Nearly sweet spots in capacitively coupled singlet-triplet spin qubits under magnetic field

Guo Xuan Chan,¹ J. P. Kestner,² and Xin Wang¹,*

¹Department of Physics, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong SAR, China, and City University of Hong Kong Shenzhen Research Institute, Shenzhen, Guangdong 518057, China
²Department of Physics, University of Maryland Baltimore County, Baltimore, Maryland 21250, USA

(Dated: November 19, 2020)

Charge noise is the main hurdle preventing high-fidelity operation, in particular that of two-qubit gates, of semiconductor-quantum-dot-based spin qubits. While certain sweet spots where charge noise is substantially suppressed have been demonstrated in several types of spin qubits, the existence of one for coupled singlet-triplet qubits is unclear. We theoretically demonstrate, using full configuration-interaction calculations, that a range of nearly sweet spots appear in the coupled singlet-triplet qubit system when a strong enough magnetic field is applied externally. We further demonstrate that ranking to and from the judiciously chosen nearly sweet spot using sequences based on the shortcut to adiabaticity offers maximal gate fidelities under charge noise and phonon-induced decoherence. These results should facilitate realization of high-fidelity two-qubit gates in singlet-triplet qubit systems.

Introduction.- Singlet-triplet qubits, defined by two-electron spin states confined in semiconductor double-quantum-dot (DQD) devices, are promising candidates for realization of large-scale quantum-dot quantum computation [1–12]. In these systems, the charge noise directly affects the control over the spin qubits and is thus the key obstacle preventing high fidelity quantum control [13–19]. A useful strategy to mitigate charge noise is to operate the qubits near so-called “sweet spots” where the control (e.g. the exchange interaction between spins) is first-order insensitive to charge noise [20–32]. While this strategy has been successfully demonstrated in a variety of single-qubit devices, the existence of any sweet spot, in particular for two singlet-triplet qubits, is far less obvious.

Gate operations on two singlet-triplet qubits coupled by capacitive interactions typically have fidelities ~72% [2] and can be improved to ~90% [33] by applying large magnetic gradient. However, to meet the stringent requirement for quantum error correction, suppression of charge noise becomes emergent. Theoretical calculations [34], particularly using variations of the configuration interaction (CI) method [35–38], are widely employed to search for the sweet spots. Ref. [38] proposes that there exists a sweet spot when the two singlet-triplet qubits are aligned at an appropriate angle, while Ref. [39] claims that a sweet spot may appear at a certain detuning value. However, these results are obtained from the Hund-Mulliken approximation keeping the lowest orbital in each quantum-dot, and it is unclear whether the results hold when higher orbitals are taken into account. Furthermore, Ref. [39] assumed that the charge states of each qubit are independent of each other, but that assumption breaks down in the parameter regime where the sweet spot was claimed to occur. Refs. [40–42], using a more sophisticated CI method either by involving excited orbitals or populating the quantum-dot system with s-type Gaussian functions, have shown that, while a sweet spot may exist for the capacitive two-qubit coupling, it is not at the same time a sweet spot for single-qubit exchange interactions, which limits the usefulness of those prior results in experiments.

All these previous CI calculations were performed without an external magnetic field. In this Letter, we show, using full CI calculations, that a range of nearly sweet spots appear in the coupled singlet-triplet qubit system, when a strong enough magnetic field is applied externally. Around these nearly sweet spots, both the capacitive coupling and the single-qubit exchange interactions are very weakly dependent on the charge noise, making possible high fidelity manipulations. We demonstrate that operating in the nearly-sweet-spot regime yields the entangling gate with fidelity much higher compared to the previous proposals [40–42]. Moreover, the extended range of this nearly-sweet-spot regime allows for application of shortcuts to adiabaticity for the ramping pulses to and from the operating point, which leads to about one order of magnitude improvement in the gate fidelity. Our results should facilitate realization of high-fidelity two-qubit gates in singlet-triplet qubit systems.

Model.- We consider an n-electron system \( H = \sum h_j + \sum \epsilon_j/\epsilon |r_j - r_k| \) with the single-particle Hamiltonian \( h_j = (-i\hbar \nabla_j + e\mathbf{A}/c)^2/2m^* + V(r) + g^* \mu_B \mathbf{B} \cdot \mathbf{S} \). The confinement potential of a double-double-quantum-dot (DDQD) device can be modeled in the \( xy \) plane as (cf. Fig. 1)

\[
V(r) = \frac{1}{2} m^* \omega_n^2 \text{Min} \left( (r - R_1)^2 + \Delta_1, (r - R_2)^2 + \Delta_2, (r - R_3)^2 + \Delta_3, (r - R_4)^2 + \Delta_4 \right),
\]

*Electronic address: x.wang@cityu.edu.hk
where $\mathbf{R}_j = (\pm R \pm x_0, 0)$ are the minima of the parabolic wells.

With each DQD hosting one singlet-triplet qubit, the DDQD defines a pair of capacitively coupled singlet-triplet qubits. The two-qubit logical states are $|SS\rangle, |ST\rangle, |TS\rangle$ and $|TT\rangle$, where $|S\rangle$ and $|T\rangle$ are spin-singlet and unpolarized spin-triplet ($S_z = 0$) states respectively. Without a magnetic field gradient, the system Hamiltonian, $H_{\text{int}}$, is diagonal in the bases of logical states as $[35, 39, 43, 44]$,

$$H_{\text{int}} = J^\text{eff}_L \sigma_z \otimes I + J^\text{eff}_R I \otimes \sigma_z + \alpha \sigma_z \otimes \sigma_z,$$

(2)

where

$$\alpha = \frac{1}{4} (E_{SS} - E_{ST} - E_{TS} + E_{TT}),$$

(3a)

$$J^\text{eff}_L = \frac{1}{4} \left[ E_{TT} - E_{SS} - (E_{ST} - E_{TS}) \right],$$

(3b)

$$J^\text{eff}_R = \frac{1}{4} \left[ E_{TT} - E_{SS} + (E_{ST} - E_{TS}) \right].$$

(3c)

The effective exchange energies $J^\text{eff}_L$ and $J^\text{eff}_R$ for the qubit defined in the left (L) and right (R) DQD respectively, contain both the individual exchange energy of the DQD in absence of the other, as well as a capacitive shift caused by the neighboring DQD. $\alpha$ is the capacitive inter-qubit coupling.

We solve the problem using the full configuration interaction (Full-CI) technique [45], detailed in Sec. I of the Supplemental Material [46]. We use parameters appropriate for GaAs, where the permittivity $\epsilon = 13.1 \varepsilon_0$, effective electron mass $m^* = 0.067 m_e$, confinement strength of the quantum dots $\hbar \omega_0 = 1\text{meV}$, effective Bohr radius $a_B = \sqrt{\hbar/m^*\omega_0} \approx 34\text{nm}$, $x_0 = 2.5 a_B$ and $R_0 = 9 a_B$. The inter-qubit distance $R_0$ is chosen such that the tunneling between qubits is negligible thus only the capacitive coupling remains. Practically, we truncate the Full-CI calculation using a cutoff scheme [45], keeping orbitals up to $n = 4$ Fock-Darwin states.

**Nearly sweet spot.-** We only consider symmetric detuning of two qubits, i.e. the detuning values on both qubits are equal. There are thus three possibilities:

- **“Outer”:** $\Delta_2 = \Delta_4 > 0$, $\Delta_1 = \Delta_3 = 0$,
- **“Center”:** $\Delta_1 = \Delta_3 = \Delta > 0$, $\Delta_2 = \Delta_4 = 0$,
- **“Right”:** $\Delta_1 = \Delta_3 = \Delta > 0$, $\Delta_2 = \Delta_4 = 0$.

In the main text, we focus on the “Outer” scheme where $J^\text{eff}_L = J^\text{eff}_R \equiv J^\text{eff}$, and a discussion on others can be found in Sec. III of the Supplemental Material [46].

Fig. 2(a) and (b) show the dependence of $J^\text{eff}$ and $\alpha$ on detuning $\Delta$ under different magnetic fields, which is the key result of this paper. When $B = 0$, $\alpha$ develops two flat regimes. A sweet spot exists for $\Delta$ around $\Delta \approx 2\text{meV}$, but the same $\Delta$ range does not give any nearly sweet spots in $J^\text{eff}$. This result is consistent with Refs. [40, 41]. Another regime where both $J^\text{eff}$ and $\alpha$ have nearly sweet spots is for $\Delta \gtrsim 2.3\text{meV}$, but ramping to such high detuning would expose the qubit to severe leakage or decoherence, which is therefore impractical. Increasing $B$ moves the sweet spot for $\alpha$ at $\Delta \approx 2\text{meV}$ to the right, while a nearly-sweet-spot regime gradually appears for $J^\text{eff}$ at $B \gtrsim 0.087\text{T}$. At $B = 0.104\text{T}$, the nearly-sweet-spot regime where both $J^\text{eff}$ and $\alpha$ are very weakly dependent on $\Delta$ is quite extended, as indicated by the cyan area. At the same time, the $\alpha$ value is enhanced so as to reduce the gate time and minimize the accumulation of gate error. We shall see later that the detuning $\Delta^c$ yields the highest gate fidelity. We also note that when $\alpha$ reaches its maximal value, $\tilde{\alpha}$, $\partial \alpha / \partial \Delta = 0$, while at the same $\Delta$ value $\partial J^\text{eff} / \partial \Delta$ is small ($\sim 10^{-2}$) but not exactly zero (cf. Fig. 3(d)). This is the reason we call the region nearly sweet spots.

Fig. 2(c) and (d) show the energy level structure of the
system as the detuning is varied. The states are labeled using a Dirac ket with the first entry being the state of the left DQD and the second the right DQD. The state of one qubit (i.e., one DQD) is either a singlet (S) or a triplet (T) with the superscript showing the charge configurations. For example, the four-electron state shown in Fig. 1 can be understood as $|S^{20}T^{02}\rangle$ [47]. Detailed discussions of all relevant states in terms of the extended Hubbard model can be found in Sec. II of the Supplemental Material [46].

Fig. 2(c) shows the energy levels at zero magnetic field. All levels are parallel for $\Delta \gtrsim 2.3$ meV, consistent with the observation that both $J_{\text{eff}}$ and $\alpha$ are weakly dependent on $\Delta$ in this range. Around $\Delta \approx 2$ meV, the slopes of the curves can be combined in the fashion of Eq. (3a), implying that $\partial \alpha/\partial \Delta \approx 0$, but not for $J_{\text{eff}}$ (Eqs. (3b) and (3c)), consistent with the observations from Fig. 2(a) and (b). When a magnetic field $B = 0.104$ T is applied, however, the situation changes. Two new states becomes significant: a bonding state $|SS\rangle$, $\Delta^\ast$ and an anti-bonding state $|TT\rangle$, $\Delta^\ast$, $|\tilde{SS}\rangle$, $\Delta^\ast$ and $|\tilde{TT}\rangle$, $\Delta^\ast$ as functions of magnetic field in the “Outer”, “Right” and “Center” detuning scheme. For both $J_{\text{eff}}$ and $\alpha$, $\tilde{\alpha}$, in the nearly-sweet-spot regime, as well as $\partial J_{\text{eff}}/\partial \Delta$ evaluated at the same $\Delta$ value where $\alpha$ reaches maximum, for the “Outer” and “Right” schemes. For both schemes, $\partial J_{\text{eff}}/\partial \Delta$ is as small as $\sim 10^{-2}$ for $B \gtrsim 0.1$ T, indicating that the susceptibility to charge noise is extremely weak. On the other hand, $\tilde{\alpha}$ is much greater for the “Outer” scheme than the “Right” one, suggesting that the “Outer” scheme remains the optimal protocol to operate the coupled DQD systems.

**CPHASE gate.** The inter-qubit coupling, $\sigma_2 \otimes \sigma_z$, gives rise to a controlled-phase (CPHASE) gate [40, 41]. The system is initialized at $\Delta_{\text{init}}$ where $\alpha$ is negligible, and is then ramped to a larger detuning, $\Delta_{\text{op}}$, where the operation is performed with a reasonably strong $\alpha$. This ramping time is denoted as $\tau_{\text{ramp}}$. After operating at $\Delta_{\text{op}}$ for a time $\tau_{\text{op}}$, the system is brought back to $\Delta_{\text{init}}$ in $\tau_{\text{ramp}}$ (see Fig. 4(a)). The total gate time is therefore $\tau = 2\tau_{\text{ramp}} + \tau_{\text{op}}$.

