Inducing Spin-Correlations and Entanglement in a Double Quantum Dot through Nonequilibrium Transport

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For a double quantum dot system in a parallel geometry, we demonstrate that by combining the effects of a flux and driving an electrical current through the structure, the spin correlations between electrons localized in the dots can be controlled at will. In particular, a current can induce spin correlations even if the spins are uncorrelated in the initial equilibrium state. Therefore, we are able to engineer an entangled state in this double-dot structure. We take many-body correlations fully into account by simulating the real-time dynamics using the time-dependent density matrix renormalization group method. Using a canonical transformation, we provide an intuitive explanation for our results, related to Ruderman-Kittel-Kasuya-Yoshida physics driven by the bias.

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\textbf{Introduction:} Considerable progress in nanotechnology in the last decades has made possible the fabrication of new artificial structures \cite{1,2} such as quantum dots (QDs), quantum rings, or molecular conductors. The physics of quantum dots in a parallel geometry is intriguing, since it allows one to study interference effects between electrons traveling through different paths, most notably realized in the Aharonov-Bohm effect. Such structures have been studied in several experiments \cite{3,4}. Besides the interest in practical applications in nanoelectronics or in fundamental many-body physics such as the Kondo effect \cite{4,5}, double quantum dots (DQD) also play a vital role in the context of quantum information processing \cite{7,8,14}. Means of detecting entangled states of electrons were discussed in, e.g., Refs. \cite{11,15}. Generating, controlling, and detecting entangled states in condensed matter systems is one of the challenges for future quantum computation applications \cite{10}. Various proposals for entangling spatially separated electrons have been put forward, such as, for instance, by splitting Cooper pairs \cite{11,12} or by manipulating spins in quantum dots \cite{13}. In a DQD, an entangled state can be realized by putting the electrons into a singlet state \cite{7,8,14}. Means of detecting entangled states of electrons were discussed in, e.g., Refs. \cite{11,15}.

In this work, we demonstrate that an entangled state between electrons localized in a DQD embedded in an Aharonov-Bohm interferometer can be induced and controlled by sending an electrical current through the structure. In the presence of a flux, the initial state can even be fully uncorrelated yet the nonequilibrium dynamics results in nonzero spin correlations in the steady state. The sign and the strength of such steady-state spin correlations depend on voltage, interactions, and the flux. The generation of entanglement through nonequilibrium dynamics in quantum dots, with different set-ups, has been discussed in Refs. \cite{16}. An additional motivation for our work stems from the current interest, both from theory \cite{17,18} and from experiment (see, e.g., \cite{19}), in the nonequilibrium dynamics of nano-structures with strong electronic correlations. We emphasize that we treat both interactions and nonequilibrium dynamics in a well-controlled manner using the time-dependent density matrix renormalization group (DMRG) method \cite{20}. As we will see, the effect of inducing spin correlations is the largest at voltages \( \sim \Theta(W/4) \) (\( W \) is the bandwidth of the reservoirs) where Kondo correlations cease to matter \cite{2,3}.

\textbf{The model:} We model the quantum dots as Anderson impurities resulting in the Hamiltonian [depicted in the inset of Fig. 1(a)],

\begin{equation}
H = H_1 + H_{hy1} + H_{hy2} + H_{int},
\end{equation}

\begin{equation}
H_1 = \sum_{\alpha=L,R} \sum_{i=1}^{N-1} \left[ -t_0(c_{\alpha i}^\dagger c_{\alpha i+1} + h.c.) \right] + \sum_{\alpha=L,R} \sum_{i=1}^{N} \mu_{\alpha} n_{\alpha i},
\end{equation}

\begin{equation}
H_{hy1} = -t_1 [d_1^\dagger c_{L1}\sigma + c_{R1}\sigma d_1 + h.c.],
\end{equation}

\begin{equation}
H_{hy2} = -t_2 [d_2^\dagger c_{L1}\sigma + c_{R1}\sigma d_2 + h.c.],
\end{equation}

