state, in complete analogy to entangled eigenstates of a static systems [40]. Such capabilities are particularly useful when the weakly interacting spins are spatially separated, providing the opportunity to create long-lasting entanglement to long distances and highly connected quantum networks.

## Model

Consider a pair of magnetic dipole coupled NVCs under the driving of external microwave fields (see figure 1(a)). The Hamiltonian of the NVC  $j$  ( $j = A, B$ ) is

$$\hat{H}_j^{(0)} = D\hat{\delta}_{j,z}^2 + \gamma_e \mathbf{B} \cdot \hat{\mathbf{S}}_j + Q\hat{I}_{j,z}^2 - \gamma_n \mathbf{B} \cdot \hat{\mathbf{I}}_j + \hat{\mathbf{S}}_j \cdot \mathbf{A} \cdot \hat{\mathbf{I}}_j. \quad (1)$$

Here  $\hat{\mathbf{S}}_j = (\hat{S}_{j,x}, \hat{S}_{j,y}, \hat{S}_{j,z})$  and  $\hat{\mathbf{I}}_j = (\hat{I}_{j,x}, \hat{I}_{j,y}, \hat{I}_{j,z})$  are the electronic and nuclear spin operators with  $D$  and  $Q$  being their respective zero-field splitting,  $\mathbf{A} = \text{diag}(A_\perp, A_\perp, A_\parallel)$  are the hyperfine coupling strengths,  $\mathbf{B}$  is the applied magnetic field, and  $\gamma_\alpha = g_\alpha \mu_B$  ( $\alpha = e, n$ ) with  $g_e = 2$ ,  $g_n = 0.4$ , and  $\mu_B$  being the Bohr magneton, are the gyromagnetic ratio of the electron and the nuclei. The dipole–dipole coupling between the two NVCs is [7]

$$\hat{H}^{(1)} = (\mu_0 \gamma_e^2 / 4\pi r^3) [\hat{\mathbf{S}}_A \cdot \hat{\mathbf{S}}_B - 3(\hat{\mathbf{S}}_A \cdot \mathbf{n})(\hat{\mathbf{S}}_B \cdot \mathbf{n})], \quad (2)$$

where  $\mu_0$  is the magnetic permeability,  $\mathbf{r} = \mathbf{r}_m$  is the relative coordinate vector between the two NVCs. We consider  $^{14}\text{N}$  nuclear spin with a quadrupolar moment  $Q = -4.962$  MHz, and the hyperfine coupling strength  $A_\parallel = -2.14$  MHz.

The first two terms of equation (1) define three electronic states (see figure 1(b)) with two degenerate ones  $|m_s = \pm 1\rangle$  lifted by the magnetic field  $\mathbf{B}$ . We approximately treat  $|m_s = -1\rangle \equiv |\uparrow\rangle$  and  $|m_s = 0\rangle \equiv |\downarrow\rangle$  as an effective spin-1/2, by which our three-state system is reduced into a pseudo-two-state one. Supposing that the two NVCs orientate in the same direction and the magnetic field  $\mathbf{B} = B\mathbf{e}_z$ , we have the reduced free Hamiltonian as

$$\hat{H}_j^{(0)} = \omega_0 \hat{\sigma}_{j,z} / 2 + Q\hat{I}_{j,z}^2 - \gamma_n B \hat{I}_{j,z} + \hat{\sigma}_j \cdot \mathbf{A} \cdot \hat{\mathbf{I}}_j, \quad (3)$$

where  $\omega_0 = D - \gamma_e B$  and  $\hat{\sigma}$  are Pauli matrices defined on the two pseudo states. We apply an  $x$ -axis continuous microwave driving field (with strength  $\omega_1$  and frequency  $\omega_0$ ) resonant with the  $m_s = 0, -1$  transition to create new dressed states  $|\pm_x\rangle$  with energy separation  $\omega_1$ , and employ another  $z$ -axis periodic microwave driving field with strength  $\Omega$  and frequency  $\omega$  to manipulate the dressed states. Using the rotating-wave approximation under the condition that  $\omega_1 \gg A_\perp, \omega_1$  to neglect the time-oscillatory terms of equation (3) and the dipole–dipole coupling in the interaction picture, we get the final Hamiltonian

$$\hat{H}_j^{(0)}(t) = \omega_1 \hat{\sigma}_{j,x}/2 + \Omega \cos(\omega t) \hat{\sigma}_{j,z} + Q \hat{I}_{j,z}^2 - \gamma_n B \hat{I}_{j,z} + A_{\parallel} \hat{\sigma}_{j,z} \hat{I}_{j,z}, \quad (4)$$

$$\hat{H}^{(1)} = J[\hat{\sigma}_{A,z} \hat{\sigma}_{B,z} - (\hat{\sigma}_{A,+} \hat{\sigma}_{B,-} + \text{h.c.})], \quad (5)$$

where  $J = \mu_0 \gamma_e^2 [1 - 3(\mathbf{n} \cdot \mathbf{e}_z)^2]/4\pi r^3$ . It describes the dressed NVC with eigenstates  $|\pm_x\rangle$  driven by a periodic longitudinal field  $\Omega$  (see figure 1(c)). Thus one can switch between the weak and strong driving regimes by tuning the ratio between  $\Omega$  and  $\omega_1$ .