The evolution of the system in the logical subspace can be described by the master equation,

$$
\dot{\rho} = -i[H_{\text{int}}, \rho] + (\gamma_{\varphi_2} + \gamma_{\text{dep}_2}) D[\sigma_z \otimes I] \rho + (\gamma_{\varphi_2} + \gamma_{\text{dep}_2}) D[I \otimes \sigma_z] \rho + (\gamma_{\varphi_3} + \gamma_{\text{dep}_3}) D[\sigma_z \otimes \sigma_z] \rho + \sum_{j<k} \gamma_{\text{rel},jk} D[\sigma_j \sigma_k] \rho,
$$

where $\gamma_{\varphi_{2,3}}$ and $\gamma_{\text{dep}_{2,3}}$ are the charge-noise dephasing rates for qubit L ($\mathbb{R}$) and the capacitive coupling $\alpha$, respectively. All of them are proportional to a reference charge-noise dephasing rate $\tilde{\gamma}_{\varphi}$ [46], which we shall use as our noise amplitude. $\gamma_{\text{rel}}$ ($\gamma_{\text{dep}}$) is the phonon-mediated relaxation (pure dephasing) rate. $D[c]$ represents the dis-
sipation superoperator $\mathcal{D}[^c]\rho \equiv 2c\rho c^\dagger - c^\dagger c^\dagger c^2 - \rho c^\dagger c^2$ [48]. More details, including the derivation of the decoherence rates listed above can be found in Sec. V of the Supplemental Material [46].

We have chosen $\Delta_{a,b,c,d}^{\text{op}}$ as candidates of $\Delta_{\text{op}}$ (as indicated on Fig. 2). The $\alpha$ values as well as $\partial J_{\text{eff}}/\partial \Delta$ for these points are summarized in Table 1. On one hand, $\partial J_{\text{eff}}/\partial \Delta$ is small for $\Delta_c^{\text{op}}$, $\Delta_d^{\text{op}}$, suggesting that the charge-noise-induced dephasing is suppressed. On the other hand, ramping the system to $\Delta_{a,b,c,d}$ requires less detuning sweeps compared to $\Delta_{b,d}^{\text{op}}$, suggesting that within the same $\tau_{\text{ramp}}$, choosing $\Delta_{a,b,c,d}^{\text{op}}$ as the operating points limits the leakage. These considerations imply that $\Delta_c^{\text{op}}$ is the optimal choice as $\Delta_{\text{op}}$.

We consider two ways of detuning the DDQD system from $\Delta_{\text{init}}$ to $\Delta_{\text{op}}$: a linear (LIN) ramping scheme where $d\Delta/dt = \text{constant}$, as well as one based on shortcut to adiabaticity (STA) [49, 50] (see Fig. 3(a)). It is noticed that for $B = 0.104$ T, charge transitions of different logical states are either located at the same $\Delta$ (facilitated by the same inter-dot tunneling) or well-separated in $\Delta$ values (see Fig. 2(d)), allowing us to apply concatenated STA pulse sequences, the details of which can be found in Sec. VII of the Supplemental Material [46]. The application of STA pulse sequences allows the reduction of the total gate time $\tau$, without increasing the leakage, therefore suppressing decoherence. Note that STA pulse sequence is not available for $\Delta_{a,b}^{\text{op}}$ as the charge transitions occur very closely in $\Delta$ and cannot be individually addressed for different logical states (see Fig. 2(c)).

We numerically simulate the master equation, Eq. (4), taking into account the leakage by expanding $H_{\text{init}}$ into the effective Hamiltonian block for each logical eigenstate [46]. The results of gate infidelities, $1 - F$ [51], as functions of $\tau$ and $\gamma_{\phi}$ are shown in Fig. 4(b) and (c) respectively. From Fig. 4(b), we see a reduction of infidelities at small $\tau$ for all results, but STA with operating point $\Delta_c^{\text{op}}$ gives the lowest infidelity at the shortest gate operation time, while LIN with $\Delta_c^{\text{op}}$ gives the second lowest infidelity. When $\tau$ is large, the infidelities increase with $\tau$ due to accumulated exposure to various decoherence channels other than leakage, as expected. Fig. 4(c) shows the gate infidelities as functions of the reference charge-noise dephasing rate $\gamma_{\phi}$, with the gate time $\tau$ for each set of results chosen such that it produces the minimal gate infidelity as indicated in panel (b). We see that in the LIN scheme, results calculated at $\Delta_c^{\text{op}}$ exhibit about a factor $2 \sim 4$ reduction in infidelity compared to other $\Delta_{\text{op}}$ values, while using STA scheme offer another factor of $2 \sim 4$. Therefore the STA scheme in combination with the nearly sweet spot offers roughly an order of magnitude reduction in infidelities.

Conclusions.- We have shown, using Full CI calculations, that a range of nearly sweet spots, for both the single-qubit exchange energy as well as the capacitive coupling, appear in the coupled singlet-triplet qubit system under a strong enough external magnetic field. This range of nearly sweet spots arises due to the appearance of $|SS\rangle$ and $|TT\rangle$ states under magnetic field, which occupy detuning ranges that increase with the magnetic field. We further demonstrate that ramping to and from the judiciously chosen nearly sweet spot using sequences based on the shortcut to adiabaticity offers maximal gate fidelities under charge noise and phonon-induced decoherence. Our results should facilitate realization of high-fidelity two-qubit gates in coupled singlet-triplet qubit systems.

Acknowledgements.- G.X.C. and X.W. are supported by the Key-Area Research and Development Program of GuangDong Province (Grant No. 2018B030326001), the National Natural Science Foundation of China (Grant No. 11874312), the Research Grants Council of Hong Kong (Grant Nos. 11303617, 11304018, 11304920), and the Guangdong Innovative and Entrepreneurial Research Team Program (Grant No. 2016ZT06D348). J. P. K. acknowledges support from the National Science Foundation under Grant No. 1915064.

Table 1: Summary of the parameters for different $\Delta_{\text{op}}$.

| $B$ (T) | $\Delta_{\text{op}}$ (ns) | $|\alpha|$ (μeV) | $\partial J_{\text{eff}}/\partial \Delta$ |
|--------|-------------------|-----------------|---------------------|
| 0      | $\Delta_a^{\text{op}}$ | 3.84            | $4.94 \times 10^{-1}$ |
| 0.104  | $\Delta_b^{\text{op}}$ | 70.32           | $1.74 \times 10^{-3}$ |
|        | $\Delta_c^{\text{op}}$ | 57.24           | $7.50 \times 10^{-3}$ |
|        | $\Delta_d^{\text{op}}$ | 75.24           | $7.96 \times 10^{-3}$ |

[1] J. R. Petta, A. C. Johnson, J. M. Taylor, E. A. Laird, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Science 309, 2180 (2005).
[2] M. D. Shulman, O. E. Dial, S. P. Harvey, H. Bluhm, V. Umansky, and A. Yacoby, Science 336, 202 (2012).
[3] J. Levy, Phys. Rev. Lett. 89, 147902 (2002).
[4] X. Wu, D. R. Ward, J. R. Prance, D. Kim, J. K. Gamble, R. T. Mohr, Z. Shi, D. E. Savage, M. G. Lagally, M. Friesen, S. N. Coppersmith, and M. A. Eriksson, Proc. Natl. Acad. Sci. U.S.A. 111, 11938 (2014).
[5] B. M. Maune, M. G. Borselli, B. Huang, T. D. Ladd, P. W. Deelman, K. S. Holabird, A. A. Kiselev, I. Alvarado-Rodriguez, R. S. Ross, A. E. Schmitz, M. Sokolich, C. A. Watson, M. F. Gyure, and A. T. Hunter, Nature (London) 481, 344 (2012).
[6] C. Barthel, J. Medford, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Phys. Rev. Lett. 105, 266808 (2010).
[7] Z. Shi, C. B. Simmons, J. R. Prance, J. King Gamble, M. Friesen, D. E. Savage, M. G. Lagally, S. N. Coppersmith, and M. A. Eriksson, Appl. Phys. Lett. 99, 233108 (2011).
[8] K. Takeda, A. Noiri, J. Yoneda, T. Nakajima, and S. Tarucha, Phys. Rev. Lett. 124, 117701 (2020).
[9] P. Cerfontaine, T. Botzem, J. Ritzmann, S. S. Humphol, A. Ludwig, D. Schuh, D. Bougeard, A. D. Wieck, and H. Bluhm, Nat. Commun. 11, 4414 (2020).
[10] K. Eng, T. D. Ladd, A. Smith, M. G. Borselli, A. A. Kiselev, B. H. Fong, K. S. Holabird, T. M. Hazard, B. Huang, P. W. Deelman, I. Milosavljevic, A. E. Schmitz, R. S. Ross, M. F. Gyure, and A. T. Hunter, Sci. Adv. 1, e1500214 (2015).
[11] A. Noiri, T. Nakajima, J. Yoneda, M. R. Delbecq, P. Stano, T. Otsuka, K. Takeda, S. Amaha, G. Allioun, K. Kawasaki, Y. Kojima, A. Ludwig, A. D. Wieck, D. Loss, and S. Tarucha, Nat. Commun. 9, 5066 (2018).
[12] P. Harvey-Collard, R. M. Jock, N. T. Jacobson, A. D. Gardner, M. J. Manfra, and A. Yacoby, npj Quantum Inf. 3, 3 (2017).
[13] G. Cao, H.-O. Li, G.-D. Yu, B.-C. Wang, B.-B. Chen, X.-X. Song, M. Xiao, G.-C. Guo, H.-W. Jiang, X. Hu, and G.-P. Guo, Phys. Rev. Lett. 116, 096801 (2016).
[14] Z. Shi, C. B. Simmons, D. R. Ward, J. R. Prance, X. Wu, T. S. Koh, J. K. Gamble, D. E. Savage, M. G. Lagally, M. Friesen, S. N. Coppersmith, and M. A. Eriksson, Nat. Commun. 5, 3020 (2014).
[15] M. Russ and G. Burkard, J. Phys. Condens. Matter 29, 393001 (2017).
[16] F. K. Malinowski, F. Martins, P. D. Nissen, S. Fallahi, G. C. Gardner, M. J. Manfra, C. M. Marcus, and F. Kuemmeth, Phys. Rev. B 96, 045443 (2017).
[17] Y.-P. Shim and C. Tahan, Phys. Rev. B 97, 155402 (2018).
[18] J. M. Nichol, L. A. Onra, S. P. Harvey, S. Fallahi, G. C. Gardner, M. J. Manfra, and A. Yacoby, npj Quantum Inf. 3, 3 (2017).
[19] J. M. Taylor, H. A. Engel, W. Dür, A. Yacoby, C. M. Marcus, P. Zoller, and M. D. Lukin, Nat. Phys. 1, 177 (2005).
[20] D. Stepanenko and G. Burkard, Phys. Rev. B 75, 085324 (2007).
[21] V. Srinivasa and J. M. Taylor, Phys. Rev. B 92, 235301 (2015).
[22] R. Li, X. Hu, and J. Q. You, Phys. Rev. B 86, 205306 (2012).
[23] S. Yang and S. Das Sarma, Phys. Rev. B 84, 121306 (2011).
[24] M. A. Wolfe, F. A. Calderon-Vargas, and J. P. Kestner, Phys. Rev. B 96, 201307 (2017).
[25] E. Nielsen, R. P. Muller, and M. S. Carroll, Phys. Rev. B 85, 035319 (2012).
[26] T. Hiltunen and A. Harju, Phys. Rev. B 90, 125303 (2014).
[27] D. Buterakos, R. E. Throckmorton, and S. Das Sarma, Phys. Rev. B 100, 075411 (2019).
[28] G. Ramon, Phys. Rev. B 84, 155329 (2011).
[29] F. A. Calderon-Vargas and J. P. Kestner, Phys. Rev. B 91, 035301 (2015).
[30] E. Barnes, J. P. Kestner, N. T. T. Nguyen, and S. Das Sarma, Phys. Rev. B 84, 235309 (2011).
[31] See Supplemental Material for details.
[32] There are complications involving the \(T^{(2)}\) state and all such notations in the main text should be understood as two electrons occupying the lowest two levels respectively, i.e. \(T^{(2)}\) in \([46]\).
[33] H. M. Wiseman and G. J. Milburn, Quantum Measurement and Control (Cambridge University Press, Cam-
Supplemental Material for “Nearly sweet spots in capacitively coupled singlet-triplet spin qubits under magnetic field”

In this Supplemental Material we provide necessary details complementary to results shown in the main text.