\begin{equation}
H_{int} = \sum_{j=1,2} \left[ U n_{ja} n_{j\sigma} + V_g n_{j}\sigma \right].
\end{equation}
The system size is $2N + 2$ where $N$ is the number of sites in the left or right lead. The two dots are at the center of the system labeled by $j = 1, 2$. The Hamiltonian consists of four parts: First, the non-interacting leads $H_I$ with a constant hopping matrix element $t_0 = 1$ used as the unit of energy ($\hbar = 1, e = 1$). Second, the terms $H_{Iy1}$ and $H_{Iy2}$ give rise to the hybridization between the localized levels of the dots and the leads. We consider fully symmetric tunnel couplings, i.e., $|t'_1| = |t'_2| = t'$ (see the supplementary material [21] for a discussion of asymmetric couplings). We define the tunneling strength by $\Gamma = 2rt^2\rho_{lead}(E_F) = 2t^2$, where $\rho_{lead}(E_F)$ is the local density of states (LDOS) of the leads at the Fermi energy $E_F$. In the hopping matrix element between the second dot and the right lead we incorporate an arbitrary phase $\phi$. Finally, there is the interacting region $H_{int}$ with the two quantum dots, which are both subject to the same Coulomb repulsion $U$ and a gate potential $V_g = -U/2$ such that both dots are kept at half filling. The operator $c^\dagger_{\alpha l\sigma}$ ($c_{\alpha l\sigma}$) creates (annihilates) an electron at site $l$ in the $\alpha = L, R$ lead with spin $\sigma$ while $d^\dagger_{j\sigma}$ ($d_{j\sigma}$) acts on dot $j$: $n_{\alpha l\sigma} = c^\dagger_{\alpha l\sigma}c_{\alpha l\sigma}$ as usual. In Eq. (1), $\mu_L$ and $\mu_R$ mimic the chemical potentials of the leads.

The ground state and the linear conductance of DQDs Eq. (1) were extensively studied in Ref. [22]. A closely related DQD model with a finite flux $\phi$ and with spin-polarized electrons was discussed in Ref. [23].

The phase included in Eq. (1) may have a different meaning depending on the specific physical realization. The most obvious one is to associate $\phi$ with a magnetic flux that pierces the ring structure containing the two dots and the first site from each lead as shown in Fig. 1(b). As usual, one can use a gauge transformation such that the flux appears in only one of the four hopping matrix elements. Another situation described by Eq. (1) is a single quantum dot with two levels where by symmetry the levels can couple with a phase difference to the leads.

We use DMRG [20] to obtain the steady state in the presence of a finite bias voltage by time-evolving the wavefunction $|\Psi(t)\rangle$ and then measuring its properties such as the current and spin correlations as a function of time $t$. This method has been successfully used to study nonequilibrium transport through nano-structures with electronic correlations [17, 24, 25]. We evaluate the spin correlations from [26]

$$S_{12}(t) = \langle \Psi(t)|\vec{S}_1 \cdot \vec{S}_2|\Psi(t)\rangle. \quad (6)$$

The current between two sites in the leads is defined as

$$J_{l,m}(t) = i\hbar t_0 \sum_\sigma \langle \Psi(t)|c^\dagger_{l\sigma}c_{m\sigma} - c^\dagger_{m\sigma}c_{l\sigma}|\Psi(t)\rangle. \quad (7)$$

In the figures, we display the current $J = (J_{L2,L1} + J_{R1,R2})/2$ averaged over the first link in the left and right lead.

Our simulations start from the system in equilibrium with a finite $\Gamma \neq 0$ and a charge per spin of $\langle n_{j\sigma} \rangle = 0.5$ on both dots. At time $t = 0$, we turn on a bias voltage $V = \mu_L - \mu_R$ that drives the system out of equilibrium. We work at large values of $\Gamma = 0.25$ such that the transient dynamics to reach the steady state is short [25]. The two quantum dots are treated as a super-site permitting the use of a Trotter-Suzuki breakup of $\exp(-iHt)$ [27]. The time step is $\delta t \approx 0.1$ and we enforce a fixed discarded weight [27] of $10^{-5}$ or less, keeping a maximum of 2000 DMRG states. All runs are performed at an overall half filling of dots and leads.