## Floquet quasienergy spectrum

Periodically driven system has no meaningful energy spectrum because its energy is not conserved. Thanks to Floquet theorem, it still has well-defined quasienergy spectrum. The theorem states that the system governed by  $\hat{H}(t) = \hat{H}(t + T)$  has a complete set of basis  $|u_\alpha(t)\rangle$  determined by the Floquet equation  $\hat{H}_F |u_\alpha(t)\rangle = \epsilon_\alpha |u_\alpha(t)\rangle$  with  $\hat{H}_F = \hat{H}(t) - i\partial_t$  called Floquet operator such that the evolution of any state can be expanded as  $|\Psi(t)\rangle = \sum_\alpha c_\alpha e^{-i\epsilon_\alpha t} |u_\alpha(t)\rangle$  with  $c_\alpha = \langle u_\alpha(0) | \Psi(0) \rangle$  [41, 42]. The time-independence of  $c_\alpha$  and the evolution factor  $\epsilon_\alpha$  imply that  $\epsilon_\alpha$  and  $|u_\alpha(t)\rangle$  play the same roles in periodic system as the eigenenergies and stationary states in static system. They thus are called quasienergies and quasistationary states, respectively. One can check that  $\epsilon_\alpha$  is periodic with period  $\omega = 2\pi/T$  because  $e^{i\omega t} |u_\alpha(t)\rangle$  are also the eigenstates of  $\hat{H}_F$  with eigenvalues  $\epsilon_\alpha + l\omega$ . Carrying all the quasistationary-state characters of periodic system, the quasienergy spectrum plays a key role in periodic system. Compared with static system, the distinguished character of the periodic system is its good controllability to the quasienergy spectrum via adjusting the external periodic driving.

The Floquet operator  $\hat{H}_F$  acts on the so-called Sambe space made up of the Hilbert space and an extra temporal space [39, 42]. To calculate the quasienergies, we first expand  $|u_\alpha(t)\rangle$  in the complete basis  $\{e^{ik\omega t} |k \in Z\}$  of the temporal space and obtain  $|u_\alpha(t)\rangle = \sum_k |\tilde{u}_\alpha(k)\rangle e^{ik\omega t}$ , by which the Floquet equation is recast into

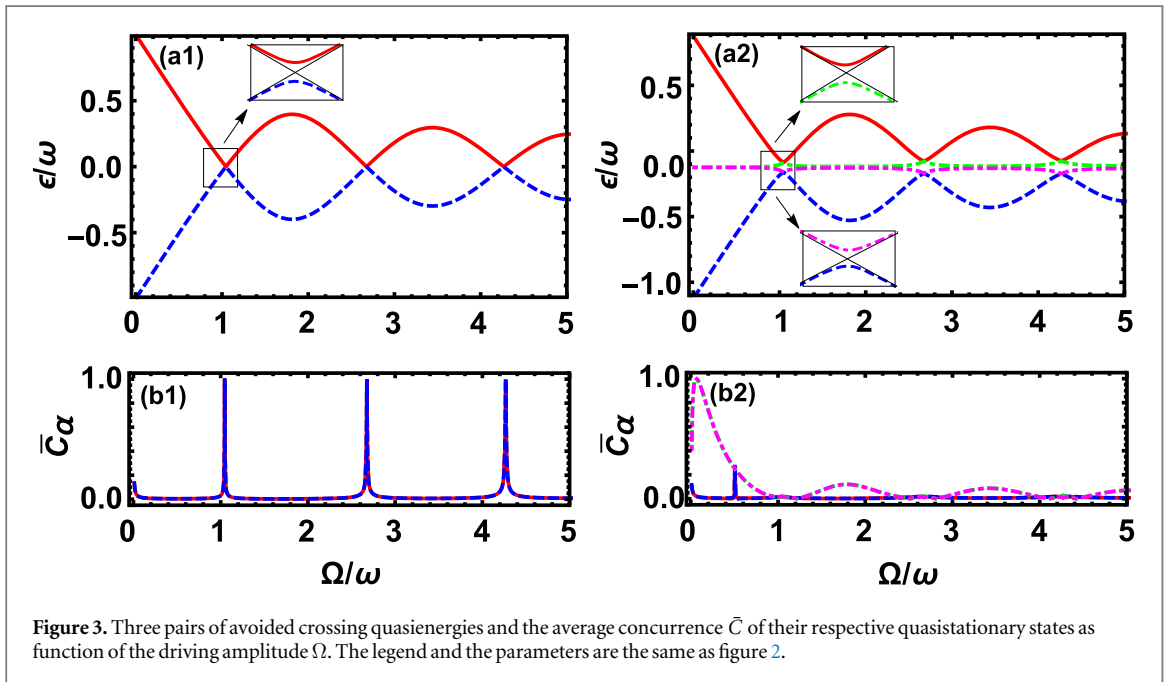
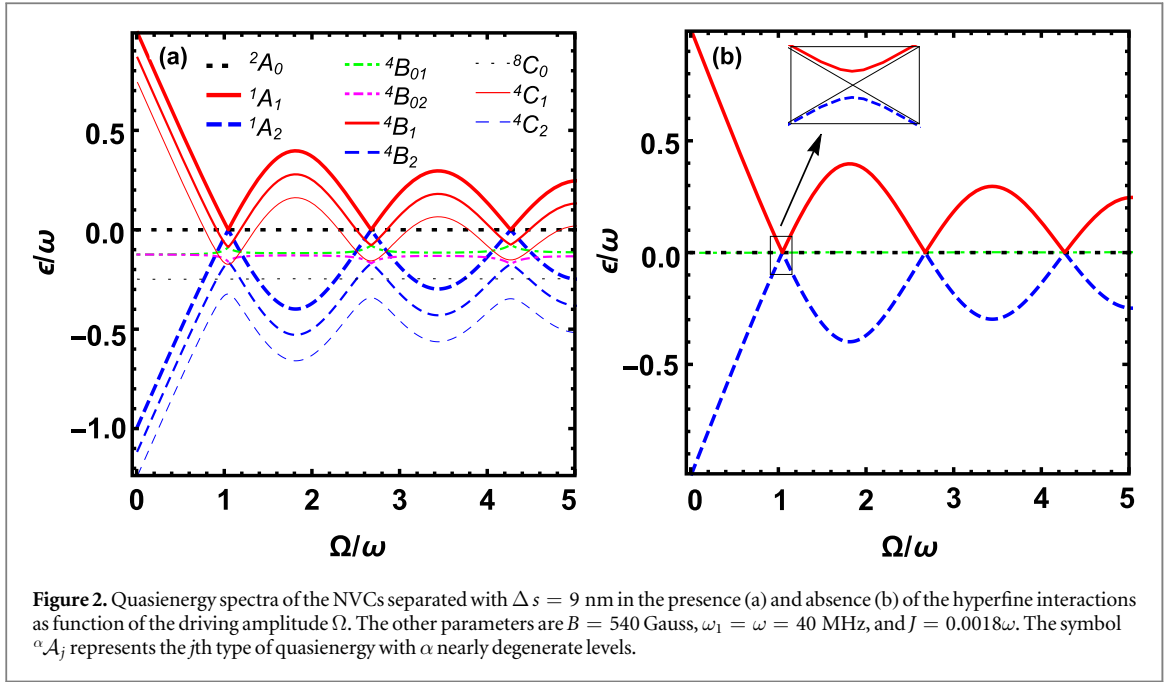
$$\sum_{k \in Z} [\hat{H}_{l-k} + k\omega \delta_{l,k}] |\tilde{u}_\alpha(k)\rangle = \epsilon_\alpha |\tilde{u}_\alpha(l)\rangle, \quad (6)$$