I. FULL CONFIGURATION INTERACTION (FULL-CI) CALCULATION

A. Single particle basis states

The Hamiltonian for $N$ electrons confined in a potential $V(r)$, in the presence of a uniform magnetic field $B = B\hat{z} = \nabla \times A$, is

$$H = \sum_{j=1}^{N} \left[ \frac{-i\hbar \nabla_j + eA/c}{2m^*} + V(r_j) \right] + \sum_{j<k} \frac{\epsilon^2}{\epsilon|r_j - r_k|} + g^* \mu_B B \cdot S,$$

(S-1)

where $m^*$ is the electron effective mass, $\epsilon$ is the permittivity of the semiconductor material, $g^*$ is the effective $g$ factor, $\mu_B$ is the Bohr magneton, and $S$ is the total electronic spin. Assuming the confining potential yields a quadratic in-plane potential for electrons in a lateral gate-defined confinement of a quantum dot,

$$V_{R_0}(x,y) = \frac{1}{2} m^* \omega^2 \left[(x-x_0)^2 + y^2 \right],$$

(S-2)

where the vector $R_0 = (x_0,0)$ is the position of potential minimum. The solution to the single-particle Hamiltonian, $\frac{p^2}{2m^*} + V(r)$, are then the Fock-Darwin (F-D) states centered at the minimum of the potential well,

$$\phi_{nm}(x,y) = \frac{1}{l_0} \sqrt{\frac{(n-\frac{1}{2})!}{\pi (n+\frac{1}{2})!}} \frac{|n|}{l_0} e^{-\frac{(x-x_0)^2+y^2}{2l_0^2}} L_n^{m-\frac{1}{2}} L_n^{m+\frac{1}{2}} \frac{(x-x_0)^2+y^2}{l_0^2},$$

(S-3)

where $l_0 = l_B/(1/4 + \omega_0^2/\omega_c^2)^{1/4}$, $l_B = \sqrt{\hbar c/eB}$, $\omega_c = eB/m^*c$, $L_n^m(x)$ is the associated Laguerre polynomial and we have adopted the symmetric gauge, $A = B/2 [-y\hat{x} + (x+x_0)\hat{y}]$. The corresponding single-particle energies of Eq. (S-3) are

$$E_{n,m} = (n+1) \sqrt{\frac{1}{4} + \frac{\omega_c^2}{\omega_0^2}} \hbar \omega_c + \frac{m}{2} \hbar \omega_c.$$

(S-4)

The derivation of the Fock-Darwin states, Eq. (S-3), and the corresponding energy spectrum can be found in [S1].

Taking electron spin into account, the spin orbitals are given by

$$\Phi_{nm\sigma}(r) = \phi_{nm}(r)\sigma(\omega),$$

(S-5)

where $\sigma$ denotes the electron spin, $\uparrow$ or $\downarrow$, and $\omega$ is the spin variable.
B. Multi-particle Slater determinant

The two electron multi-particle bases for a single DQD is a Slater determinant of relevant single particle spin orbitals,

\[ |\Psi_L\rangle = |\Phi_{L,1}^\uparrow \Phi_{L,2}^\downarrow\rangle, \]
\[ |\Psi_R\rangle = |\Phi_{R,1}^\uparrow \Phi_{R,2}^\downarrow\rangle, \]

(S-6)

where \( |\Phi_{L,j}^\sigma\rangle \) (\( |\Phi_{R,j}^\sigma\rangle \)) refers to the single-particle Fock Darwin state in the left (right) DQD of the \( j \)th electron with spin \( \sigma \). For a DDQD device, the four-electron multi-particle bases are constructed by a direct product of the left and right DQD two-particle states,

\[ |\Psi\rangle = |\Phi_{L,1}^\uparrow \Phi_{L,2}^\downarrow\rangle |\Phi_{R,1}^\uparrow \Phi_{R,2}^\downarrow\rangle. \]

(S-7)

C. General procedure for Full-CI calculation

The general procedure to perform Full-CI calculation is shown in Fig. S1. The calculation loop is escaped when the newly obtained ground state energy is converged (see below).

D. Truncation and convergence

In principle, the Full-CI calculation should include an infinite number of electronic orbitals which is intractable. In practice, one introduces a cutoff energy, defined as the total non-interacting energy of the system above the ground state configuration [S1]. The convergence is verified by raising the cutoff energy and examining the change in relevant physical quantities.

Our verification of convergence is specific to the quantum-dot device parameters chosen in this paper: \( \hbar \omega_0 = 1 \) meV, \( x_0 = 2.5a_B \), \( R_0 = 9a_B \), \( B = 0 \). In Fig. S2, we demonstrate that orbitals included in the Full-CI calculation up until 4 meV is sufficient to achieve convergence. This implies that the orbitals required are up until \( n = 4 \) Fock-Darwin states, corresponding to 60 orbitals for a DDQD system and \( \approx 1.6 \) million number of Coulomb interaction terms to be evaluated. Results shown in this paper are therefore obtained by keeping relevant orbitals up to \( n = 4 \).

II. EXTENDED HUBBARD MODEL

A. Hamiltonian

Although the Full-CI calculations allow us to obtain an accurate description of the energy spectrum of a DDQD device, it is computationally forbiddingly expensive to simulate a range of parameters. The extended Hubbard model
Supplementary Figure S2: (a) Effective single-qubit exchange energy, \( J_{\text{eff}}^L = J_{\text{eff}}^R \) as a function of cutoff energy. (b) Ground state energy, \( E_{(GS)} \) as a function of the cutoff energy. \( \Delta = 0 \).

Supplementary Figure S3: Schematic figure of Fock-Darwin states in a four-quantum-dot device.

on one hand gives us a computationally efficient way to calculate physical quantities of the system, and on the other hand reveals important insights on relevant eigenstates involved in our problem. The Hamiltonian is,

\[
H = \sum_{j,\sigma} \varepsilon_j \sigma c_j^{\dagger} c_j + \sum_{j,k,\sigma} t_{j\sigma k} \sigma c_j^{\dagger} c_k \sigma + \text{H.c.} \right) + \sum_j U_j n_j^{\dagger} n_j + \sum_{\sigma, \sigma'} \sum_{j<k} U_{j\sigma k}^{\sigma'} n_j^{\sigma} n_k^{\sigma'}, \tag{S-8}
\]

where \( j \) and \( k \) are orbital indices (cf. Fig. S3), while \( \sigma \) and \( \sigma' \) are spins. The summations over orbitals \((j, k)\) are from 1 to 12 and spins \((\sigma, \sigma')\) are for up and down. \( \varepsilon_j \sigma \) denotes the on-site energy at dot \( j \), \( t_{j\sigma k} \sigma \) the tunneling between the \( j \)th and \( k \)th orbital, while \( U_j \) denotes the on-site Coulomb interaction in the \( j \)th orbital and \( U_{j\sigma k} \) the Coulomb interaction between the \( j \)th and \( k \)th orbital. To avoid confusion of the Coulomb interaction between the 1st and 2nd orbital, \( U_{12} \), and the on-site Coulomb interaction on the 12th orbital (also labelled as \( U_{12} \)), we take the symmetry into consideration. We use \( U_{34} \) to label the former one (inter-site Coulomb interaction) in all situations (“Outer”, “Center”, “Right”) as they are equal to each other. For the latter one (on-site Coulomb interaction), we use \( U_9 \) in the “Outer” and “Center” cases, and \( U_{10} \) in the “Right” case. This is clear from Fig. S3.

Theses parameters are calculated from,

\[
U_{j\sigma k} = \int \Phi_j^{\sigma}(r_1) \Phi_k^{\sigma}(r_2) \frac{e^2}{\kappa |r_1 - r_2|} \Phi_j(r_1) \Phi_k(r_2) dr^2, \\
t_{j\sigma k} = \int \Phi_j^{\sigma}(r) \left[ \frac{\hbar^2}{2m^*} \nabla^2 + V(r) \right] \Phi_k(r) dr, \\
\varepsilon_j = \int \Phi_j^{\sigma}(r) \left[ \frac{\hbar^2}{2m^*} \nabla^2 + V(r) \right] \Phi_j(r) dr. \tag{S-9}
\]

Since the Hamiltonian, Eq. (S-8), commutes with \( S^2 \), each logical manifold forms a diagonal block, allowing us to evaluate the eigenvalues of \( |SS\rangle, |ST\rangle, |TS\rangle \) and \( |TT\rangle \) by diagonalizing the corresponding individual blocks. In this section, we focus on the effective Hamiltonian blocks for the “Outer” detuning case. Results can also be obtained
using Eq. (S-8) for “Center” and “Right” detuning cases, but we will not present the results in this section. In the “Outer” detuning case, the symmetry around the center of the DDQD system allows us to simplify some parameters, e.g. $\varepsilon_1 = \varepsilon_4$, inter-site Coulomb interaction $U_{12} = U_{34}$ as mentioned above, $U_{16} = U_{47}$ etc. When treating the blocks solely relevant to the singlet $|S\rangle$ (i.e. the $|SS\rangle$ states), we keep orbitals 1 through 4 only as contribution from higher orbitals can be safely neglected. However, all 12 orbitals are retained whenever the triplet state plays a role.

We assume $U_1 = U_2 = U_3 = U_4 = U_{os}$ (where “os” means on-site) and $t_{12\sigma} = t_{34\sigma} = t$ for convenience. However, $U_j \neq U_k$ for $j$ from 1 to 4 and $k$ from 5 to 12, due to different forms of wave functions with the former yielding even Gaussian functions, and the later being odd oscillating functions. Also, it should be pointed out that the single-particle tunneling between DDQDs is assumed to be zero due to the large distance $R_0$ between them, i.e. $t_{jk} = 0$ when the $j$th and $k$th orbitals belong to different DDQDs.

The Hamiltonian block in each logical manifold is written in the bases of four-electron multi-particle states, constructed from the direct product of two-electron Slater determinants, Eq. (S-7). We therefore write the relevant two-electron Slater determinants as:

\[
|S^{20}\rangle = |\Phi_{1\uparrow}\Phi_{4\downarrow}\rangle,
\]

\[
|S^{02}\rangle = |\Phi_{3\uparrow}\Phi_{4\downarrow}\rangle,
\]

\[
|S^{11}\rangle = \frac{1}{\sqrt{2}}(|\Phi_{1\uparrow}\Phi_{2\downarrow}\rangle + |\Phi_{2\uparrow}\Phi_{1\downarrow}\rangle) = \frac{1}{\sqrt{2}}(|\Phi_{3\uparrow}\Phi_{4\downarrow}\rangle + |\Phi_{4\uparrow}\Phi_{3\downarrow}\rangle),
\]

\[
|T^{11}\rangle = \frac{1}{\sqrt{2}}(|\Phi_{1\uparrow}\Phi_{2\downarrow}\rangle - |\Phi_{2\uparrow}\Phi_{1\downarrow}\rangle) = \frac{1}{\sqrt{2}}(|\Phi_{3\uparrow}\Phi_{4\downarrow}\rangle - |\Phi_{4\uparrow}\Phi_{3\downarrow}\rangle),
\]

\[
|T^{20}_{S}\rangle = \frac{1}{\sqrt{2}}(|\Phi_{1\uparrow}\Phi_{5\downarrow}\rangle - |\Phi_{5\uparrow}\Phi_{1\downarrow}\rangle),
|T^{20}_{H}\rangle = \frac{1}{\sqrt{2}}(|\Phi_{1\uparrow}\Phi_{9\downarrow}\rangle - |\Phi_{9\uparrow}\Phi_{1\downarrow}\rangle),
\]

\[
|T^{02}_{S}\rangle = \frac{1}{\sqrt{2}}(|\Phi_{4\uparrow}\Phi_{8\downarrow}\rangle - |\Phi_{8\uparrow}\Phi_{4\downarrow}\rangle),
|T^{02}_{H}\rangle = \frac{1}{\sqrt{2}}(|\Phi_{12\uparrow}\Phi_{14\downarrow}\rangle - |\Phi_{14\uparrow}\Phi_{12\downarrow}\rangle).
\]

The subscript $L$ ($H$) in Eq. (S-10d) and Eq. (S-15) carries the meaning of “lower” (“higher”) as it involves the lower (higher) state for first-excited orbital, labeled as $(n, m) = (1, -1) ((n, m) = (1, 1))$.

As mentioned above, the effective Hamiltonian block for the $|SS\rangle$ manifold can be simplified by assuming the electrons only occupy the lowest single-particle state. This is due to the fact that for the ground $|SS\rangle$ eigenstate in large detuning, both electrons in a DDQD can occupy the lowest orbitals (Eq. (S-10a)), in contrast to the “repulsion” experienced by electrons for $|TT\rangle$ eigenstate (Eq. (S-10d)). In the bases $|S^{20}\rangle|S^{02}\rangle, |S^{11}\rangle|S^{02}\rangle, |S^{20}\rangle|S^{11}\rangle, |S^{11}\rangle|S^{11}\rangle$,

\[
H_{|SS\rangle} - E_{|S^{20}\rangle|S^{02}\rangle} = \begin{pmatrix}
0 & \sqrt{2}t & \sqrt{2}t & 0 \\
\sqrt{2}t & U_{\chi} - U^{\odot} + \varepsilon & 0 & \sqrt{2}t \\
\sqrt{2}t & 0 & U_{\chi}^{\ast} - U^{\odot} + \varepsilon & \sqrt{2}t \\
0 & \sqrt{2}t & \sqrt{2}t & U_{\chi}^{\ast} - 2U^{\odot} + 2\varepsilon
\end{pmatrix},
\]

where

\[
U_{\chi} = U_{23} + U_{14} - 2U_{24},

U^{\odot} = U_{os} - (U_{34} + U_{23} - U_{14}),

\varepsilon = \varepsilon_2 - \varepsilon_1 = \varepsilon_3 - \varepsilon_4 = \varepsilon_6 - \varepsilon_5 = \varepsilon_7 - \varepsilon_8 = \varepsilon_{10} - \varepsilon_9 = \varepsilon_{11} - \varepsilon_{12},

E_{|S^{20}\rangle|S^{02}\rangle} = 2\varepsilon_1 + 2\varepsilon_4 + 2U_{os} + 4U_{14}.
\]
\(|ST\) and \(|TS\).