Results: In Fig. 1 we elucidate the time dependence of the current and spin correlations, comparing the behavior of $\phi = 0$ to $\phi = \pi$. Similar to a single quantum dot [25], the current undergoes transient dynamics, and then takes a quasi-stationary value (i.e., a plateau in time), which we shall refer to as the steady-state regime. Note that on finite systems, there is a system-size dependent revival time, resulting in a decay of the steady state current and a sign change (realized for $t \gtrsim 38$). For a discussion of transient time scales as well as an analysis of time dependent data for currents, see Ref. [25].

For the spin correlations shown in Fig. 1(b), we first observe that in the initial state, $S_{12} > 0$ for $\phi = 0$ whereas the correlation vanishes for $\phi = \pi$. The application of the bias voltage does virtually not affect the value of $S_{12}$ for $\phi = 0$, which remains positive. The more interesting behavior is realized for $\phi = \pi$. As a function of time, $S_{12}$ decreases and approaches a roughly constant value. The transient time is comparable to the one for the current and is of order $1/\Gamma$. Moreover, the transients are suppressed by increasing the bias, similar to a single quantum dot [25]. This finite and large spin correlation between the spins localized in the dots that emerges in the steady state and that is induced by driving a current through the structure is the main aspect of our work. It implies that nonequilibrium dynamics can be used to prepare a DQD in a correlated and thus entangled state.

To link the spin correlations to entanglement we use the concurrence $C_{12}$ [28, 29]. For instance, the concurrence approaches $C_{12} = 1$ if the spin correlation is $-3/4$ and if there are no charge fluctuations on the dots [21]. In Fig. 1(b) we include the concurrence versus time calculated for $\phi = \pi$. We observe that for $t = 0$ the concurrence is zero showing that
the dots are not entangled. Applying the bias, and after reaching the steady state for the spin correlations, the concurrence takes a value $C_{12} \sim 0.3$ corresponding to a finite entanglement between the dots.

The qualitative behavior of the spin correlations can be understood by using a canonical transformation of the states of the leads, which is given by (see, e.g., [23, 30]):

$$c_{\gamma\ell\sigma} = (c_{R\ell\sigma} \pm c_{L\ell\sigma})/\sqrt{2},$$

where $\gamma = s, a$ are the symmetric and antisymmetric combinations, respectively. The result of this transformation is sketched in Fig. 2, where the leads shown represent the new states obtained from Eq. (8). In the absence of a bias voltage, there is no direct coupling between these new states, as depicted in Figs. 2(a,1) and (b,1). Most importantly, the dots are coupled to only the symmetric states for $\phi = 0$, whereas for $\phi = \pi$, dot $j = 1$ is coupled to the symmetric states and dot $j = 2$ to the antisymmetric ones. For $\phi = 0$, the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction gives rise to a ferromagnetic correlation between the dots since each lead, which is given by (see, e.g., [23, 30]):

$$\Gamma = \frac{1}{2} \sum_{\ell} \left( c_{R\ell\sigma} c_{L\ell\sigma} + c_{R\ell\sigma} c_{L\ell\sigma}^* \right),$$

where $\Gamma$ is the steady-state values by $\langle S_{12} \rangle$ and $\langle J \rangle$, obtained from averaging over time-dependent data in the steady-state regime (compare Ref. [23]). Figure 3(a) shows $\langle J \rangle/V$ versus $V$ for phases $\phi = 0$ and $\pi$. For $\phi = 0$, $\langle J \rangle/V$ approaches a constant value at low bias [23]. For $\phi = \pi$, the linear conductance vanishes due to the Aharonov-Bohm effect [23]. A finite voltage causes a finite current to flow in both cases, but $\langle J \rangle/V$ for $\phi = 0$ is always larger than in the $\phi = \pi$ case.