with  $\hat{H}_{l-k} \equiv T^{-1} \int_0^T \hat{H}(t) e^{-i(l-k)\omega t} dt$ . Then expanding each  $\hat{H}_l$  in the complete basis of the Hilbert space, we get an infinite matrix equation. The eigenstates of  $\hat{H}_{j,z}$  are  $|1, m\rangle$  ( $m = \pm 1, 0$ ). Thus the dimension of Hilbert space of the whole system is  $2^2 \times 3^2$ . The quasienergies are obtained by truncating the basis of the temporal space to the rank such that the obtained magnitudes converge.

Figure 2(a) shows the dependence of the obtained 36 quasienergies on the driving amplitude  $\Omega$  by numerically calculating equation (6). The quasienergies in the absence of the hyperfine interactions are plotted in figure 2(b) for comparison. They exhibit linear variation in the weak driving regime with  $\Omega/\omega < 1$ , and change into periodic oscillations with damping tendency in the strong driving regime with  $\Omega/\omega > 1$ , where the rotating-wave approximation breaks down. Compared to figure 2(b), where the quasienergies have an obvious avoided level crossing at certain values of  $\Omega$ , in figure 2(a) with the switching on of the hyperfine interactions, although each quasienergy level has a distinctive splitting, the character of the avoided level crossing is kept. We can figure out three pairs of levels with avoided crossing, i.e.  $A_1$  and  $A_2, B_j$  and  $B_{0j}$  ( $j = 1, 2$ ). It suggests an extremely useful scheme to manipulate the quasienergy spectrum by the external periodic driving. It could lift the experimental difficulty in adjusting the energy spectrum of the system with its parameters unchangeable once the material sample is fabricated.

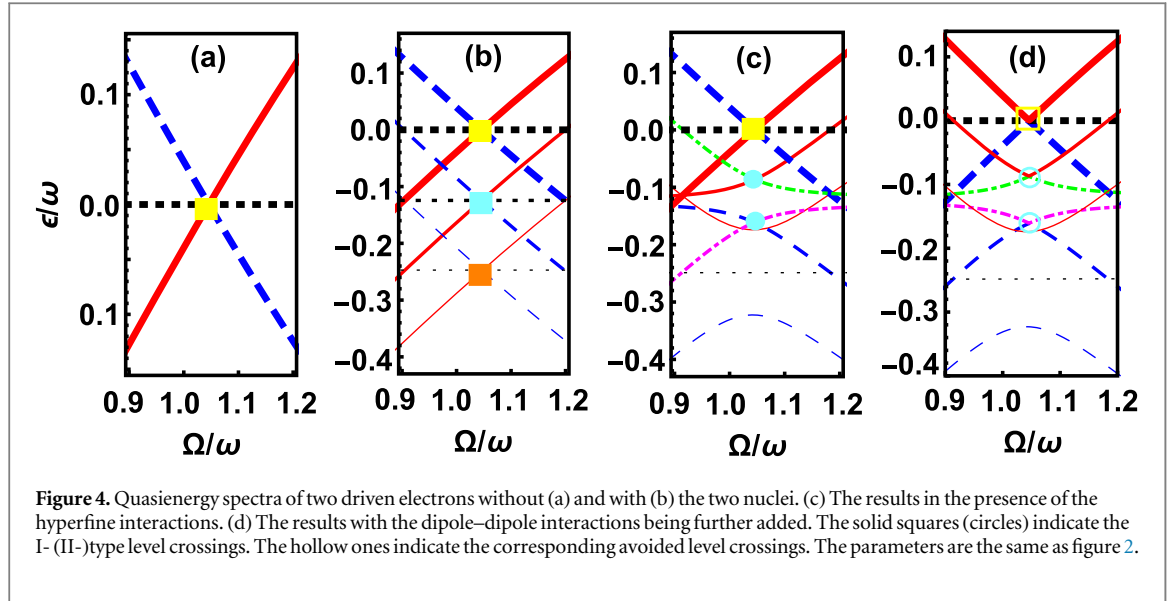
## Entanglement protection

The avoided crossing in the quasienergy spectrum characterizes a resonant coupling between the involved quasistationary states, which plays the essential role in Landau–Zener transition [43] and coherent destruction of tunneling [44, 45]. We here show that it could also be used to generate a maximal entanglement for the two NVCs even when they are far separated. For static system, to generate a long-lasting entanglement, one generally



prepares the system in one of its eigenstates with large entanglement by adjusting the system interactions [40]. The entanglement in the eigenstate, as a stationary state, is dynamically preserved. However, one always confronts the difficulties that the system parameters are hard to adjust and the interactions are not sufficiently available. In our periodically driven system, these difficulties could be solved easily. As long as the system is in a quasistationary state with notable entanglement by appropriately choosing the parameter of external driving field, the entanglement could persist.