In the bases of \( \{ |T^{11}\rangle |T^{11}\rangle, |T^{11}\rangle |T^{02}\rangle, |T^{11}\rangle |T^{20}\rangle |T^{11}\rangle, |T^{20}\rangle |T^{11}\rangle, |T^{11}\rangle |T^{02}\rangle, |T^{20}\rangle |T^{02}\rangle, |T^{20}\rangle |T^{02}\rangle, |T^{20}\rangle |T^{02}\rangle \} \),

\[
H_{TT} = \begin{pmatrix}
E_{|T^{11}\rangle |T^{11}\rangle} & -t_{25} & -t_{29} & t_{25} & t_{29} & 0 & 0 & 0 & 0 \\
-t_{25} & E_{|T^{11}\rangle |T^{02}\rangle} & 0 & 0 & t_{25} & t_{29} & 0 & 0 & 0 \\
-t_{29} & 0 & E_{|T^{11}\rangle |T^{20}\rangle} & 0 & 0 & t_{25} & t_{29} & 0 & 0 \\
t_{25} & 0 & 0 & E_{|T^{02}\rangle |T^{11}\rangle} & -t_{25} & t_{29} & 0 & 0 & 0 \\
t_{29} & 0 & 0 & 0 & E_{|T^{02}\rangle |T^{20}\rangle} & 0 & 0 & 0 & 0 \\
0 & t_{25} & 0 & -t_{25} & 0 & E_{|T^{20}\rangle |T^{02}\rangle} & 0 & 0 & 0 \\
0 & 0 & t_{25} & -t_{25} & 0 & 0 & E_{|T^{20}\rangle |T^{02}\rangle} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & E_{|T^{20}\rangle |T^{02}\rangle} & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & E_{|T^{20}\rangle |T^{02}\rangle} \\
\end{pmatrix},
\]

(S-13)

where

\[
\begin{align*}
E_{|T^{11}\rangle |T^{11}\rangle} - E_{|T^{02}\rangle |T^{20}\rangle} &= U^\Delta_L + U^\nabla_L - U^\Box_M + 2\varepsilon - \hbar\omega_{1,-1} - \hbar\omega_{1,1}, \\
E_{|T^{11}\rangle |T^{02}\rangle} - E_{|T^{20}\rangle |T^{02}\rangle} &= \frac{1}{2} \left[ U^\Delta_L + U^\nabla_L - 2(U^\Box_M - \varepsilon + \hbar\omega_{1,1}) \right] + \varepsilon, \\
E_{|T^{11}\rangle |T^{20}\rangle} - E_{|T^{02}\rangle |T^{20}\rangle} &= \frac{1}{2} \left[ U^\Delta_H + U^\nabla_H + 2(U^\Box_M - \varepsilon + \hbar\omega_{1,1}) \right] + \varepsilon, \\
E_{|T^{20}\rangle |T^{11}\rangle} - E_{|T^{02}\rangle |T^{02}\rangle} &= \frac{1}{2} \left[ U^\Delta_L + U^\nabla_L - 2(U^\Box_M - \varepsilon + \hbar\omega_{1,1}) \right] + \varepsilon, \\
E_{|T^{20}\rangle |T^{20}\rangle} - E_{|T^{02}\rangle |T^{02}\rangle} &= U^\Delta_H + U^\nabla_H - 2(U^\Box_M - \varepsilon + \hbar\omega_{1,1}) + \varepsilon, \\
E_{|T^{20}\rangle |T^{02}\rangle} - E_{|T^{02}\rangle |T^{20}\rangle} &= U^\Delta_L - U^\Box_M + \hbar\omega_{1,-1} - \hbar\omega_{1,1}, \\
E_{|T^{20}\rangle |T^{20}\rangle} - E_{|T^{02}\rangle |T^{20}\rangle} &= U^\Delta_H - U^\Box_M - \hbar\omega_{1,-1} + \hbar\omega_{1,1}, \\
E_{|T^{20}\rangle |T^{02}\rangle} &= E_{|T^{20}\rangle |T^{02}\rangle} = 2\varepsilon_1 + 2\varepsilon_4 + U_{1,12} + U_{5,12} + U_{15} + U_{19} + U_{14} + U_{15} + \hbar\omega_{1,-1} - \hbar\omega_{1,1},
\end{align*}
\]

and

\[
\begin{align*}
U^\Delta_L &= 2U_{34} + 2U_{13} + 2U_{35} + U_{14} - U_{58}, \\
U^\Delta_H &= 2U_{34} + 2U_{13} + 2U_{39} + U_{14} - U_{9,12}, \\
U^\nabla_L &= U_{23} + U_{58} - 2U_{35}, \\
U^\nabla_H &= U_{23} + U_{9,12} - 2U_{39}, \\
U^\Box_L &= U_{14} + (U_{15} + U_{15}) + 2U_{18} + U_{58}, \\
U^\Box_H &= U_{14} + (U_{15} + U_{15}) + (U_{18} + U_{9,12}) + U_{5,12}, \\
\hbar\omega_{p,q} &= \hbar\omega_0 + q\hbar\omega_c.
\end{align*}
\]

The effective Hamiltonian block for \(|ST\) and \(|TS\) can be obtained using the same techniques used for \(|TT\). In the bases of \( \{ |S^{11}\rangle |T^{11}\rangle, |S^{20}\rangle |T^{11}\rangle, |S^{11}\rangle |T^{02}\rangle, |S^{11}\rangle |T^{20}\rangle, |S^{20}\rangle |T^{02}\rangle, |S^{20}\rangle |T^{11}\rangle \} \),

\[
H_{ST} = \begin{pmatrix}
E_{|S^{11}\rangle |T^{11}\rangle} & \sqrt{2}t & t_{25} & t_{29} & 0 & 0 & 0 & 0 \\
\sqrt{2}t & E_{|S^{20}\rangle |T^{11}\rangle} & 0 & 0 & t_{25} & t_{29} & 0 & 0 \\
t_{25} & 0 & E_{|S^{11}\rangle |T^{02}\rangle} & \sqrt{2}t_{12} & 0 & \sqrt{2}t_{12} & 0 & \sqrt{2}t_{12} \\
t_{29} & 0 & 0 & E_{|S^{11}\rangle |T^{20}\rangle} & 0 & 0 & \sqrt{2}t_{12} & 0 \\
0 & t_{25} & \sqrt{2}t_{12} & 0 & E_{|S^{20}\rangle |T^{02}\rangle} & 0 & 0 & \sqrt{2}t_{12} \\
0 & t_{29} & 0 & \sqrt{2}t_{12} & 0 & E_{|S^{20}\rangle |T^{02}\rangle} & 0 & 0 \\
\end{pmatrix},
\]

(S-16)
Hence, $H_{TS}$ yields a similar form as $H_{ST}$, and we would not give its explicit form here.

B. Correspondence between Full-CI and extended Hubbard model

Supplementary Fig. S4 compares the energy levels obtained using the Hamiltonian given by extended Hubbard Model (Sec. II A, dashed black lines) and those from the Full-CI calculation (solid colored lines). The similarity validates the accuracy of the extended Hubbard model when used in our problem.

III. COMPARISON BETWEEN “OUTER, “CENTER” AND “RIGHT” DETUNING SCHEMES

In this section we compare the “Outer,” “Center” and “Right” detuning schemes. The eigenenergies of four logical states, $|SS\rangle$, $|ST\rangle$, $|TS\rangle$ and $|TT\rangle$ are shown in Supplementary Fig. S5. The left, middle and right column are for “Outer,” “Center” and “Right” schemes respectively, while the upper and lower rows are for $B = 0$ and $B = 0.087$ T respectively. It is found that the overlap in the $\Delta$ range between $|SS\rangle$ and $|TT\rangle$ under external magnetic field is only found for the “Outer” and “Right” detuning schemes, but not for the ”Center” scheme. Therefore the nearly-sweet-spot regime only exists for the “Outer” and “Right” , but not ”Center” scheme. This is also consistent with results shown in Fig. 3 of the main text.

Among the “Outer” and “Right” schemes, the value of capacitive coupling in the nearly-sweet-spot regime, $\alpha$ is stronger for “Outer” as compared to the “Right” scheme, as shown in Supplementary Fig. S6, indicating that a choice of detuning value in the nearly-sweet-spot regime of the “Outer” scheme is superior than others.

IV. EXISTENCE OF NEARLY SWEET SPOT REGION

In this section, we show that the existence of nearly-sweet-spot regime is not specific to the parameters chosen in our paper, but rather general under reasonable assumptions. We assume that in the DDQD system, a maximum of one electron is allowed to be excited from the ground configuration, as configurations involving two or more excited electron would have much lower probability. This assumption allows us to safely discard higher energy excitations and
Supplementary Figure S5: Eigenenergies of four logical states, $|SS\rangle$, $|ST\rangle$, $|TS\rangle$ and $|TT\rangle$ for (a,d) “Outer”, (b,e) “Center”, (c,f) “Right” detuning scheme. (a)-(c) are results for $B = 0$ while (d)-(f) are results for $B = 0.087$ T.

Supplementary Figure S6: Capacitive coupling strength, $\alpha$, as a function of detuning deduced from Supplementary Fig. S5 (d)-(f) for $B = 0.087$ T.

simplifies the calculations. We will use $(n_1, n_2, n_3, n_4)$ to denote the number of electrons occupying each dot, where $n_j$ indicates number of electrons in the $j$th dot. As noted in the main text, here we focus on the “Outer” scheme only.

A. $\Delta$ range of $|SS\rangle$

The relevant charge states for $|SS\rangle$ in the entire range of detuning are

| Charge state | Multi-particle antisymmetrized state |
|--------------|-------------------------------------|
| $(1,1,1,1)$  | $|S(\Phi_1\Phi_2)\rangle |S(\Phi_3\Phi_4)\rangle$ |
| $(1,1,1,1)^*$ | $|S(\Phi_1\Phi_2)\rangle |S(\Phi_3\Phi_5)\rangle |S(\Phi_2\Phi_3)\rangle |S(\Phi_4\Phi_1)\rangle$ |
| $(1,1,1,1)^{**}$ | $|S(\Phi_1\Phi_2)\rangle |S(\Phi_3\Phi_5)\rangle |S(\Phi_2\Phi_1)\rangle |S(\Phi_4\Phi_3)\rangle$ |
| $(1,1,0,2)/(2,0,1,1)$ | $|S(\Phi_1\Phi_2)\rangle |S(\Phi_3\Phi_5)\rangle |S(\Phi_2\Phi_4)\rangle |S(\Phi_3\Phi_1)\rangle$ |
| $(1,1,0,2)^*/(2,0,1,1)^*$ | $|S(\Phi_1\Phi_2)\rangle |S(\Phi_3\Phi_5)\rangle |S(\Phi_2\Phi_4)\rangle |S(\Phi_3\Phi_1)\rangle$ |
| $(1,1,0,2)^{**}/(2,0,1,1)^{**}$ | $|S(\Phi_1\Phi_2)\rangle |S(\Phi_3\Phi_5)\rangle |S(\Phi_2\Phi_4)\rangle |S(\Phi_3\Phi_1)\rangle$ |
| $(2,0,0,2)$ | $|S(\Phi_1\Phi_4)\rangle |S(\Phi_3\Phi_4)\rangle$ |
| $(2,0,0,2)^*$ | $|S(\Phi_1\Phi_4)\rangle |S(\Phi_3\Phi_4)\rangle |S(\Phi_2\Phi_4)\rangle |S(\Phi_3\Phi_1)\rangle$ |
| $(2,0,0,2)^{**}$ | $|S(\Phi_1\Phi_4)\rangle |S(\Phi_3\Phi_4)\rangle |S(\Phi_2\Phi_4)\rangle |S(\Phi_3\Phi_1)\rangle$ |