In Fig. 3(b), we display the steady-state spin correlations $\langle S_{12} \rangle$ versus $V$. First, let us emphasize that data for the steady-state values obtained from systems of different lengths are included, showing that all our main results are quantitatively robust against finite-size effects. For $\phi = 0$, a constant value of $\langle S_{12} \rangle > 0$ is found. A slight decrease appears for $V \gtrsim 1$, which we trace back to the variation of the LDOS of the leads seen by the dots. Since in our simulations we work with tight-binding bands with a finite band-width and band curvature, this LDOS decreases with $V$. For the case of $\phi = \pi$, the value of the steady-state correlations can be tuned by the bias voltage and in fact, $\langle S_{12} \rangle$ increases with $U$. Therefore, to obtain a strong correlation a large voltage is needed putting the system out of the Kondo regime. As the figure clearly shows, we can get to $\langle S_{12} \rangle \approx -0.25$ for $U/T = 2$. We therefore realize a mixed state with singlet correlations dominating over triplet correlations. If there were no charge fluctuations then this value of $S_{12}$ would correspond to a Werner state [51, 32] with 50% of the weight in the singlet. In our case, however, the current needed to obtain the entangled state typically induces charge fluctuations at large $V$. Therefore, $\langle S_{12} \rangle \leq -1/4$ implies an even larger relative contribution of the singlet over the triplet than in a situation without any charge fluctuations. The fact that a finite voltage unavoidably induces charge fluctuations is the reason why the steady-state spin correlations do not reach their largest possible negative value of $-3/4$.

The steady-state values further depend on $U/T$. To elucidate this, we plot $\langle S_{12} \rangle$ versus $U$ in the inset of Fig. 3(b) for a fixed value of $V = 0.5$, by increasing $U/T$ to 4, i.e., in a regime where charge fluctuations are still relevant even in equilibrium. As expected, the larger $U$, the more strongly charge fluctuations are suppressed, leading to larger steady-state spin correlations. Therefore, either a large $U/T$ at a fixed voltage or applying a large voltage order of $V \sim t$ induces the largest steady-state correlations. Fortunately, many experiments with DQDs realize $U/T \gtrsim 10$ [4, 5], thus relaxing the requirement on voltage. An important role of $U$ is to define a local spin as in many other quantum information application
of QDs \[8\].

So far we have investigated the dependence of correlations on \(V\), \(U\) and \(\phi\) in nonequilibrium, comparing the cases of \(\phi = 0\) to \(\phi = \pi\). An additional degree of tunability can be added if the phase can take arbitrary values (see Fig. S1 in the supplementary material [21]). As expected from the discussion of Fig. 2, \(\langle S_{12} \rangle\) is positive for small \(\phi\) at \(V = 0\) and then decreases to zero as \(\phi = \pi\) is approached. This transition to the uncorrelated case of \(\phi = \pi\) is continuous. At a finite voltage, it is possible to go from positive steady-state correlations to negative ones by changing \(\phi\). For the parameters of Fig. 3a, the steady-state correlations change sign at \(\phi_c \approx 0.18\pi\) (see Fig. S1). This value depends both on \(U\) and \(V\). To summarize, the steady-state correlations can be tuned both in sign and magnitude by changing \(V\), \(\phi\), and \(U\). We have further verified that the steady-state correlations are independent of the initial conditions [21].

Based on the qualitative picture developed so far, we conclude that the steady-state correlations are a result of mixing the symmetric and antisymmetric states of lead electrons in nonequilibrium. At finite \(U\), this may be viewed as an RKKY effect in nonequilibrium. A discussion on how to estimate the effective indirect coupling \(J_{eff}(V)\) induced by the bias can be found in the supplementary material [21]. As is well known from the physics of the RKKY effect in equilibrium the spin correlation induced by indirect exchange is destroyed for temperatures larger than \(T \approx 33\) [23]. For the nonequilibrium version of RKKY discussed here, we expect that temperatures should be smaller than the effective strength \(J_{eff}(V)\) shown in [21] for thermal fluctuations not to affect the induced correlations.

Finally, we study the behavior of \(S_{12}^{(t)}\) under quenches of parameters of the Hamiltonian Eq. (1). We proceed as before, i.e., a finite bias voltage \(V \neq 0\) is turned on at \(t = 0\), and in addition we instantaneously change some of the tunnel couplings at a time \(t_q \geq 0\).

We find that if we disconnect the quantum dots from the leads at time \(t_q > 0\) by setting \(t'_1 = t'_2 = 0\) after the steady state has been established, as expected, the spins remain in a correlated state after isolating them from the reservoirs (see Fig. S4(b) in [21]).

In a second example, after reaching the steady state, we isolate one of the dots while the current continues to flow through the other. This results in the loss of the spin correlations after a short transient time (see Fig. S4(c) in [21]). Therefore, control over the tunneling matrix elements allows one to put the system back into its original uncorrelated state. Both the generation of entanglement and the removal happen on short times scales, similar to the proposals discussed in [16].