We use the average concurrence  $\bar{C}_\alpha = \frac{1}{T} \int_0^T C_\alpha(t) dt$  with  $C_\alpha(t)$  the instantaneous concurrence of  $|u_\alpha(t)\rangle$  during one period to quantify the entanglement of the Floquet state  $|u_\alpha(t)\rangle$  [46]. Here  $\bar{C}_\alpha = 1$  implies that  $|u_\alpha(t)\rangle$  is maximally entangled for all times due to its quasistationary-state nature. To provide a complete picture on how to achieve maximal entanglement of the NVCs, the dependence of  $\bar{C}$  for the three pairs of Floquet states with avoided level crossing on the driving amplitude  $\Omega$  is plotted in figure 3. It is remarkable to see from figures 3(a1) and (b1) that the optimal amplitudes of the periodic driving to achieve the maximal entanglement corresponds exactly to the values occurring the avoided level crossing. This maximal entanglement induced by the periodic driving is called entanglement resonance [47]. It is significant that we can achieve multiple points

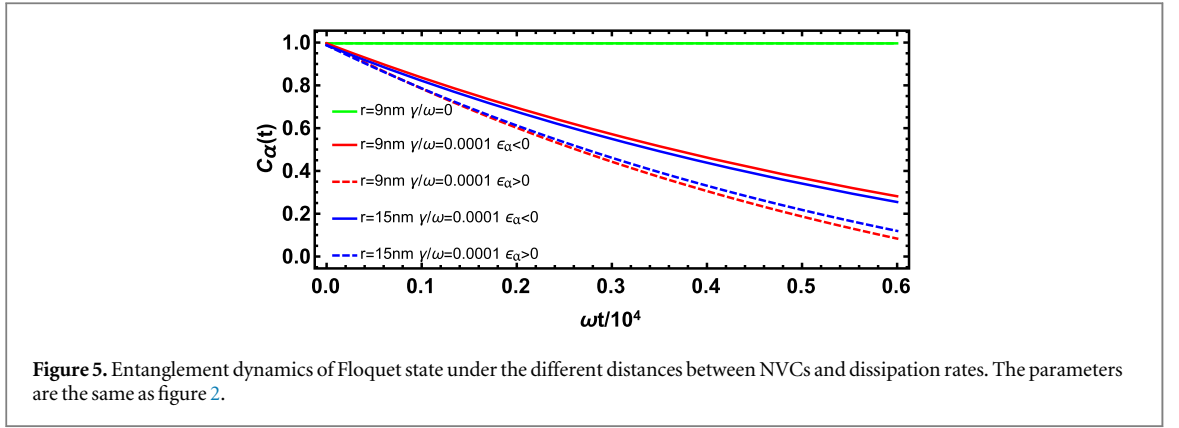


for entanglement resonance under the same static conditions by changing the driving parameters. This is instructive for experiment to engineering the resonance points. Unfortunately, such entanglement resonance is not valid in the other two avoided crossing cases (see figures 3(a2) and (b2)).

To understand the physics behind the entanglement protection and entanglement resonance, we analyze how each avoided level crossing is formed in figure 4. Figure 4(a) shows the quasienergies of two independent periodically driven electrons. We can analytically construct the Floquet states as  $|u_{1,4}(t)\rangle = |\phi_{\pm}(t)\rangle, \phi_{\pm}(t)\rangle$  with  $\epsilon_{1,4} = \pm\mu$  and  $|u_{2,3}(t)\rangle = \{|\phi_{+}(t), \phi_{-}(t)\rangle, |\phi_{-}(t), \phi_{+}(t)\rangle\}$  with  $\epsilon_{2,3} = 0$ , where  $|\phi_{\pm}(t)\rangle \simeq \exp[-i\Omega \sin(\omega t) \hat{\sigma}_z / \omega] |\pm_x\rangle$  are the single-electron Floquet states with the quasienergies  $\pm\mu \simeq \pm\omega_1 \mathcal{J}_0(2\Omega/\omega)$  under the high-frequency driving condition. Here  $\mathcal{J}_0(x)$  is the Bessel function. Details are presented in appendix<sup>7</sup>. The inclusion of the two independent nuclei shifts the electronic quasienergies totally by  $jQ$  ( $j = 0, 1, 2$ ) and form three independent level crossings of quasienergies (see figure 4(b)), which we call I-type level crossings. Here the weak magnetic field term gives negligible contribution. The corresponding Floquet states are  $|u_{\alpha}(t); \Phi_j\rangle$  with  $|\Phi_0\rangle = |0, 0\rangle, |\Phi_1\rangle = \{|0, \pm 1\rangle, |\pm 1, 0\rangle\}$ , and  $|\Phi_2\rangle = \{|1, \pm 1\rangle, |-1, \pm 1\rangle\}$ . Since the hyperfine interactions commute with free nuclei Hamiltonian, they do not cause the mixture of different  $|\Phi_j\rangle$ . The quasienergies in the  $|\Phi_0\rangle$  sector do not change due to the vanishing contribution of the hyperfine interactions. The ones in the  $|\Phi_1\rangle$  sector split into four levels  $Q \pm \mu/2 - (\mu^2/4 + A_{\parallel}^2)^{1/2}$  and  $Q \pm \mu/2 + (\mu^2/4 + A_{\parallel}^2)^{1/2}$  and form two new level crossings, which we call II-type crossing. The former two quasienergies correspond to the Floquet states  $|\phi_{-}(t)\rangle \otimes (|\phi_{+}(t)\rangle \mp |\phi_{-}(t)\rangle) \otimes |0, \pm 1\rangle$  and  $|\phi_{+}(t)\rangle \otimes (|\phi_{+}(t)\rangle \mp |\phi_{-}(t)\rangle) \otimes |0, \pm 1\rangle$  and their permutation between the two electronic and nuclear states at the upper II-type crossing point. The similar forms can also be obtained for the lower crossing point. Our above analysis matches with figure 4(c). When the dipole–dipole interactions are further considered, the I-type crossing in the  $|\Phi_0\rangle$  sector is opened (see figure 4(d)), which causes the superposition of the involved states as  $[|u_1(t)\rangle \pm |u_4(t)\rangle] \otimes |\Phi_0\rangle$ . It is a maximally entangled electronic state. This is the reason of entanglement resonance of the I-type avoided level crossing in figure 3(b1). However, the opening of the II-type crossing by the dipole–dipole interactions achieves the superposition  $(|\phi_{+}(t)\rangle \pm |\phi_{-}(t)\rangle) \otimes (|\phi_{+}(t)\rangle \mp |\phi_{-}(t)\rangle) \otimes |0, \pm 1\rangle$ , which is a product state. Thus no entanglement can be obtained at the II-type avoided level crossing points in figure 3(b2).