Supplementary Table S-I: Charge states of interest of $|SS\rangle$ in the entire range of detuning.
In Supplementary Table S-I we have defined:

\[
\left| S(\Phi_j \Phi_k) \right| = \frac{1}{\sqrt{2}} \left( \left| \Phi_j \Phi_k \right| + \left| \Phi_k \Phi_j \right| \right),
\]

\[
\left| S(\Phi_j \Phi_j) \right| = \left| \Phi_j \Phi_j \right|,
\]

\[
|\Phi_j \Phi_k\rangle = \left| \Phi_j(r_1) \uparrow (\omega_1) \Phi_j(r_2) \uparrow (\omega_2) \right| \left| \Phi_k(r_1) \downarrow (\omega_1) \Phi_k(r_2) \downarrow (\omega_2) \right|,
\]

where \( \uparrow (\downarrow) \) is the spin up (down) state and \( \Phi_j \) is \( j \)th single-particle orbital as shown in Supplementary Fig. S3. The single and double asterisk(s) on the superscript of \( \left| (n_1, n_2, n_3, n_4) \right| \) indicate an electron occupying the orbital indexed as \( 5 \leq j \leq 8 \) (\( n = 1 \), \( m = -1 \), for a single asterisk) and \( 9 \leq j \leq 12 \) (\( n = 1 \), \( m = 1 \), for double asterisks), respectively. The calculation can be further simplified by recognizing that some charge states are degenerate, e.g. \( \left| S(\Phi_1 \Phi_2) \right| \left| S(\Phi_3 \Phi_8) \right| \) and \( \left| S(\Phi_1 \Phi_5) \right| \left| S(\Phi_2 \Phi_4) \right| \). Hence, each charge state can be represented by only one of its corresponding multi-particle state, e.g. \((1,1,1,1)^*\) by \( \left| S(\Phi_1 \Phi_2) \right| \left| S(\Phi_3 \Phi_8) \right| \), and \((1,1,0,2)\) by \( \left| S(\Phi_1 \Phi_2) \right| \left| S(\Phi_3 \Phi_4) \right| \). We denote the probability of electron excitation to higher orbitals by \( \eta \), and the corresponding charge state \( \text{ex}(n_1, n_2, n_3, n_4) \). The expressions of \( |S^{11}S^{11}| \), \( \left| \bar{S}S \right| \) and \( |S^{20}S^{02}| \) are thus linear combinations of the corresponding charge states,

\[
|S^{11}S^{11}| = c_{(1,1,1,1)}|(1,1,1,1)\rangle + c_{(1,1,1,1)^*}||(1,1,1,1)^*\rangle + c_{(1,1,1,1)^{**}}||1,1,1,1)^{**}\rangle + \eta_{(1,1,1,1)}\text{ex}(1,1,1,1),
\]

\[
\left| \bar{S}S \right| = c_{(1,1,0,2)}||1,1,0,2\rangle + c_{(1,1,0,2)^*}||(1,1,0,2)^*\rangle + c_{(1,1,0,2)^{**}}||(1,1,0,2)^{**}\rangle
\]

\[
+ \eta_{(1,1,0,2)}\text{ex}(1,1,0,2),
\]

\[
|S^{20}S^{02}| = c_{(2,0,0,2)}||(2,0,0,2)\rangle + c_{(2,0,0,2)^*}||(2,0,0,2)^*\rangle + c_{(2,0,0,2)^{**}}||(2,0,0,2)^{**}\rangle
\]

\[
+ \eta_{(2,0,0,2)}\text{ex}(2,0,0,2),
\]

where the coefficients satisfy the normalization condition,

\[
|c_{(1,1,1,1)}|^2 + |c_{(1,1,1,1)^*}|^2 + |c_{(1,1,1,1)^{**}}|^2 + |\eta_{(1,1,1,1)}|^2 = 1,
\]

\[
|c_{(1,1,0,2)}|^2 + |c_{(1,1,0,2)^*}|^2 + |c_{(1,1,0,2)^{**}}|^2 + |\eta_{(1,1,0,2)}|^2 = 1,
\]

\[
|c_{(2,0,0,2)}|^2 + |c_{(2,0,0,2)^*}|^2 + |c_{(2,0,0,2)^{**}}|^2 + |\eta_{(2,0,0,2)}|^2 = 1.
\]

The relationships between coefficients \( c_{(\cdots)} \) in Eq. (S-19) are summarized in Eq. (S-21) below. Eqs. (S-21a)-(S-21c) are obtained by noting that electrons energetically favor the lowest orbitals. In \((1,1,0,2)/(2,0,0,2)\) charge configurations, double electron occupancy on one dot implies that the two electrons favor to occupy different orbitals on the same dot as the Coulomb repulsion on the same orbital is stronger, e.g. \( U_{15} < U_1 \). Such repulsion effect is less pronounced in the \((1,1,1,1)\) charge configuration. Similar reasoning applies when comparing the \((1,1,0,2)\) and \((2,0,0,2)\) charge configurations, resulting in Eqs. (S-21d–S-21f). Also, the enhancement of the magnetic field lowers the energies of orbitals with \( m < 0 \), increasing the probability for electrons to occupy those orbitals to mitigate strong on-site Coulomb interaction, \( U_1 \). Such effect is more significant for eigenstates with more doubly-occupied dots, i.e. the effect for \((1,1,0,2)\) is stronger than \((1,1,1,1)\), but the effect is strongest for \((2,0,0,2)\). These considerations lead to Eq. (S-21g).

\[
|c_{(1,1,1,1)}| \gg |c_{(1,1,1,1)^*}| \gg |c_{(1,1,1,1)^{**}}|,
\]

\[
|c_{(1,1,0,2)}| \gg |c_{(1,1,0,2)^*}| \gg |c_{(1,1,0,2)^{**}}|,
\]

\[
|c_{(2,0,0,2)}| \gg |c_{(2,0,0,2)^*}| \gg |c_{(2,0,0,2)^{**}}|,
\]

\[
|c_{(1,1,1,1)}| \gg |c_{(1,1,0,2)}| \gg |c_{(2,0,0,2)}|,
\]

\[
|c_{(1,1,1,1)^*}| \gg |c_{(1,1,0,2)^*}| \gg |c_{(2,0,0,2)^*}|,
\]

\[
|c_{(1,1,1,1)^{**}}| \gg |c_{(1,1,0,2)^{**}}| \gg |c_{(2,0,0,2)^{**}}|,
\]

\[
|\eta_{(2,0,0,2)}|^2 \gg |\eta_{(1,1,0,2)}|^2 \gg |\eta_{(1,1,1,1)}|^2.
\]
We can therefore obtain the starting, \( \Delta^*_{(SS),s} \) and ending, \( \Delta^*_{(SS),e} \) points of the \( \Delta \) range of \( \langle SS \rangle \), which are also the avoided-crossing points, by equating the eigenenergies of \( |S^{11}S^{11}\rangle \) with \( \langle SS \rangle \) and \( |SS\rangle \) with \( |S^{20}S^{02}\rangle \) respectively. The results are,

\[
\Delta^*_{(SS),s} \approx \tilde{E}_{1111,1102} + \hbar \omega_c \left[ \left( |c_{1111}|^2 - |c_{1102}|^2 \right) + 2 \left( |c_{1111}|^2 - |c_{1102}|^2 \right) \right] \\
= \tilde{E}_{1111,1102} + \hbar \omega_c \left[ \left( |c_{1111}|^2 + 2 |c_{1111}|^2 \right) - \left( |c_{1102}|^2 + 2 |c_{1102}|^2 \right) \right] \\
\approx \tilde{E}_{1111,1102} + \hbar \omega_c \left[ \left( |c_{1102}|^2 + 2 |c_{1102}|^2 \right) \right] \\
= \tilde{E}_{1111,1102} + \hbar \omega_c \left[ \left( |c_{1111}|^2 + |c_{1102}|^2 \right) \right] (S-22a)
\]

\[
\Delta^*_{(SS),e} \approx \tilde{E}_{1102,2002} + \hbar \omega_c \left[ \left( |c_{1102}|^2 - |c_{2002}|^2 \right) + 2 \left( |c_{1102}|^2 - |c_{2002}|^2 \right) \right] \\
= \tilde{E}_{1102,2002} + \hbar \omega_c \left[ \left( |c_{1102}|^2 + 2 |c_{1102}|^2 \right) - \left( |c_{2002}|^2 + 2 |c_{2002}|^2 \right) \right] \\
\approx \tilde{E}_{1102,2002} + \hbar \omega_c \left[ \left( |c_{1102}|^2 + |c_{2002}|^2 \right) \right] (S-22b)
\]

where

\[
\tilde{E}_{1111,1102} = U_{15} + U_{14} - U_{34} - U_{23} + |c_{1102}|^2 \left( U_1 - U_{15} \right) + \hbar \omega_0 \left( |c_{1111}|^2 - |c_{1102}|^2 \right),
\]

\[
\tilde{E}_{1102,2002} = 2U_{14} - U_{34} - 2U_{13} + U_1 \left( 1 - |c_{1102}|^2 \right)^2 + |c_{2002}|^2 \right) + \hbar \omega_0 \left( |c_{1111}|^2 - |c_{2002}|^2 \right) \right) .
\]

(S-23)

It is reasonable to assume that although electrons may excite to higher orbitals, the probability of such excitation is low, i.e. \( |c_{\ldots\ldots}| \ll |c_{\ldots\ldots}| \), as suggested by Eqs. (S-21a) and (S-21b), giving the third lines of Eq. (S-22a) and Eq. (S-22b). The forth lines of Eq. (S-22a) and Eq. (S-22b) are obtained by applying the normalization condition, Eq. (S-20).

We are interested in the magnetic field dependence of the avoided-crossing points, i.e. \( \partial \Delta^*_{(SS),s}/\partial (\hbar \omega_c) \) and \( \partial \Delta^*_{(SS),e}/\partial (\hbar \omega_c) \). Implied by Eqs. (S-21d)-(S-21f)), the square brackets in the first lines of Eq. (S-22a) yield positive values. Also Eq. (S-21g) suggests that the value of the square bracket in the fourth line of Eq. (S-22b) is greater than that in Eq. (S-22a). These considerations imply that both \( \Delta^*_{(SS),e} \) and \( \Delta^*_{(SS),s} \) increase with increasing magnetic field, while the former increases faster. We therefore have,

\[
\Delta^*_{(SS),e} - \Delta^*_{(SS),s} \propto \hbar \omega_c .
\]

(S-24)

This behavior is consistent with the result of Full-CI calculation, shown in Supplementary Fig. S7(a).
B. \( \Delta \) range of \(|\hat{T}\hat{T}|\)

The relevant charge states for \(|\hat{T}\hat{T}|\) in the entire range of detuning are

| Charge state | Multi-particle antisymmetrized state |
|--------------|--------------------------------------|
| \(|(1, 1, 1, 1)|\) | \(|T(\Phi_1\Phi_2)\rangle |T(\Phi_3\Phi_4)\rangle| \)
| \(|(1, 1, 0, 2)^R\rangle\) | \(|T(\Phi_1\Phi_2)\rangle |T(\Phi_3\Phi_5)\rangle| \)
| \(|(1, 1, 0, 2)^R^{***}\rangle\) | \(|T(\Phi_1\Phi_2)\rangle |T(\Phi_3\Phi_6)\rangle| \)
| \(|(2, 0, 0, 2)^L^{***}\rangle\) | \(|T(\Phi_1\Phi_3)\rangle |T(\Phi_4\Phi_5)\rangle| \)
| \(|(2, 0, 0, 2)^L^{***}R^{***}\rangle\) | \(|T(\Phi_5\Phi_3)\rangle |T(\Phi_4\Phi_6)\rangle| \)

Supplementary Table S-II: Charge states of interest of \(|\hat{T}\hat{T}|\) in the entire range of detuning.

In Supplementary Table S-II we have defined

\[
|T(\Phi_j\Phi_k)\rangle = \frac{1}{\sqrt{2}} (|\Phi_j\Phi_k\rangle - |\Phi_k\Phi_j\rangle).
\]  

(S-25)

We use the superscripts to indicate which electron occupies higher energy states. In the superscripts, \(L\) and \(R\) indicate that the electron concerned is in the left and right DQD respectively. The number of asterisks indicates the number of excitation to the higher orbital with respect to ground orbital. For example, in the \((1, 1, 0, 2)\) charge state, the ground state configuration of \(|\hat{T}\hat{T}|\) is \(|T(\Phi_1\Phi_2)\rangle |T(\Phi_3\Phi_5)\rangle| \), of which an electron in the right DQD occupies the first excited orbital, \(\Phi_8\), is denoted as \(|(1, 1, 0, 2)^R\rangle\). If we move an electron in \(\Phi_4\) in the right DQD to the second higher orbitals, \(\Phi_{12}\) (state No. 12), the resultant charge state \(|T(\Phi_1\Phi_2)\rangle |T(\Phi_8\Phi_{12})\rangle| \) is then denoted as \(|(1, 1, 0, 2)^R^{***}\rangle\) as the total number of excitations of the electrons from ground orbitals is three. For degenerate states, we keep only one of them and discard others as their coefficients can be absorbed into those shown in Table S-II. Also, we empirically found that the Coulomb interaction between two dots in a DQD is similar regardless of the level of excitations, e.g. \(U_{34} = U_{16} = U_{1,10}\). This allows us to neglect the excitation when each DQD is in \((1, 1)\) charge configuration, as it is energetically unfavorable to occupy higher orbitals in the inter-dot case. On the other hand, the on-site (inter-orbital) Coulomb repulsion is weaker when higher orbitals are involved, e.g. \(U_{59} < U_{15} < U_1\). Therefore, for a doubly-occupied quantum dot, an electron will favor to occupy excited orbitals. Near the avoid-crossing between \(|T^{11}|T^{11}\rangle\) and \(|\hat{T}\hat{T}|\) at small detuning, the charge states are

\[
|T^{11}|T^{11}\rangle \approx |(1, 1, 1, 1)\rangle,
\]

(S-26a)

\[
|\hat{T}\hat{T}| \approx |(1, 1, 0, 2)^R\rangle.
\]

(S-26b)