Summary: In this work, we demonstrated that spin correlations between spatially separated electrons localized in a parallel QD embedded in the rings of an Aharonov-Bohm interferometer can be induced and modified by driving a current through the structure. The steady-state correlations depend on voltage, the flux, and Coulomb interactions. Control over the individual tunneling couplings would allow one to isolate the entangled spins from the environment or to remove the entanglement again. The mechanism behind this time-dependent formation of correlations can be thought of as an RKKY effect in nonequilibrium. Our results may be relevant for applications of DQD structures in quantum information processing.

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Supplemental material for Inducing Spin Correlations and Entanglement in a Double Quantum Dot through Nonequilibrium Transport

DEPENDENCE OF STEADY-STATE SPIN CORRELATIONS ON THE PHASE

Figure 4 shows the dependence of the spin correlations on the phase $\phi$, both in equilibrium ($V = 0$), and in the steady state ($V = 0.5$). At a finite voltage, it is possible to go from positive spin correlations to negative ones by changing $\phi$. For this set of parameters, the steady-state correlations change sign at $\phi_c = 0.18$. See main article for further discussion.

CONCURRENCE

In this Section we use the concurrence to quantify the entanglement (see Refs. [26] and [27] in the main paper). First we define the single fermion operator at the dots 1,2 as

$$N_i^s = n_{i\uparrow} + n_{i\downarrow} - 2 n_{i\uparrow} n_{i\downarrow}. \quad (9)$$

This operator projects onto the subspace with exactly one fermion on the dot $i$. Using $N_i^s$ the concurrence (see Ref. [27]) can be written as

$$C_{12}(t) = \max \left\{ 0, -\frac{1}{2} - \frac{2}{\langle \Psi(t) | N_1^s N_2^s | \Psi(t) \rangle} \right\}. \quad (10)$$

Note that the concurrence takes its maximum value 1 when $S_{12}(t)/\langle \Psi(t) | N_1^s N_2^s | \Psi(t) \rangle \rightarrow -3/4$. One example to get this is $\langle S_{12} \rangle = -3/4$ and $\langle \Psi(t) | N_1^s N_2^s | \Psi(t) \rangle = 1$.

In Figure 5 we present the results for the average of the concurrence in the steady state as a function of the bias $V$ and the phase $\phi$. In panel (a) we see that for $V > 0.25$ the concurrence starts to be non-zero showing that the singlet starts to be relevant. Increasing the bias the concurrence grows monotonically at the same time as the spin correlations.

In this reference system we then calculate the equilibrium correlations $S_{12} = S_{12}(J_{\text{eff}})$ in this reference system. In the next step, we use the numerically determined function $\langle S_{12} \rangle = \langle S_{12} \rangle(V)$ and by equating it to $\langle S_{12} \rangle$, we obtain $J_{\text{eff}} = J_{\text{eff}}(V)$, keeping all other parameters fixed in both the time-dependent simulation and in the reference system. The results are presented in Fig. 6 and its inset.

EFFECTIVE RKKY COUPLING

As discussed in the main article an effective spin coupling, similar to the RKKY effect, emerges when the bias is applied. To estimate the strength $J_{\text{eff}}$ of the effective coupling between the localized spins in the dots in the steady state, we proceed as before. First, in the set-up shown in Fig. 2(b.1) of the main article, we connect the impurities by an additional term $J_{\text{eff}} \hat{S}_1 \cdot \hat{S}_2$ and we then calculate the equilibrium correlations $S_{12} = S_{12}(J_{\text{eff}})$ in this reference system. In the next step, we use the numerically determined function $\langle S_{12} \rangle = \langle S_{12} \rangle(V)$ and by equating it to $\langle S_{12} \rangle(J_{\text{eff}})$, we obtain $J_{\text{eff}} = J_{\text{eff}}(V)$, keeping all other parameters fixed in both the time-dependent simulation and in the reference system. The results are presented in Fig. 6 and its inset.