We come to conclude that the degeneracy of the non-interacting NVCs is lifted by the weak dipole–dipole interaction, which transforms the NVCs from separable into maximally entangled states at the avoided level crossing point of Floquet levels. We have shown that the peaks of the entanglement resonance seriously depend on the driving field parameters. It allows us to accurately predict the positions of entanglement resonances then control and protect the long-distance entanglement between NVCs in a transparent way, i.e. tailoring the Floquet spectrum through adjusting the strong-driving parameter setting, and to investigate the relevant applications and control protocol in such a realistic system. Note that recent experiment [10] has realized high-fidelity entanglement between two distant NVCs. We think that our idea can also be employed in their setup if

<sup>7</sup> For detailed derivations of the solution of the single-electron Floquet equation and the derivation of the Floquet–Markovian master equation, see appendix.



we changed the weak-driving regime in [10] into the strong-driving regime in our scheme, the entanglement protection of NVCs could be observed used the current technology.

### Effect of decoherence

The NVCs inevitably interact with their outer environments. Such interactions cause the decoherence of NVCs. It is significant to evaluate the performance of our scheme when the decoherence of NVCs is present. We concentrate here on the influence from the dissipation effects of the environments on NVCs, which affect the quantum dynamics and result in the deviation from the desired target states. Considering each NVC weakly coupled to a zero-temperature environment, the Floquet–Markovian master equation reads [48]

$$\begin{aligned} \dot{\rho}(t) &= -i[\hat{H}(t), \rho(t)] + \hat{\mathcal{L}}(t)\rho(t), \\ \hat{\mathcal{L}}(t)\rho(t) &= \sum_{j=1,2} \sum_{\bar{\omega}} \frac{\gamma_j(\bar{\omega})}{2} [2\hat{X}_j(\bar{\omega}, t)\rho(t)\hat{X}_j(\bar{\omega}, t) \\ &\quad - \hat{X}_j(\bar{\omega}, t)^2\rho(t) - \rho(t)\hat{X}_j(\bar{\omega}, t)^2], \end{aligned} \quad (7)$$

where  $\hat{X}_j(\bar{\omega}, t) = \sum_{\alpha,\beta,q} \sum_p \langle \tilde{u}_\alpha(p) | \hat{\sigma}_{j,x} | \tilde{u}_\beta(p+q) \rangle |u_\alpha(t)\rangle \langle u_\beta(t)|$  with  $\bar{\omega} = \epsilon_\beta - \epsilon_\alpha - q\omega$ . Details are presented in (see footnote 7). Figure 5 shows the entanglement dynamics of Floquet state with I-type avoided level crossing through numerically solving this master equation. It can be found that, although the maximal entanglement of the Floquet state shows dynamical decaying with the dissipation considered, the entanglement can still be established in quite a long distance between the two NVCs. Also, these highly entangled Floquet states can be made more robust in the presence of dissipation effect if we suppress these unbeneficial effects as much as we can [49]. The entanglement in this system is most susceptible to environmental noise coupling to the system via the operators  $\hat{\sigma}_{j,z}$ . Since the driving field does not commute with these terms, it automatically suppresses most of the detrimental effects of environmental noise and stabilize the entanglement present in the system. If necessary, the environmental noise can be suppressed further by combining the driving with DD pulses, provided these pulses are short compared to the Floquet period [50, 51]. Other options include additional driving fields that drive transitions between the dressed states. Note that our work is based on recent experimental achievements of strong-driving technology in the NVC system [52–56], which makes our scheme more feasible. The Floquet states have also been experimentally observed in the superconducting flux-qubit system [23] and quantum dot system [57]. This paves the way to realize our proposed Floquet engineering to generate entanglement between remote subsystems via entanglement resonance induced by avoided level crossing.

### 6. Conclusions

In summary, we have proposed a Floquet engineering strategy to protect high-fidelity heralded entanglement between a pair of distant NVCs using the periodic driving in the picture of radiation-dressed states. Our scheme relies firmly on the physical mechanism of entanglement resonance induced by the avoided level crossings in the Floquet quasienergy spectrum of the NVCs. It opens an avenue to create and preserve the entanglement between two exceedingly weakly coupled NVCs by periodic driving. Our results can be generalized to other spin systems and multi-partite quantum systems. Resorting only to the parameter manipulation of the local external driving field on the NVCs, our scheme lifts greatly the experimental difficulty in changing the static parameters of the

system to establish quantum correlation. This offers our scheme an attractive perspective in the application of quantum information processing.

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## Appendix A. Analytical solutions in the limit of high-frequency driving

First, we consider the case that both of the dipole–dipole and the hyperfine interactions of the two NVCs are absent. The two NVCs in this case are independent. Each NVC is periodically driven by an external longitudinal laser field. The corresponding Hamiltonian reads

$$\hat{H}(t) = \hat{H}_0 + \hat{H}_d(t), \quad (8)$$

$$\hat{H}_0 = \frac{\omega_1}{2} \sum_j \hat{\sigma}_{j,x}, \quad (9)$$

$$\hat{H}_d(t) = \Omega \cos(\omega t) \sum_j \hat{\sigma}_{j,z}. \quad (10)$$

According to Floquet theorem, the solutions  $|u_\alpha(t)\rangle$  of the Floquet equation

$$\hat{H}_F |u_\alpha(t)\rangle = \epsilon_\alpha |u_\alpha(t)\rangle \quad (11)$$

with  $\hat{H}_F = \hat{H}(t) - i\partial_t$  characterize the quasi-stationary-state properties of the system. Thus, the corresponding eigenvalues are called quasienergies of the system. To obtain the analytical solution of the Floquet equation, we expand the Floquet state  $|u_\alpha(t)\rangle$  by a complete basis  $\{\hat{U}_t e^{ik\omega t} |k \in Z\}$  in the rotating frame referring to

$$\hat{U}_t = \exp[-i \int_0^t \hat{H}_d(t') dt']$$

$$|u_\alpha(t)\rangle = \sum_k \hat{U}_t e^{ik\omega t} |\tilde{u}_\alpha(k)\rangle. \quad (12)$$