At larger detuning, near the avoided-crossing between \(|\hat{T}\hat{T}|\) and \(|T^{20}|T^{02}\rangle\), the charge states are

\[
|\hat{T}\hat{T}| = c_{(1,1,0,2)^R}|(1, 1, 0, 2)^R\rangle + c_{(1,1,0,2)^R^{***}}|(1, 1, 0, 2)^R^{***}\rangle + \eta_{(1,1,0,2)}|\text{ex}(1, 1, 0, 2)\rangle,
\]

(S-27a)

\[
|T^{20}|T^{02}\rangle = c_{(2,0,0,2)^L^{***}R^{***}}|(2, 0, 0, 2)^L^{***}R^{***}\rangle + c_{(2,0,0,2)^L^{***}R^{***}}|(2, 0, 0, 2)^L^{***}R^{***}\rangle + \eta_{(2,0,0,2)}|\text{ex}(2, 0, 0, 2)\rangle.
\]

(S-27b)

The relationship between the coefficients in Eq. (S-27) are given as:

\[
|c_{(1,1,0,2)^R}|^2 + |c_{(1,1,0,2)^R^{***}}|^2 + |\eta_{(1,1,0,2)}|^2 = 1,
\]

(S-28a)

\[
|c_{(2,0,0,2)^L^{***}R^{***}}|^2 + |c_{(2,0,0,2)^L^{***}R^{***}}|^2 + |c_{(2,0,0,2)^L^{***}R^{***}}|^2 + |\eta_{(2,0,0,2)}|^2 = 1,
\]

(S-28b)

\[
|c_{(1,1,0,2)^R}|^2 > |c_{(2,0,0,2)^L^{***}R^{***}}|^2.
\]

(S-28c)

Eqs. (S-28a)-(S-28b) are obtained from the normalization condition, while Eq. (S-28c) is found by recognizing that the Coulomb repulsion effect is more pronounced for \(|(2, 0, 0, 2)^L^{***}R^{***}\rangle\) than \(|(1, 1, 0, 2)^R\rangle\), so that electrons \(|(2, 0, 0, 2)^L^{***}R^{***}\rangle\) favor occupying highly-lying orbitals, resulting in a higher energy state with lower probability than the latter. We
then obtain $\Delta^*_{\{\bar{T}\},s}$ by equating the eigenenergies of $|T^{11}\rangle$ and $|\bar{T}\rangle$ at smaller detuning and $\Delta^*_{\{T\},e}$ by equating the eigenenergies of $|T\rangle$ and $|T^{20}\rangle$ at larger detuning, giving

$$
\Delta^*_{\{\bar{T}\},s} \approx \bar{E}_{\{1,1,1,1\},\{(1,1,0,2)\}} - \hbar \omega_c,
$$

(S-29a)

$$
\Delta^*_{\{T\},e} - \Delta^*_{\{\bar{T}\},s} \approx \bar{E}_{\{1,1,1,1\},\{(1,1,0,2)\}} + \bar{E}_{\{(1,1,0,2)\},\{2,0,0,2\}} + \hbar \omega_c \left\{ \left( \left| c_{\{(2,0,0,2)\}^{2}} \right|^2 - \left| c_{\{(1,1,0,2)\}^{2}} \right|^2 \right) + \left( \left| c_{\{(1,1,0,2)\}^{2}} \right|^2 - \left| c_{\{(2,0,0,2)\}^{2}} \right|^2 \right) \right\},
$$

(S-29b)

where

$$
\bar{E}_{\{1,1,1,1\},\{(1,1,0,2)\}} = U_{14} + U_{34} - U_{14} + (U_{15} - U_{59}) \left| c_{\{(1,1,0,2)\}^{2}} \right|^2 + \hbar \omega_c \left( 2 - \left| c_{\{(1,1,0,2)\}^{2}} \right|^2 \right),
$$

$$
\bar{E}_{\{(1,1,0,2)\},\{2,0,0,2\}} = U_{59} + 2U_{14} - U_{34} - 2U_{13} + (U_{59} - U_{15} + \hbar \omega_c) \left( \left| c_{\{(1,1,0,2)\}^{2}} \right|^2 - 2 \left| c_{\{(2,0,0,2)\}^{2}} \right|^2 - \left| c_{\{(2,0,0,2)\}^{2}} \right|^2 \right) + 2\hbar \omega_c.
$$

(S-30)

Again, we are interested in the magnetic-field dependence of the avoided-crossing points, i.e. $\partial \Delta^*_{\{\bar{T}\},s}/\partial (\hbar \omega_c)$ and $\partial \Delta^*_{\{T\},e}/\partial (\hbar \omega_c)$. Eq. (S-29a) shows that a stronger magnetic field leads to a smaller value of $\Delta^*_{\{\bar{T}\},s}$. Also, Eq. (S-28b) and Eq. (S-28c) suggest that the curly bracket in Eq. (S-29b) yields a positive value, so that the $\Delta$ range of $|\bar{T}\rangle$ increases with the magnetic field as

$$
\left( \Delta^*_{\{\bar{T}\},e} - \Delta^*_{\{\bar{T}\},s} \right) \propto \hbar \omega_c.
$$

(S-31)

These results are again consistent with those from the Full-CI calculation, as can be seen in Supplementary Fig. S7(b).

C. $\Delta$ range of $|S^{20}T^{11}\rangle$ and $|T^{11}S^{02}\rangle$

We denote the starting and ending $\Delta$ values of $|S^{20}T^{11}\rangle$ as $\Delta^*_{\{ST\},s}$ and $\Delta^*_{\{ST\},e}$ respectively (cf. Supplementary Fig. S5(d)). Near the avoided crossing between $|S^{11}T^{11}\rangle$ and $|S^{20}T^{11}\rangle$, the relevant charge states for $|ST\rangle$ are

| Charge state | Multi-particle antisymmetrized state |
|-------------|--------------------------------------|
| $|(1,1,1,1)\rangle$ | $|S(\Phi_1\Phi_2)\rangle|T(\Phi_3\Phi_4)\rangle$ |
| $|(1,1,1,1)^*\rangle$ | $|S(\Phi_1\Phi_2)\rangle|T(\Phi_3\Phi_5)\rangle|S(\Phi_2\Phi_5)\rangle|T(\Phi_3\Phi_4)\rangle$ |
| $|(1,1,1,1)^{**}\rangle$ | $|S(\Phi_1\Phi_2)\rangle|T(\Phi_3\Phi_4)\rangle|S(\Phi_2\Phi_5)\rangle|T(\Phi_3\Phi_4)\rangle$ |
| $|(1,1,0,2)/(2,0,1,1)\rangle$ | $|S(\Phi_1\Phi_2)\rangle|T(\Phi_3\Phi_4)\rangle|S(\Phi_2\Phi_5)\rangle|T(\Phi_3\Phi_4)\rangle$ |
| $|(1,1,0,2)^*/(2,0,1,1)^*\rangle$ | $|S(\Phi_1\Phi_2)\rangle|T(\Phi_3\Phi_4)\rangle|S(\Phi_2\Phi_5)\rangle|T(\Phi_3\Phi_4)\rangle$ |
| $|(1,1,0,2)^{**}/(2,0,1,1)^{**}\rangle$ | $|S(\Phi_1\Phi_2)\rangle|T(\Phi_3\Phi_4)\rangle|S(\Phi_2\Phi_5)\rangle|T(\Phi_3\Phi_4)\rangle$ |

Supplementary Table S-III: Charge states of interest of $|ST\rangle$ near the avoided crossing between $|S^{11}T^{11}\rangle$ and $|S^{20}T^{11}\rangle$.

To avoid confusion, we emphasize that even though the notations for charge states here are same as those shown in Supplementary Sec. IV A, they refer to $|ST\rangle$ instead of $|SS\rangle$. We have

$$
|S^{11}T^{11}\rangle = c_{\{(1,1,1,1)\}}|(1,1,1,1)\rangle + c_{\{(1,1,1,1)^{*}\}}|(1,1,1,1)^{*}\rangle + c_{\{(1,1,1,1)^{**}\}}|(1,1,1,1)^{**}\rangle + \eta_{\{(1,1,1,1)\}}|\text{ex}(1,1,1,1)\rangle,
$$

(S-32a)

$$
|S^{20}T^{11}\rangle = c_{\{(1,1,0,2)\}}|(1,1,0,2)\rangle + c_{\{(1,1,0,2)^{*}\}}|(1,1,0,2)^{*}\rangle + c_{\{(1,1,0,2)^{**}\}}|(1,1,0,2)^{**}\rangle + \eta_{\{(1,1,0,2)\}}|\text{ex}(1,1,0,2)\rangle.
$$

(S-32b)

Equating the eigenenergies of $|S^{11}T^{11}\rangle$ with $|S^{20}T^{11}\rangle$ gives that

$$
\Delta^*_{\{ST\},s} \approx \bar{E}_{\{1111\},\{1102\}} + \hbar \omega_c \left( \left| c_{\{1111\}} \right|^2 - \left| c_{\{1102\}} \right|^2 \right) + 2 \left( \left| c_{\{1111\}} \right|^2 - \left| c_{\{1102\}} \right|^2 \right),
$$

(S-33)
\[
\tilde{E}_{(1,1,1,1),(1,1,0,2)} = U_{15} + U_{14} - U_{34} - U_{23} + |c_{(1,1,1,1)}|^2 (U_1 - U_{15}) + \hbar \omega_0 \left( |c_{(1,1,1,1)}|^2 - |c_{(1,1,0,2)}|^2 \right).
\]  
(S-34)

Eq. (S-33) yields the same expression as the first line of Eq. (S-22a), implying that equating the eigenenergies of between two dots is similar regardless of the level of excitations, e.g. between the charges states with excited orbitals as shown in Eq. (S-27) are not considered here for the purpose of comparison.

\[
\partial J / \partial \tilde{\alpha}_{ST},e \approx \partial \Delta / \partial \tilde{\alpha}_{TT},e.
\]  
(S-35)

Near the avoided crossing between \(|S^{20}T^{11}\rangle\) and \(|S^{20}T^{02}\rangle\), we focus on the main composition of states \(|ST\rangle\) and \(|TT\rangle\). The relevant charge states are

\[
\begin{align*}
|S^{20}T^{11}\rangle &\approx |S(\Phi_1 \Phi_1)|T(\Phi_2 \Phi_4)\rangle, \\
|S^{20}T^{02}\rangle &\approx |S(\Phi_1 \Phi_1)|T(\Phi_4 \Phi_8)\rangle, \\
|TT\rangle &\approx |T(\Phi_1 \Phi_3)|T(\Phi_3 \Phi_4)\rangle, \\
|T^{20}T^{02}\rangle &\approx |T(\Phi_1 \Phi_5)|T(\Phi_4 \Phi_8)\rangle.
\end{align*}
\]  
(S-36a)

The charges states with excited orbitals as shown in Eq. (S-27) are not considered here for the purpose of comparison between the \(|ST\rangle\) and \(|TT\rangle\) charge state. For degenerate states, we keep only one of them and discard others as their coefficients can be absorbed into those shown in Eq. (S-36). Also, we empirically found that the Coulomb interaction between two dots is similar regardless of the level of excitations, e.g. \(U_{14} \approx U_{18} \approx U_{45}\). We then obtain \(\Delta_\ast \) by equating the eigenenergies of \(|TT\rangle\) and \(|T^{20}T^{02}\rangle\), and \(\Delta_\ast \) by equating the eigenenergies of \(|S^{20}T^{11}\rangle\) and \(|S^{20}T^{02}\rangle\), giving

\[
\begin{align*}
\Delta_\ast_{|ST\rangle} &\approx U_{15} - (U_{13} + U_{14}) - U_{34} + (U_{45} + U_{45}) + \hbar \omega_0 - \hbar \omega_c, \\
\Delta_\ast_{|TT\rangle} &\approx U_{15} - (U_{13} + U_{14}) - U_{34} + (U_{45} + U_{85}) + \hbar \omega_0 - \hbar \omega_c \\
&\approx U_{15} - (U_{13} + U_{14}) - U_{34} + (U_{45} + U_{45}) + \hbar \omega_0 - \hbar \omega_c.
\end{align*}
\]  
(S-37a)

Eq. (S-37) shows that

\[
\Delta_\ast_{|ST\rangle} \approx \Delta_\ast_{|TT\rangle}.
\]  
(S-38)

Eq. (S-35) and Eq. (S-38) are consistent with results from the Full-CI calculation, as can be seen in Supplementary Fig. S5(d). Since the eigenenergy of \(|TS\rangle\) is the same as that of \(|ST\rangle\) for the “Outer” scheme, the starting and ending points of the \(\Delta\) range of \(|T^{11}S^{02}\rangle\) are identical to those of \(|ST\rangle\).