QUENCHES

In this section we show how robust is the steady-state spin correlations against quenching certain parameters of the Hamiltonian Eq. (1). As explained in the main text we proceed as before. The finite bias voltage $V > 0$ is turned on at $t = 0$. In addition we perform quenches of either the phase $\phi$ or some of the tunnel couplings at a time $t_q \geq 0$.

The first example, presented in Fig. 6(a), consists of changing the phase from 0 to $\pi$. We show results for $t_q = 0$ and 10
and observe that in both cases, the spin correlations evolve from their initial constant and positive value to a negative value that only depends on parameters in the steady state (i.e., \( V \) and \( \phi \)), but as this comparison shows, is virtually independent of the specific transient dynamics.

In the second case, presented in Fig. 7(b), we simply disconnect the quantum dots from the leads at time \( t_q > 0 \) after the steady state has been established. The spins remain in a correlated state after isolating them from the reservoirs.

Third, after reaching the steady state, we isolate one of the dots while the current continues to flow through the other one. As a consequence the spin correlations is destroyed after a short transient time [see Fig. 4(c)].

### ASYMMETRIC TUNNEL COUPLINGS

In this section we show that the key results of our work described in the main text for a DQD with symmetric tunnel couplings carrying over to the asymmetric case. We start from the most general form of the Hamiltonian from Eqs. (3) and (4) of the main text. The hybridization between the localized levels of the dots and the leads can be written as

\[
H_{hy1} = -t'_{L1}d^\dagger_{\sigma}c_{L1\sigma} - t'_{R1}d^\dagger_{\sigma}c_{R1\sigma} + \text{H.c.},
\]

\[
H_{hy2} = -t'_{L2}d^\dagger_{\sigma}c_{L1\sigma} - t'_{R2}d^\dagger_{\sigma}c_{R1\sigma} + \text{H.c.},
\]

(12)

where tunnel matrix elements \( t_{\alpha j} \), are considered that can in general be different for each dot \( (j = 1, 2) \) and lead \( (\alpha = L, R) \). The tunneling strength is then defined by \( \Gamma_j = t'_{Lj}^2 + t'_{Rj}^2, j = 1, 2. \) For each dot we introduce an angle \( \theta_j \), defined in Eq. (14) appears directly in \( H_{hy1} \) and \( H_{hy2} \) can be re-expressed as

\[
H_{hyj} = -\sqrt{\Gamma_j}d^\dagger_{\sigma} (\sin \theta_j c_{L1\sigma} + \cos \theta_j c_{R1\sigma} + \text{H.c.}) .
\]

We next generalize the canonical transformation of the lead operators that was discussed in the main text in the following way:

\[
c_{\mu i\sigma} = (\sin \theta_1 c_{L1\sigma} + \cos \theta_1 c_{R1\sigma}) ,
\]

(14)

\[
c_{\nu i\sigma} = (\cos \theta_1 c_{L1\sigma} - \sin \theta_1 c_{R1\sigma}) .
\]

(15)

In the special case of \( \theta_1 = \pi/4 \), this reduces to Eq. (8) from the main text with \( \mu = s \) and \( \nu = a \). Note that the operator \( c_{\mu i\sigma} \) defined in Eq. (14) appears directly in \( H_{hy1} \). Therefore, the Quantum Dot (QD) \( j = 1 \) is just connected to the \( \mu \) channel. The resulting configuration, i.e., QD 1 coupled to the \( \mu \)-channel and QD \( j = 2 \) coupled to either the \( \mu \), \( \nu \), or both channels is depicted in Fig. 8.

The Hamiltonian of the leads \( H_L \) with chemical potential \( \mu_R = -\mu_L = V/2 \) is, under this transformation, given by

\[
H_1 = H_c + H_V ,
\]

(16)

\[
H_c = -t_0 \sum_{\gamma = \mu, \nu} \sum_{i = 1: \sigma} \left( c^\dagger_{\gamma i\sigma}c_{\gamma i+1\sigma} + \text{H.c.} \right) ,
\]

(17)

\[
H_V = \sum_{i = 1: \sigma} \left[ t_p (c^\dagger_{\nu i\sigma}c_{\mu i\sigma} + \text{H.c.}) + V_\mu n_{\mu i\sigma} \right] ,
\]

(18)

with

\[
t_p = -V \sin \theta_1 \cos \theta_1 ,
\]

(19)

\[
V_\mu = -V_\nu = \frac{V}{2} (\cos^2 \theta_1 - \sin^2 \theta_1) .
\]

(20)

Similar to the case of fully symmetric couplings shown in Fig. 2 of the main text, the bias potential connects the channels \( \mu \) and \( \nu \) through an effective coupling \( t_p \). This coupling takes its maximum value in the case of symmetric couplings for QD \( j = 1 \), i.e., \( t'_{L1} = t'_{R1} \) and therefore, \( \theta_1 = \pi/4 \).