Inserting equation (12) into equation (11), we can obtain

$$\sum_k \hat{H}_{l-k} |\tilde{u}_\alpha(k)\rangle = \epsilon_\alpha |\tilde{u}_\alpha(l)\rangle, \quad (13)$$

$$\hat{H}_{l-k} = \frac{1}{T} \int_0^T e^{i(k-l)\omega t} \hat{U}_t^\dagger \hat{H}_0 \hat{U}_t dt. \quad (14)$$

In the limit of high-frequency driving, we can neglect the fast oscillation terms in  $\hat{H}_{l-k}$ , i.e. the terms with  $l \neq k$ . The Floquet equation is reduced into an eigen equation as a static system

$$\begin{aligned} \hat{H}_0 |\tilde{u}_\alpha(k)\rangle &= \epsilon_\alpha |\tilde{u}_\alpha(k)\rangle, \\ \hat{H}_0 &= \frac{\mu}{2} \sum_j \hat{\sigma}_{j,x}, \end{aligned} \quad (15)$$

where  $\mu = \omega_1 \mathcal{J}_0(2\Omega/\omega)$  and  $\mathcal{J}_0(x)$  is the zeroth order Bessel function of the first kind. Its eigensolution can be readily obtained as

$$\epsilon_{1,4} = \pm\mu, \quad |\tilde{u}_{1,4}(k)\rangle = |\pm_x, \pm_x\rangle, \quad (16)$$

$$\epsilon_{2,3} = 0, \quad |\tilde{u}_{2,3}(k)\rangle = |\pm_x, \mp_x\rangle. \quad (17)$$

The obtained quasienergies  $\epsilon_\alpha$  ( $\alpha = 1, \dots, 4$ ) match the plot shown in figure 4(a) in the main text. Substituting equations (16) and (17) into equation (12), we finally obtain the quasistationary states of the periodic system as

$$|u_{1,4}(t)\rangle = \hat{U}_t |\pm_x, \pm_x\rangle \equiv |\phi_\pm(t), \phi_\pm(t)\rangle, \quad (18)$$

$$|u_{2,3}(t)\rangle = \hat{U}_t |\pm_x, \mp_x\rangle \equiv |\phi_\pm(t), \phi_\mp(t)\rangle. \quad (19)$$

With the consideration of the nuclear degree of freedom, the term  $\hat{H}_1 = \sum_j [Q \hat{I}_{j,z}^2 - \gamma_n B \hat{I}_{j,z}]$  is further added in equation (8). Because  $\hat{I}_{j,z}$  is conserved, the quasistationary states are just the product states of the electrons and nuclei. According the action of  $\hat{H}_1$  on the eigenstates of  $\hat{I}_{j,z}$ , the three quasienergy levels of the NVCs split into three sectors with  $\epsilon_{\pm,0} + lQ$  ( $l = 0, 1, 2$ ) and



$$|u_{\alpha,l}(t)\rangle = |u_{\alpha}(t)\rangle \otimes |\Phi_l\rangle,$$

where the tiny contribution of  $\gamma_n \hat{B}_{j,z}^{\dagger}$  terms has been neglected and  $|\Phi_0\rangle = |0, 0\rangle$ ,  $|\Phi_1\rangle = \{|0, \pm 1\rangle, |\pm 1, 0\rangle\}$ , and  $|\Phi_2\rangle = \{|+1, \pm 1\rangle, |-1, \pm 1\rangle\}$ . The obtained three sectors of quasienergies match the plot shown in figure 4(b) in the main text.

Second, we consider the case when the hyperfine interactions of the NVCs are present. The Hamiltonian is  $\hat{H}_2(t) = \sum_j A_{\parallel} \hat{\sigma}_{j,z} \hat{I}_{j,z}$  is further added in the original Hamiltonian. Because  $[\hat{H}_2(t), \hat{H}_1] = 0$ , the hyperfine interactions do not change the nuclear eigenstates  $|\Phi_l\rangle$ . In the subspace with  $l = 0$ , the total Hamiltonian can be expanded in the complete basis of  $|u_{\alpha}(t)\rangle$  as

$$\hat{h}_0(t) = \begin{pmatrix} \mu & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -\mu \end{pmatrix}. \quad (20)$$

The quasienergies are not changed, as shown in the upper sector of figure 4(c). In the subspace with  $l = 1$ , the Hamiltonian is expanded as

$$\hat{h}_1(t) = \begin{pmatrix} \mu + Q & \pm A_{\parallel} & 0 & 0 \\ \pm A_{\parallel} & Q & 0 & 0 \\ 0 & 0 & Q & \pm A_{\parallel} \\ 0 & 0 & \pm A_{\parallel} & -\mu + Q \end{pmatrix} \quad (21)$$

for  $|0, \pm 1\rangle$ , and

$$\hat{h}'_1(t) = \begin{pmatrix} \mu + Q & 0 & \pm A_{\parallel} & 0 \\ 0 & Q & 0 & \pm A_{\parallel} \\ \pm A_{\parallel} & 0 & Q & 0 \\ 0 & \pm A_{\parallel} & 0 & -\mu + Q \end{pmatrix} \quad (22)$$

for  $|\pm 1, 0\rangle$ . Their eigenvalues are the same, i.e.

$$\bar{\epsilon}_{1,2} = Q \pm \mu/2 - (\mu^2/4 + A_{\parallel}^2)^{1/2}, \quad (23)$$

$$\bar{\epsilon}_{3,4} = Q \pm \mu/2 + (\mu^2/4 + A_{\parallel}^2)^{1/2} \quad (24)$$

For  $|0, +1\rangle$ , the corresponding Floquet states at the crossing point  $\mu = 0$  are

$$|\bar{u}_{1,2}(t)\rangle = |\phi_{\pm}(t)\rangle \otimes (|\phi_{+}(t)\rangle - |\phi_{-}(t)\rangle) \otimes |0, +1\rangle, \quad (25)$$

$$|\bar{u}_{3,4}(t)\rangle = |\phi_{\pm}(t)\rangle \otimes (|\phi_{+}(t)\rangle + |\phi_{-}(t)\rangle) \otimes |0, +1\rangle. \quad (26)$$