**D. Nearly sweet spot regime and strong inter-qubit coupling**

To perform a high fidelity two-qubit gate, we need on one hand substantially suppressed charge noises, i.e. \(\partial J_{1L, e} / \partial \Delta \rightarrow 0\), \(\partial J_{2L, e} / \partial \Delta \rightarrow 0\) and \(\partial J_{1L, e} / \partial \Delta \rightarrow 0\), and on the other hand a strong inter-qubit coupling \(\alpha\) to limit the exposure to other decoherence channels. To satisfy the first condition, we need a relatively wide \(\Delta\) range where \(\partial E_{|SS\rangle} / \partial \Delta \approx \partial E_{|ST\rangle} / \partial \Delta \approx \partial E_{|TS\rangle} / \partial \Delta \approx \partial E_{|TT\rangle} / \partial \Delta\). Concerning the second condition, we note that an electron of \(|T^{02}\rangle\) \((|T^{20}\rangle)\) has to occupy an excited orbital, while both electrons of \(|S^{02}\rangle\) \((|S^{20}\rangle)\) can occupy the ground orbital. Therefore the difference of single-particle energy due to orbital effect, \(\hbar \omega_0\) contributes to strong inter-qubit capacitive coupling, \(\tilde{\alpha}\), as

\[
\tilde{\alpha} = (2U_{24} + U_{17} - 3U_{13}) + (U_{27} - U_{23}) + (U_{26} - U_{2}) + (\hbar \omega_0 - \hbar \omega_c).
\]  
(S-39)
We emphasize that $\tilde{\alpha}$ is the $\alpha$ value where $\partial \alpha / \partial \Delta = 0$, (cf. Supplementary Fig. S6). To facilitate later discussions, we note that

\[
\frac{\partial \Delta^*_\text{SS},e}{\partial (\hbar \omega_c)} > 0, \tag{S-40a}
\]
\[
\frac{\partial \Delta^*_\text{TT},s}{\partial (\hbar \omega_c)} \approx -1, \tag{S-40b}
\]

where Eq. (S-40a) is given by the last line of Eq. (S-22b) while Eq. (S-40b) is derived from Eq. (S-29a).

When the magnetic field increases, $\Delta^*_\text{SS},e$ increases (Eq. (S-40a)), while $\Delta^*_\text{TT},s$ decreases (Eq. (S-40b)), making it possible an overlap between the $\Delta$ ranges of $\ket{\text{SS}}$ and $\ket{\text{TT}}$ above certain magnetic field strength, satisfying

\[
\Delta^*_\text{SS},e - \Delta^*_\text{TT},s > 0. \tag{S-41}
\]

Also, since $\Delta^*_\text{ST},s \approx \Delta^*_\text{SS},e$ (Eq. (S-35)) and $\Delta^*_\text{ST},s \approx \Delta^*_\text{TT},e > \Delta^*_\text{TT},s$ (Eq. (S-38)), the overlap will occur simultaneously for all logical eigenstates $\ket{\text{SS}}$, $\ket{S^{20}T^{11}}$, $\ket{T^{11}S^{02}}$, and $\ket{\text{TT}}$. In addition, the $\Delta$ ranges of $\ket{\text{SS}}$ and $\ket{\text{TT}}$ increase with the magnetic field, given by Eq. (S-24) and (S-31), making the overlap range wide enough. Therefore, we have demonstrated that at sufficiently large magnetic field, the DDQD system will exhibit a nearly-sweet-spot regime for both single-qubit exchange interactions and the inter-qubit capacitive coupling, and at the same time the capacitive coupling can be strong.

V. SYSTEM BATH HAMILTONIAN

The Hamiltonian of a DDQD in a noisy environment can be expressed as [S2]:

\[
H = H_\text{int} + H_z + H_\text{SOI} + H_\text{hyp} + H_\text{el-ph} + H_\text{ph} + H_\varphi
\]
\[
= \tilde{H} + H_\text{el-ph} + H_\text{ph} + H_\varphi, \tag{S-42}
\]

where $H_\text{int}$ is the system Hamiltonian of interest, $H_z$ the Zeeman term, $H_\text{SOI}$ the spin-orbit coupling, $H_\text{hyp}$ the hyperfine coupling, $H_\text{el-ph}$ the electron-phonon interaction, $H_\text{ph}$ the phonon bath, and $H_\varphi$ the dephasing Hamiltonian. We have

\[
\tilde{H} = H_\text{int} + H_z + H_\text{SOI} + H_\text{hyp}, \tag{S-43a}
\]
\[
H_z = \frac{E_z}{2} \sigma_B, \tag{S-43b}
\]
\[
H_\text{SOI} \simeq g^* \mu_B (r \times B) \cdot \sigma, \tag{S-43c}
\]
\[
H_\text{hyp} \simeq \frac{\delta B}{2} \langle S^{11} \rangle \langle T^{11} \rangle, \tag{S-43d}
\]
\[
H_\text{el-ph} = \sum_{\mathbf{q},s} W_s(\mathbf{q}) a^{\dagger}_{\mathbf{q}s} a_{\mathbf{q}s} e^{i\mathbf{q} \cdot \mathbf{r}} + \text{H.c.}, \tag{S-43e}
\]
\[
H_\text{ph} = \sum_{\mathbf{q},s} \hbar \nu_{\mathbf{q}s} \left( a^{\dagger}_{\mathbf{q}s} a_{\mathbf{q}s} + \frac{1}{2} \right), \tag{S-43f}
\]
\[
H_\varphi = H_\varphi_L + H_\varphi_R + H_\varphi_X, \tag{S-43g}
\]
\[
H_\varphi_L = \hbar \nu_L f_\varphi(t) (\sigma_z \otimes I),
\]
\[
H_\varphi_R = \hbar \nu_R f_\varphi(t) (I \otimes \sigma_z),
\]
\[
H_\varphi_X = \hbar \nu_{L,R} f_\varphi(t) (\sigma_z \otimes \sigma_z), \tag{S-43h}
\]

where $E_z = g^* \mu_B B$ is the Zeeman energy, $g^*$ the effective Landé-$g$ factor, $\mu_B$ the Bohr magneton, $B = |\mathbf{B}|$ the magnetic field strength, and $\sigma_B = \sigma \cdot e_B$ the Pauli spin operator along the direction of the magnetic field. The hyperfine coupling strength $\delta B$ couples states $\ket{S^{11}}$ and $\ket{T^{11}}$ of a DQD. $W_s(\mathbf{q})$ is the strength of electron-phonon interaction with phonon of type $s$ and wave vector $\mathbf{q}$, while $a^{\dagger}_{\mathbf{q}s}$ ($a_{\mathbf{q}s}$) creates (annihilates) a phonon. The dispersion relation for the phonon is assumed to be $\omega_{\mathbf{q}s} = q \nu_s$ [S3]. $H_\varphi_L$, $H_\varphi_R$, and $H_\varphi_X$ are charge-noise-induced dephasing
terms on the single-qubit exchange energies, $J_{\text{leff}}$ (for the left DQD), $J_{\text{re}}$ (for the right DQD) and the capacitive coupling between the left and right DQD, $\alpha$, respectively.

To obtain the master equation, we first apply a unitary transformation, $\tilde{U}$, that diagonalizes $\tilde{H}$, resulting in an effective Hamiltonian, $H_\text{q} + H_{\text{q-ph}} + H_{\text{ph}}$ as

$$
\tilde{U} \tilde{H} \tilde{U}^\dagger = H_\text{q} + \frac{1}{2} g^* \mu_B \left[ B_{\text{le}}^\text{eff} (\sigma_z \otimes I) + B_{\text{re}}^\text{eff} (I \otimes \sigma_z) + B_{\text{LR}}^\text{eff} \sigma_z \otimes \sigma_z \right],
$$

(S-44a)

$$
\tilde{U} H_{\text{ch-ph}} \tilde{U}^\dagger = H_{\text{q-ph}} = H_{\text{q-ph}_\text{dep}} + H_{\text{q-ph}_\text{rel}},
$$

(S-44b)

$$
\text{where}
$$

$$
H_{\text{q-ph}_\text{dep}} = \frac{1}{2} g^* \mu_B \left[ \delta B_\text{L} (\sigma_z \otimes I) + \delta B_\text{R} (I \otimes \sigma_z) + \delta B_{\text{LR}} (\sigma_z \otimes \sigma_z) \right],
$$

(S-45a)

$$
H_{\text{q-ph}_\text{rel}} = \frac{1}{2} g^* \mu_B \sum_{j<k} \delta B_{jk} (\sigma_{jk} + \text{H.c.}),
$$

(S-45b)

$B_{\text{le}}^\text{eff}$, $B_{\text{re}}^\text{eff}$ and $B_{\text{LR}}^\text{eff}$ are the effective magnetic field while $\delta B_\text{L}$, $\delta B_\text{R}$, $\delta B_{\text{LR}}$ and $\delta B_{jk}$ are phonon-induced noisy terms. $\sigma_{jk} = \eta k$ are the coupling operators between logical states, with $\{j, k\} \in \{|TT, |TS, |ST, |SS\}$. The subscripts “dep” and “rel” refers to phonon induced pure dephasing and relaxation effect respectively.

A. Charge noise dephasing $\gamma_{\varphi}$

The charge-noise dephasing effect can be modeled as time-varying fluctuations on exchange energies and the capacitive coupling, i.e.

$$
H_{\varphi_\text{L}} = \delta J_{\text{leff}}^\text{eff}(t) (\sigma_z \otimes I) = h \nu_\text{L} (\sigma_z \otimes I) f_{\varphi}(t) = h \nu_\text{L} (\sigma_z \otimes I) \int_{-\infty}^{\infty} f_{\varphi}(\omega) e^{i\omega t} d\omega,
$$

$$
H_{\varphi_\text{R}} = \delta J_{\text{re}}^\text{eff}(t) (I \otimes \sigma_z) = h \nu_\text{R} (I \otimes \sigma_z) f_{\varphi}(t) = h \nu_\text{R} (I \otimes \sigma_z) \int_{-\infty}^{\infty} f_{\varphi}(\omega) e^{i\omega t} d\omega,
$$

(S-46)

$$
H_{\varphi_{\text{LR}}} = \delta \alpha(t) (\sigma_z \otimes \sigma_z) = h \nu_{\text{LR}} (\sigma_z \otimes \sigma_z) f_{\varphi_{\text{LR}}}(t) = h \nu_{\text{LR}} (\sigma_z \otimes \sigma_z) \int_{-\infty}^{\infty} f_{\varphi}(\omega) e^{i\omega t} d\omega,
$$

where $\delta J_{\text{leff}}^\text{eff}(t)$, $\delta J_{\text{re}}^\text{eff}(t)$, and $\delta J_{\text{LR}}^\text{eff}(t)$ are the fluctuations of exchange energies and the capacitive coupling as functions of time, respectively, $f_{\varphi}(t)$ is a random function of time with zero mean, while $\nu_\text{L, R}$ and $\nu_{\text{LR}}$ are the coupling magnitudes with exchange energies, $J_{\text{leff}}$, $J_{\text{re}}$ and the capacitive coupling, $\alpha$ respectively. Also, we use the identity $f_{\varphi}(t) = \int_{-\infty}^{\infty} f_{\varphi}(\omega) e^{i\omega t} d\omega$ to write $H_{\varphi}$ in the frequency space.

We apply the unitary transformation $\tilde{U}$, on the Hamiltonian Eq. (S-42), and move to the rotating frame defined by the transformation $\tilde{U} = \exp[-i(H_\text{q} + H_{\text{ph}})t/\hbar]$. We have

$$
\tilde{U} \tilde{U} H_{\varphi_\text{L}} \tilde{U}^\dagger = h \nu_\text{L} (\sigma_z \otimes I) \int_{-\infty}^{\infty} f_{\varphi}(\omega) e^{i\omega t} d\omega,
$$

(S-47a)

$$
\tilde{U} \tilde{U} H_{\varphi_\text{R}} \tilde{U}^\dagger = h \nu_\text{R} (I \otimes \sigma_z) \int_{-\infty}^{\infty} f_{\varphi}(\omega) e^{i\omega t} d\omega,
$$

(S-47b)

$$
\tilde{U} \tilde{U} H_{\varphi_{\text{LR}}} \tilde{U}^\dagger = h \nu_{\text{LR}} (\sigma_z \otimes \sigma_z) \int_{-\infty}^{\infty} f_{\varphi}(\omega) e^{i\omega t} d\omega.
$$

(S-47c)

Eq. (S-47) implies the dephasing rates [S4]:

$$
\gamma_{\varphi_\text{L}} = 2\nu_\text{L}^2 S(\omega \to 0),
$$

$$
\gamma_{\varphi_\text{R}} = 2\nu_\text{R}^2 S(\omega \to 0),
$$

(S-48)

$$
\gamma_{\varphi_{\text{LR}}} = 2\nu_{\text{LR}}^2 S(\omega \to 0).
$$

Since $\nu_\text{L}$, $\nu_\text{R}$ and $\nu_{\text{LR}}$ indicate the sensitivity of exchange energies and the capacitive coupling to the charge noise,
they can be parameterized by $\partial J^\text{eff}_L/\partial \Delta$, $\partial J^\text{eff}_R/\partial \Delta$ and $\partial \alpha/\partial \Delta$, respectively. We therefore have

\[
\nu_L \propto \partial J^\text{eff}_L/\partial \Delta, \\
\nu_R \propto \partial J^\text{eff}_R/\partial \Delta, \\
\nu_{LR} \propto \partial \alpha/\partial \Delta, \\
\]

resulting in

\[
\gamma_{\varphi_L} = \tilde{\gamma}_{\varphi} \left( \frac{\partial J^\text{eff}_L/\partial \Delta}{[\partial J/\partial \Delta]_{\text{ref}}} \right)^2, \\
\gamma_{\varphi_S} = \tilde{\gamma}_{\varphi} \left( \frac{\partial J^\text{eff}_R/\partial \Delta}{[\partial J/\partial \Delta]_{\text{ref}}} \right)^2, \\
\gamma_{\varphi_{L_S}} = \tilde{\gamma}_{\varphi} \left( \frac{\partial \alpha/\partial \Delta}{[\partial J/\partial \Delta]_{\text{ref}}} \right)^2, \\
\]

where $\tilde{\gamma}_{\varphi}$ is the reference dephasing rate while $[\partial J/\partial \Delta]_{\text{ref}} \approx 5 \times 10^{-3}$ is the reference derivative of the exchange energy with respect to detuning, whose explicit value is extracted from $[S5]$. 