Next, the second QD is connected to the new channels \( \mu \)
and $\nu$ through:

$$H_{\text{hy2}} = \sum_{\sigma} \left( t'_{\mu2} d_{\mu\sigma}^\dagger c_{\mu\sigma} + t'_{\nu2} d_{\nu\sigma}^\dagger c_{\nu\sigma} + \text{H.c.} \right). \quad (21)$$

It is straightforward to verify that $|t'_{\mu2}|^2 + |t'_{\nu2}|^2 = \Gamma_2$.

Figure 9 shows, for $\phi = 0, \pi/2$ and $\pi$, the square modulus of $t'_{\mu2}$ and $t'_{\nu2}$ as a function of $\theta_1$ and $\theta_2$ for $U = 0.5$, $\Gamma_1 = \Gamma_2 = 0.25$, $N = 35$. For $\phi = 0$ and $\phi = \pi$ we observe diagonal stripes where $t'_{\mu2}$ or $t'_{\nu2}$ vanish resulting in the situation depicted in Figs. 2 (a) and (b) from the main text. For $\phi = 0$ it is obvious that, if $\theta_2 = \theta_1$, the canonical transformation will decouple both QDs from the $\nu$-channel at the same time as both dots have the same asymmetry.

In order to reproduce the features of Fig. 2 of the main text for the case of asymmetric couplings, we require that either $t'_{\mu2}$ or $t'_{\nu2}$ be close to zero. To illustrate we have selected eight parameter sets for the tunneling matrix elements and the flux $\phi$ that reproduce such situations, which we list in Table I. Each set of parameters contained in this table is labeled by a letter A, B, . . . , G.

Set A is the completely symmetric situation with $\phi = 0$ that we discussed in the main text. In this case, $t'_{\mu2}/\sqrt{2} = 1$ and $t'_{\nu2} = 0$. Therefore, QD $j = 1$ and $j = 2$ are connected to the same effective lead and the spin correlations are positive in the steady state. A similar behavior is realized for sets B and C. Set B corresponds to a situation in which $\phi = \pi$ while set for C, we choose $\phi = 0.2\pi$. Note that in set B the phase changes the sign of $t'_{R2}$ and the situation is thus similar to set A. In set C, all four couplings, $t_{L1}, t'_{L1}, t'_{L2}$, and $t'_{R2}$, are different from each other and for the selected value of the phase $\phi = 0.2\pi$, no signs are affected.

The examples labeled D to G are cases in which $t'_{\mu2}$ is small, and therefore, where, according to the picture put forward in the discussion of Fig. 2(b) of the main text, we expect negative spin correlations in the steady state. Set D is the completely symmetric case with $\theta_1 = \theta_2 = \pi/4$ and $\phi = \pi$. This case was analyzed in the main article and is shown here for comparison. For set E, although $\phi = 0$, there is a difference of $\pi/2$ between $\theta_1$ and $\theta_2$, resulting in a behavior similar to set D, despite the left-right asymmetry present in set E. Note that $t'_{L2}$ is negative as a consequence, and therefore, the steady-state spin correlations are also negative, in accordance with the discussion of the main article. The set F realizes a case with $\phi = \pi/2$ and a small $t'_{R2} \sim 0.09$. In this example, left-right symmetry is broken but there are still just two different values for the four tunnel matrix elements $t'_{\nu2}$. Finally, in set G, $\phi = 0.2\pi$ and all four tunnel matrix elements are different. The very small value of $t'_{\mu2}$ implies negative steady-state spin correlation as before.