The similar forms for the degenerate levels of  $|0, -1\rangle$  and  $|\pm 1, 0\rangle$  can be found. The quasienergies create two crossing points at  $\mu = 0$ , which we call II-type crossing. The four quasienergy levels are shown in the middle sector of figure 4(c). In the subspace with  $l = 2$ , the Hamiltonian reads

$$\hat{h}_2(t) = \begin{pmatrix} \mu + 2Q & \pm A_{\parallel} & A_{\parallel} & 0 \\ \pm A_{\parallel} & 2Q & 0 & A_{\parallel} \\ A_{\parallel} & 0 & 2Q & \pm A_{\parallel} \\ 0 & A_{\parallel} & \pm A_{\parallel} & -\mu + 2Q \end{pmatrix} \quad (27)$$

for  $|+1, \pm 1\rangle$ , and

$$\hat{h}_2(t) = \begin{pmatrix} \mu + 2Q & \pm A_{\parallel} & -A_{\parallel} & 0 \\ \pm A_{\parallel} & 2Q & 0 & -A_{\parallel} \\ -A_{\parallel} & 0 & 2Q & \pm A_{\parallel} \\ 0 & -A_{\parallel} & \pm A_{\parallel} & -\mu + 2Q \end{pmatrix} \quad (28)$$

for  $|-1, \pm 1\rangle$ . Their eigenvalues are  $\{2Q, 2Q \mp (\mu^2 + A_{\parallel}^2)^{1/2}\}$ . No crossing point is created in this case, as shown in the lower sector of figure 4(c).

Third, we consider that the electric dipole–dipole interactions  $\hat{H}^{(1)} = J[\hat{\sigma}_{A,z} \hat{\sigma}_{B,z} - (\hat{\sigma}_{A,+} \hat{\sigma}_{B,-} + \text{h.c.})]$  are present. The three sectors with different nuclear state  $|\Phi_l\rangle$  are not mixed by the dipole–dipole interactions, as it should be. In  $|\Phi_0\rangle$  sector, leaving  $|u_{2,3}(t)\rangle$  unchanged, the dipole–dipole interaction causes the interchange between  $|u_1(t)\rangle$  and  $|u_4(t)\rangle$ . It induces the crossed quasienergies at  $\mu = 0$  opened and thus causes the superposition of  $|u_1(t)\rangle$  and  $|u_4(t)\rangle$ , i.e.  $(|\phi_{+}(t)\rangle, \phi_{-}(t)\rangle \pm |\phi_{-}(t)\rangle, \phi_{+}(t)\rangle)|\Phi_0\rangle$ , which is maximally entangled. We call such maximal entanglement occurring at the avoided level crossing point entanglement resonance. In  $|\Phi_1\rangle$  sector, the dipole–dipole interaction makes the two II-type crossings at  $\mu = 0$  opened and causes the

superposition  $|\bar{u}_1(t)\rangle \pm |\bar{u}_2(t)\rangle$  and  $|\bar{u}_3(t)\rangle \pm |\bar{u}_4(t)\rangle$ , which are product states. Therefore, there is no entanglement resonance at the II-type avoided level crossing points. The dipole–dipole coupling does not induce the avoided level crossing in the  $|\Phi_2\rangle$  sector. Therefore, no notable entanglement is generated in  $|\Phi_2\rangle$  sector.

## Appendix B. Derivation of the Floquet–Markovian master equation

Consider a periodic system weakly interacting with a reservoir. The total Hamiltonian reads

$$\hat{H}(t) = \hat{H}_S(t) + \hat{H}_R + \hat{H}_I,$$

where  $\hat{H}_S(t)$  and  $\hat{H}_R$  are the Hamiltonians of the periodic system and the environment, respectively, and  $\hat{H}_I = \hat{S} \otimes \hat{R}$  describes their interactions. After tracing the reservoir degree of freedom from the unitary dynamics of the whole system, we can obtain the Born–Markovian approximate master equation of the system in the interaction picture as [58]

$$\dot{\rho}_I(t) = - \int_0^\infty d\tau \text{tr}_R[\tilde{H}_I(t), [\tilde{H}_I(t - \tau), \rho_I(t) \otimes \rho_R]], \quad (29)$$

where  $\rho_I(t) = \hat{U}^\dagger(t)\rho(t)\hat{U}(t)$  and  $\tilde{H}_I(t) = \hat{S}(t) \otimes \hat{R}(t)$  with  $\hat{S}(t) = \hat{U}^\dagger(t)\hat{S}\hat{U}(t)$ ,  $\hat{R}(t) = e^{i\hat{H}_R t}\hat{R}e^{-i\hat{H}_R t}$ , and  $\hat{U}(t) = \text{T exp}(-i \int_0^t \hat{H}_S(\tau)d\tau)$ .

To obtain the Floquet–Markovian master equation for the periodic system, we need to expand  $\hat{S}(t)$  in the quasistationary state space [59]. From Floquet theorem, we know that if the system is initially prepared in one Floquet state  $|u_\alpha(0)\rangle$ , it evolves to  $e^{-i\epsilon_\alpha t}|u_\alpha(t)\rangle$ . Thus, we obtain

$$\hat{U}(t)|u_\alpha(0)\rangle = e^{-i\epsilon_\alpha t}|u_\alpha(t)\rangle. \quad (30)$$

Using the completeness relation  $\sum_\alpha |u_\alpha(0)\rangle \langle u_\alpha(0)| = 1$ , one can obtain

$$\hat{U}(t) = \sum_\alpha e^{-i\epsilon_\alpha t}|u_\alpha(t)\rangle \langle u_\alpha(0)|. \quad (31)$$

With equation (31), we have

$$\hat{S}(t) = \sum_{\alpha,\beta} e^{-i(\epsilon_\beta - \epsilon_\alpha)t} \langle u_\alpha(t)|\hat{S}|u_\beta(t)\rangle |u_\alpha(0)\rangle \langle u_\beta(0)|. \quad (32)$$