**B. Phonon mediated relaxation $\gamma_{\text{rel}}$ and the pure dephasing $\gamma_{\text{dep}}$**

Phonon mediated relaxation and pure dephasing rate can be obtained by adopting the Golden-Redfield theory $[S2]$. They are

\[
\gamma_{\text{rel}_{jk}} = J^+_j(\omega_{jk}), \\
\gamma_{\text{dep}_j} = J^+_\text{dep}_j(0), \\
\]

where $\gamma_{\text{rel}_{jk}}$ is the relaxation rate between the logical states $j$ and $k$, $\gamma_{\text{dep}_j}$ the pure dephasing rate of logical state $j$ and $\omega_{jk} = (E_k - E_j)/h$. The pure dephasing rates are

\[
\gamma_{\text{dep}_L} = J^+_{\text{dep}_{TT}}(0) - J^+_{\text{dep}_{SS}}(0) - \left( J^+_{\text{dep}_{ST}}(0) - J^+_{\text{dep}_{TS}}(0) \right), \\
\gamma_{\text{dep}_S} = J^+_{\text{dep}_{TT}}(0) - J^+_{\text{dep}_{SS}}(0) + \left( J^+_{\text{dep}_{ST}}(0) - J^+_{\text{dep}_{TS}}(0) \right), \\
\gamma_{\text{dep}_{L_S}} = J^+_{\text{dep}_{SS}}(0) - J^+_{\text{dep}_{ST}}(0) - J^+_{\text{dep}_{TS}}(0) + J^+_{\text{dep}_{TT}}(0). \\
\]

The r.h.s. of Eq. (S-51) can be expressed as:

\[
J^+_j(\omega) = \text{Re} \left[ J_{jk}(\omega) + J_{jk}(-\omega) \right] = \frac{g^2 \mu_B^2}{2h^2} \int_{-\infty}^{\infty} \cos(\omega \tau) \langle \delta B_{jk}(0) \delta B_{jk}(\tau) \rangle d\tau, \\
J^+_\text{dep}_j(0) = 2\text{Re} \left[ J_{\text{dep}_j}(0) \right] = \frac{g^2 \mu_B^2}{h^2} \int_{-\infty}^{\infty} \cos(\omega \tau) \langle \delta B_j(0) \delta B_j(\tau) \rangle d\tau \bigg|_{\omega \to 0}, \\
\]

where $\delta B(\tau) = e^{iH_{ph}\tau/h} \delta B e^{-iH_{ph}\tau/h}$ and $\delta B_{jk} = \text{Re} \left[ \delta B_{jk} \right] + \text{Im} \left[ \delta B_{jk} \right]$. The temperature-dependent correlator $\langle \delta B_j(0) \delta B_j(\tau) \rangle$ can be calculated by exploiting the relation: $\langle a_{q}^{\dagger} a_{q'}^{\dagger} \rangle = \delta_{q,q'} \delta_{\nu,\nu'} n_B(\omega_{\nu})$, where $n_B(\omega)$ is the Bose-Einstein distribution:

\[
n_B(\omega) = \frac{1}{e^{\hbar \omega/(k_B T)} - 1}. \\
\]

$k_B$ the Boltzmann constant and $T$ the temperature. In order to calculate $\tilde{U} H_{\text{ph}} \tilde{U}^\dagger$, we perform the Schrieffer-Wolff transformation up to the second order of electron-phonon coupling, corresponding to the contribution from two-phonon
Supplementary Figure S8: (a) Schematic figure of an STA pulse to perform high-fidelity state transfer in arbitrary time for a two-level system, based on Lewis-Riesenfeld invariants [S8]. The black (red) dashed line shows the detuning (tunnel coupling) as a function of time. (b) The leakage, $\eta$, as a function of the normalized time $t/\tau$, where $t/\tau = 0$ and 1 indicate the DDQD system residing at $(1,1,1,1)$ and $(2,0,0,2)$ charge configuration, respectively. At $t/\tau = 0$, the system is initialized as $|\Psi_0\rangle = \sum_{jk} 1/2 |jk\rangle$, where the sum is over all logical eigenstates, $(j,k) = (S,T)$. The dashed line shows the leakage when linear ramping (LIN) is adopted, while the solid line shows results from concatenated STA pulses as shown in (d). To facilitate comparisons, the ramping time for both LIN and STA schemes is $\tau_{\text{ramp}} = 7\text{ ns}$. (c) and (d): Concatenated STA pulses applicable for energy spectra of the logical states given by Fig. 2(d) in the main text and Supplementary Fig. S8(d). (c): Pulses performing a piecewise state transfer from $(1,1,1,1)$ to $(1,1,0,2)/(2,0,1,1)$) charge regime. (d): Pulses performing piecewise state transfer from $(1,1,1,1)$ to $(2,0,0,2)$ charge regime.

processes to the $\delta B$ term. The relaxation $\gamma_{\text{rel}}$, and the pure-dephasing rates $\gamma_{\text{dep}}$ are obtained for temperature $T = 20\text{ mK}$ [S5]. The phonon parameters are extracted from [S2] for the GaAs quantum-dot device.

VI. TWO-QUBIT GATE FIDELITY

In the main text, we evaluate the average gate fidelity $F$ as [S6, 7]:

$$F = \frac{dF_e + 1}{d + 1}, \quad (S-55)$$

where $d$ is the dimension of the system ($d = 4$ for a two-qubit system). The entanglement fidelity, $F_e$, for a noisy two-qubit gate is defined by setting the initial state as a maximally entangled state $|\Psi_0\rangle$ of four qubits, two of which is applied upon by the gate. To calculate $F$ for two-qubit gates on singlet-triplet qubits, the initial state is $|\Psi_0\rangle = \frac{1}{2} \sum_{j,k=S,T} |jk\rangle$, with the initial density matrix $\rho_{\Psi_0} = |\Psi_0\rangle \langle \Psi_0 | = \frac{1}{2} \sum_{j,k,m,n=S,T} |jk\rangle \langle mn| $. The resulting state after evolution in the noisy environment is then $|\tilde{\Psi}\rangle = (\mathcal{N}_g \otimes I) |\rho_{\Psi_0}\rangle$, where $\mathcal{N}_g = \tilde{U} \rho \tilde{U}^\dagger$ and $\tilde{U}$ encapsulates the noisy effects. The entanglement fidelity is then $F_e = \langle \Psi | (\mathcal{N}_g \otimes I) |\rho_{\Psi_0}| \langle \Psi \rangle$, where $|\Psi\rangle$ is the resulting state after an ideal evolution (without noise).

VII. PULSE SEQUENCES FOR AN ENTANGLING GATE

In this section, we provide more details on pulse sequences performing an entangling gate. We focus on two ramping schemes: linear ramp (LIN), and shortcut to adiabaticity (STA).

We assume that the system is initialized in the $(1,1,1,1)$ charge configuration, the detuning value for which is labeled by $\Delta_{\text{init}}$. This is to ensure that the capacitive coupling $\alpha$ is “off” (i.e. negligible). This setup also retains the coupling between logical states by the magnetic gradient $\Delta B$, i.e. $S[H_{\text{init}}|T] = \Delta B$. In order to perform an entangling gate operation, we need to ramp up the $\alpha$ value to another detuning point, which we denote as $\Delta_{\text{op}}$. $\Delta_{\text{op}}$ should be in the $(1,1,0,2)/(2,0,1,1)$ or $(2,0,0,2)$ charge regimes, where the nearly-sweet-spot regime exists.

The simplest technique to detune the system from $\Delta_{\text{init}}$ to $\Delta_{\text{op}}$ is a linear ramp on the detuning, i.e. $d\Delta/dt = \text{constant}$ (LIN scheme). The ramping time, denoted as $\tau_{\text{ramp}}$, must be on one hand long enough such that leakage due to the non-adiabatic effect is suppressed, and on the other hand short enough such that the exposure
of the system to various decoherence channels is limited. These conflicting requirements severely limit the practicality of the LIN scheme.

For a two-level system, the state transfer can be performed using shortcut to adiabaticity (STA) pulses generated by Lewis-Riesenfeld invariants [S8]. An example of the STA pulse is shown in Supplementary Fig. S8(a). To tailor the STA pulses for our purpose, it is useful to note that when the magnetic field is sufficiently strong, the avoided-crossing points for relevant charge states are well separated in detuning, making it possible to concatenate several elementary STA pulses. Taking the energy level structure shown in Supplementary Fig. S5(d) as an example: The (1,1,1,1) to (1,1,0,2)/(2,0,1,1) transitions for |S⟩, |ST⟩ and |TS⟩ occur at similar detuning values, Δ \( \approx 1.85\text{meV} \); The next charge transition, (1,1,1,1) to (1,1,0,2)/(2,0,1,1) transitions for |TT⟩ occurs at Δ \( \approx 2.15\text{meV} \); Transition into (2,0,0,2) from (1,1,0,2)/(2,0,1,1) for |SS⟩ and |ST⟩/|TS⟩/|TT⟩ occur at Δ \( \approx 2.24\text{meV} \) and 2.43meV respectively. In addition, the tunnel coupling between different charge configurations are all controlled by inter-dot tunneling, i.e. \( t_{12} = \langle \Phi_1|H|\Phi_2 \rangle \) for |SS⟩,|ST⟩ and |TS⟩ near \( \Delta^*_{[S\bar{S}],s} \), and \( t_{16} = \langle \Phi_1|H|\Phi_6 \rangle \) for |ST⟩, |TS⟩ and |TT⟩ near \( \Delta^*_{[T\bar{T}],e} \), both controlled by the barrier height between two dots within a DQD. Therefore, we place elementary STA pulses given by Supplementary Fig. S8(a) centering at the corresponding charge transition points, forming the concatenated piecewise STA pulses as shown in Supplementary Fig. S8(c) and (d). These concatenated STA pulse sequences allow state transfer of any arbitrary superposition of logical states as the input state.

We define the leakage \( \eta \) as

\[
\eta = \langle \Omega(t)|\Psi(t) \rangle, \quad (S-56)
\]

where \( \Omega(t) \) and \( \Psi(t) \) are the wavefunctions under adiabatic and non-adiabatic ramping at time \( t \). Supplementary Fig. S8(b) shows the leakage, \( \eta \) as a function of the normalized time, \( t/\tau \), for a DDQD system with \( B = 0.104T \) initialized in (1,1,1,1) and evolved into the (2,0,0,2) charge configuration. To illustrate the concatenated STA pulse sequences to perform state transfer on an arbitrary input state, the initial state is chosen to be the two-qubit superposition, \( |\Psi_0 \rangle = \sum \frac{1}{2}|j,k \rangle \), of a four-qubit maximally entangled state, \( |\Psi_0 \rangle = \sum \frac{1}{2}|j,k \rangle \) (cf. Sec. VI), where the sum is over all logical eigenstates, \( (j,k) = (S,T) \). It can be observed that the leakage is substantially suppressed for STA pulses as compared to the LIN scheme. The residual leakage observed for STA pulses arises from the weak, yet non-negligible, coupling to other higher-lying states apart from the effective two-level system centered at each charge-transition point.

[S1] E. Barnes, J. P. Kestner, N. T. T. Nguyen, and S. Das Sarma, Phys. Rev. B 84, 235309 (2011).
[S2] V. Kornich, C. Kloeffel, and D. Loss, Phys. Rev. B 89, 085410 (2014).
[S3] C. G. Van de Walle, Phys. Rev. B 39, 1871 (1989).
[S4] M. Boissonneault, J. M. Gambetta, and A. Blais, Phys. Rev. A 79, 013819 (2009).
[S5] J. M. Nichol, L. A. Orona, S. P. Harvey, S. Fallahi, G. C. Gardner, M. J. Manfra, and A. Yacoby, npj Quantum Inf. 3, 3 (2017).
[S6] M. A. Nielsen, Phys. Lett. A 303, 249 (2002).
[S7] M. Horodecki, P. Horodecki, and R. Horodecki, Phys. Rev. A 60, 1888 (1999).
[S8] X. Chen, E. Torrontegui, and J. G. Muga, Phys. Rev. A 83, 062116 (2011).