Nonmonotonic buildup of spin-singlet correlations in a double quantum dot

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Dynamical buildup of spin-singlet correlations between the two quantum dots is investigated by means of the time-dependent numerical renormalization group method. By calculating the time evolution of the spin-spin expectation value upon a quench in the hopping between the quantum dots, we examine the timescales associated with the development of an entangled spin-singlet state in the system. Interestingly, we predict a nonmonotonic buildup of entanglement between the two dots. In particular, we find that in short timescales the effective exchange interaction between the quantum dots is of ferromagnetic type, favoring spin-triplet correlations, as opposed to the long-time limit, when strong antiferromagnetic correlations develop and eventually an entangled spin-singlet state is formed between the dots. We also numerically determine the relevant timescales and show that the physics is generally governed by the interplay between the Kondo correlations on each dot and exchange interaction between the spins of both quantum dots.

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I. INTRODUCTION

Double quantum dot systems coupled by a tunable tunnel barrier are important, prototypical structures allowing for convenient control and manipulation of the electronic occupation and spin degrees of freedom [1–14]. Such systems have more complex electronic structure than single quantum dots [15–17] and therefore reveal further interesting properties, phases, and dynamics resulting from strong correlations [18–24]. Moreover, the ability to trap and control electron spins in quantum dots opens up new possibilities for promising applications in the field of quantum computing [25–32]. In fact, the generation and manipulation of quantum states of individual electrons in the dots enabled the creation of qubits with high fidelity and long coherence times [33–44]. Coupled double quantum dot systems are in particular widely considered as hosts for exchange qubits, since they grant an overall tunability and increased stability due to noise and decoherence suppression. Additionally, such systems provide several schemes for fast manipulation and also the possibility to form three-electron qubits [45–53].

Nevertheless, for efficient exploitation of quantum dots in solid-state quantum information, it is of importance to fully understand the relevant dynamical behavior, when also correlation effects may play an important role. In this regard, the dynamics and transient behavior of double quantum dot systems remains still a quite unexplored problem, demanding very precise theoretical and experimental studies [54–67]. In this work we therefore focus on theoretical study of quench dynamics of a half-filled double quantum dot following an abrupt switching on of the coupling between the two quantum dots, that were initially coupled to external electronic reservoirs, but isolated from each other. The dynamical buildup of spin-singlet correlations between the two quantum dots is investigated by means of the time-dependent numerical renormalization group (tdNRG) method [68–73]. By calculating the time-evolution of the spin-spin expectation value upon a quench, we examine the timescales associated with the development of an entangled spin-singlet state in the system. We analyze the development of the spin correlations in the double quantum dot mediated by a direct hopping between the dots. We show that the spin-singlet correlations start to compete with Kondo correlations formed in the initial state, and for considerable values of exchange interaction lead to a singlet state formed between two dots. Interestingly, when the dynamics is driven by exchange coupling values significantly exceeding the Kondo temperature T_K, there is a development of ferromagnetic order at short timescales, which precedes the formation of an entangled spin-singlet state in the long-time limit.

The following is an outline of how this paper is organized. In Sec. II we describe the model and briefly introduce the method used in numerical calculations. In Sec. III the relevant static properties of the system are presented and discussed, while the main results and discussion of spin dynamics are in Sec. IV. The paper is concluded in Sec. V.

II. MODEL AND METHOD

The considered system is presented in Fig. 1(a). It consists of two quantum dots, each attached to its own metallic reservoir, and coupled to each other through the hopping matrix element $v(t)$. The Hamiltonian of such a device can be written as

$$H = H_r + H_{DD} + H_T, \tag{1}$$

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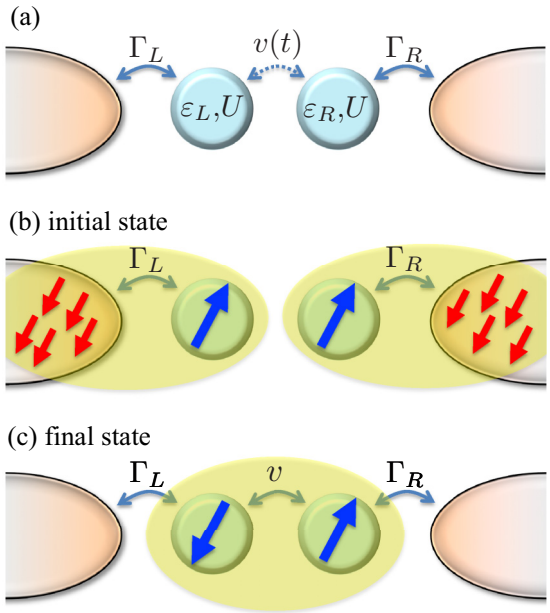