According to  $|u_\alpha(t)\rangle = \sum_k e^{ik\omega t}|\tilde{u}_\alpha(k)\rangle$ , we further have

$$\begin{aligned} \langle u_\alpha(t)|\hat{S}|u_\beta(t)\rangle &= \sum_{m,n} e^{i(n-m)\omega t} \langle \tilde{u}_\alpha(m)|\hat{S}|\tilde{u}_\beta(n)\rangle \\ &= \sum_{p,q} e^{iq\omega t} \langle \tilde{u}_\alpha(p)|\hat{S}|\tilde{u}_\beta(p+q)\rangle. \end{aligned} \quad (33)$$

Substituting equation (33) into equation (32) and setting  $\hat{S}(t) = \sum_{\bar{\omega}} e^{-i\bar{\omega}t}\hat{S}(\bar{\omega})$ , we have the Fourier transform of  $\hat{S}(t)$  as

$$\hat{S}(\bar{\omega}) = \sum_{\alpha,\beta,q} \sum_p \langle \tilde{u}_\alpha(p)|\hat{S}|\tilde{u}_\beta(p+q)\rangle |u_\alpha(0)\rangle \langle u_\beta(0)| \quad (34)$$

with fixed frequency  $\bar{\omega} = \epsilon_\beta - \epsilon_\alpha - q\omega > 0$  summing over all sets of  $(\alpha, \beta, q)$ . It enables us to cast the interaction Hamiltonian  $\tilde{H}_I(t)$  into

$$\begin{aligned} \tilde{H}_I(t) &= \sum_{\bar{\omega}} e^{-i\bar{\omega}t}\hat{S}(\bar{\omega}) \otimes \hat{R}(t) \\ &= \sum_{\bar{\omega}} e^{i\bar{\omega}t}\hat{S}^\dagger(\bar{\omega}) \otimes \hat{R}(t). \end{aligned} \quad (35)$$

Inserting equation (35) into equation (29), we obtain

$$\begin{aligned} \dot{\rho}_I(t) &= \sum_{\bar{\omega},\bar{\omega}'} e^{i(\bar{\omega}' - \bar{\omega})t} \Gamma(\bar{\omega}) [\hat{S}(\bar{\omega})\rho_I(t)\hat{S}^\dagger(\bar{\omega}') \\ &\quad - \hat{S}^\dagger(\bar{\omega}')\hat{S}(\bar{\omega})\rho_I(t)] + \text{h.c.}, \end{aligned} \quad (36)$$

where  $\Gamma(\bar{\omega}) = \int_0^\infty d\tau e^{i\bar{\omega}\tau} \text{tr}_R[\hat{R}(t)\hat{R}(t - \tau)\rho_R]$  is the Fourier transform of the correlation function of the reservoir. Supposing that the state of the reservoir is stationary, the reservoir correlation function  $\text{tr}_R[\hat{R}(t)\hat{R}(t - \tau)\rho_R]$  is homogeneous in time. It yields

$$\text{tr}_R[\hat{R}(t)\hat{R}(t - \tau)\rho_R] = \text{tr}_R[\hat{R}(\tau)\hat{R}(0)\rho_R], \quad (37)$$

which guarantees  $\Gamma(\bar{\omega})$  to be time-independent. Under the secular approximation, we can neglect the rapid oscillating terms with  $\bar{\omega}' \neq \bar{\omega}$  in the master equation, then we obtain

$$\begin{aligned} \dot{\rho}_I(t) = & \sum_{\bar{\omega}} \Gamma(\bar{\omega}) [\hat{S}(\bar{\omega}) \rho_S(t) \hat{S}^\dagger(\bar{\omega}) \\ & - \hat{S}^\dagger(\bar{\omega}) \hat{S}(\bar{\omega}) \rho_S(t)] + \text{h.c.} \end{aligned} \quad (38)$$

We further decompose the time-independent  $\Gamma(\bar{\omega})$  as

$$\Gamma(\bar{\omega}) = \frac{1}{2} \gamma(\bar{\omega}) + i \lambda(\bar{\omega}). \quad (39)$$

Then the master equation can be written

$$\begin{aligned} \dot{\rho}_I(t) = & -i[\hat{H}_{LS}, \rho_I(t)] + \sum_{\bar{\omega}} \gamma(\bar{\omega}) [\hat{S}(\bar{\omega}) \rho_S(t) \hat{S}^\dagger(\bar{\omega}) \\ & - \frac{1}{2} \{\hat{S}^\dagger(\bar{\omega}) \hat{S}(\bar{\omega}), \rho_S(t)\}], \end{aligned} \quad (40)$$

where  $\hat{H}_{LS} = \sum_{\bar{\omega}} \lambda(\bar{\omega}) \hat{S}^\dagger(\bar{\omega}) \hat{S}(\bar{\omega})$  acts as a Lamb shift, which can be generally neglected. Returning back to the Schrödinger picture, we obtain the Floquet–Markovian master equation

$$\begin{aligned} \dot{\rho}(t) = & -i[\hat{H}_S(t), \rho(t)] + \sum_{\bar{\omega}} \gamma(\bar{\omega}) [\hat{S}(\bar{\omega}, t) \rho(t) \hat{S}^\dagger(\bar{\omega}, t) \\ & - \frac{1}{2} \{\hat{S}^\dagger(\bar{\omega}, t) \hat{S}(\bar{\omega}, t), \rho(t)\}], \end{aligned} \quad (41)$$

where  $\hat{S}(\bar{\omega}, t) = \sum_{\alpha, \beta, q} \langle \tilde{u}_\alpha(p) | \hat{S} | \tilde{u}_\beta(p+q) \rangle | u_\alpha(t) \rangle \langle u_\beta(t) |$  sums over all sets of  $(\alpha, \beta, q)$  satisfying the condition  $\epsilon_\beta - \epsilon_\alpha - q\omega = \bar{\omega} > 0$ .

In our consideration, the system operator interacting with the reservoir is  $\hat{S} = \hat{\sigma}_x$ . We also assume that the reservoir is a white noise with constant spectral density. Thus, we have  $\gamma(\bar{\omega}) = \gamma$ .

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