To verify our predictions for the sign of steady-state spin correlations that are based on the numbers listed in Table I, we have calculated the spin correlation as a function of time. The results are shown in Fig. 10. Sets A, B and C, as they all have a small value of $t'_{\mu2}$, lead to a spin correlation that is positive in the initial state ($t = 0$) whose sign and absolute value are barely affected by the bias at all. Sets D, E, F and G all have a small value for $t'_{\nu2}$. As a consequence, the dots are predominantly connected to one of the two channels $\mu$ or $\nu$ (compare Fig. 2(b.1) of the main text) and spin correlations vanish in equilibrium, i.e., at for $V = 0$. Upon applying a bias, negative spin correlations emerge, as expected. Note the small value of the steady-state spin correlation in set F, consistent with the observation that in this case, the value of the coupling between the effective leads $t_p$ is the smallest.

As a conclusion, even for asymmetric tunnel couplings between the DQD and the leads the application of a voltage can

| Set | $\theta_1/\pi$ | $\theta_2/\pi$ | $t'_{L1}$ | $t'_{L2}$ | $t'_{R1}$ | $t'_{R2}$ | $\phi/\pi$ | $|t'_{\mu2}|^2/\Gamma_2$ | $|t'_{\nu2}|^2/\Gamma_2$ | $t_p/V$ | $V_L/V$ | $\langle S_{12} \rangle$ |
|-----|----------------|----------------|------------|------------|------------|------------|-------------|----------------|----------------|-----------|-------------|----------------|
| A   | 0.25           | 0.25           | 0.353      | 0.353      | 0.353      | 0.353      | 0.0         | 1.000         | 0.000         | 0.500     | 0.000       | +             |
| B   | 0.09           | 0.89           | 0.480      | 0.169      | -0.470     | 1.000      | 0.996       | 0.004         | 0.267         | 0.422     | +           |               |
| C   | 0.48           | 0.47           | 0.499      | 0.497      | -0.047     | 0.2         | 0.997       | 0.003         | 0.067         | -0.496    | +           |               |
| D   | 0.25           | 0.25           | 0.353      | 0.353      | 0.353      | 1.000      | 0.000       | 1.000         | 0.500         | 0.000     | –           |               |
| E   | 0.14           | 0.64           | 0.213      | 0.452      | -0.213     | 0.452      | 0.0         | 0.000         | 1.000         | 0.385     | –           |               |
| F   | 0.07           | 0.43           | 0.488      | 0.488      | 0.110      | 0.5         | 0.090       | 0.910         | 0.212         | 0.452     | –           |               |
| G   | 0.12           | 0.63           | 0.184      | 0.465      | 0.459      | -0.198     | 0.0         | 0.049         | 0.951         | 0.342     | –           |               |

TABLE I. Relationship between the bare parameters of the system [see Eqs. (11) and (12)] and the tunnel matrix elements of the effective channels $\mu$ and $\nu$ [compare Eqs. (13) and (21)]. In this table we show a few examples for which the discussion of the main article applies even for an asymmetric coupling. We choose $\Gamma_1 = \Gamma_2 = 0.25$ for simplicity. In the last column we list the expected sign of the spin correlations.

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**FIG. 8.** Sketch of the representation of the DQD structure and its coupling (a) the original leads and (b) the effective leads defined in Eqs. (13) and (15). We refer to the effective leads as the $\mu$- and $\nu$-channel, respectively.
FIG. 9. Square modulus of the couplings between QD $j = 2$ and the effective channels $\mu$ and $\nu$ defined in Eq. (14) and (15). All curves are for $\Gamma_2 = 0.25$, $U = 0.5$, $V_g = -U/2$ and $N = 35$. To reproduce the features of Figs. 2(a) and (b) of the main text, we need to require $|t'_{\mu2}|^2 = 0$ or $|t'_{\nu2}|^2 = 0$, respectively. Note that $t'_{\mu2}$ and $t'_{\nu2}$ do not depend on $\Gamma_1$.

FIG. 10. Spin correlations as a function of time for the parameter sets presented in Table I. Note that for sets with small values of $t'_{\nu2}$, such as A, B and C, the spin correlations do practically not change when the bias is applied. By contrast, whenever $t'_{\mu2}$ is small, as is the case in sets D, E, F and G, the spin correlations in the steady state are negative. Sets A and D are the cases discussed in the main text, compare Fig. 1(b).