FIG. 1. (a) The schematic of the considered double quantum dot system. Each quantum dot is coupled to its metallic lead with coupling strength Γ_α, with $\alpha = L$ ($\alpha = R$) for the left (right) lead. ε_L and ε_R denote the orbital level energies in the corresponding dots, while U stands for the Coulomb correlations. The quantum dots are coupled through the hopping matrix element $v(t)$. We study the evolution of the system following the quench in $v(t)$, as schematically presented in (b) and (c). (b) In the initial state, $t < 0$, $v = 0$ and the Kondo state develops separately on each quantum dot. (c) In the final state, $t \to \infty$, a spin singlet state forms between the two quantum dots, provided that the antiferromagnetic exchange interaction between the dots is larger than the corresponding Kondo temperature.

where the first term describes the electrodes in the free quasiparticle approximation,

$$H_r = \sum_{\alpha=L,R} \sum_{k\sigma} \varepsilon_{\alpha k\sigma} c^\dagger_{\alpha k\sigma} c_{\alpha k\sigma}. \tag{2}$$

Here, $c^\dagger_{\alpha k\sigma}$ ($c_{\alpha k\sigma}$) creates (annihilates) an electron of momentum k and spin σ in the left ($\alpha = L$) or right ($\alpha = R$) lead with the corresponding energy $\varepsilon_{\alpha k\sigma}$. The second term H_{DD} models the double quantum dot system and is given by

$$H_{DD} = \sum_{\alpha=L,R} \sum_{\sigma} \varepsilon_\alpha d^\dagger_{\alpha\sigma} d_{\alpha\sigma} + \sum_{\alpha=L,R} U d^\dagger_{\alpha\uparrow} d_{\alpha\uparrow} d^\dagger_{\alpha\downarrow} d_{\alpha\downarrow} + v \sum_{\sigma} (d^\dagger_{L\sigma} d_{R\sigma} + d^\dagger_{R\sigma} d_{L\sigma}), \tag{3}$$

with $d^\dagger_{\alpha\sigma}$ ($d_{\alpha\sigma}$) being the creation (annihilation) operator on the left ($\alpha = L$) or right ($\alpha = R$) quantum dot for an electron of spin σ, ε_α denotes the corresponding orbital level energy, and U stands for the Coulomb correlations assumed to be equal for both dots. The hopping amplitude between the two dots is given by v. Without loss of generality, we assume that each dot is at half filling, $\varepsilon_\alpha = -U/2$, and that the system is symmetric, $\Gamma_L = \Gamma_R = \Gamma$. We also note that generally in double quantum dot systems, depending on particular material realization, there might be a finite spin-orbit interaction [74–76]; however, in our analysis we assume that the effects associated with this interaction are negligible, i.e., the strength of spin-orbit coupling is the smaller than any other relevant energy scale.

We are interested in the development of spin correlations between the two quantum dots when turning on the hopping v; see Figs. 1(b) and 1(c). In particular, we determine the time dependence of the spin-spin expectation value,

$$S_{LR}(t) \equiv \langle \Psi(t) | \vec{S}_L \cdot \vec{S}_R | \Psi(t) \rangle, \tag{4}$$

which quantifies the formation of the singlet state between the quantum dots. Here, $\vec{S}_\alpha = \frac{1}{2} \sum_{\sigma\sigma'} d^\dagger_{\alpha\sigma} \vec{\sigma}_{\sigma\sigma'} d_{\alpha\sigma'}$ is the spin operator of the dot α, with $\vec{\sigma}$ denoting the vector of Pauli spin matrices. The many-body state of the system $|\Psi(t)\rangle$ evolves according to the full Hamiltonian H,

$$|\Psi(t)\rangle = e^{-iHt} |\Psi(0)\rangle, \tag{5}$$

where $|\Psi(0)\rangle$ is the initial state that we prepare assuming $v = 0$. To be able to resolve the system's dynamics in the presence of strong electronic correlations in the most accurate manner, we employ here the time-dependent numerical renormalization group method [68–73].

The general form of the considered time-dependent Hamiltonian is given by

$$H(t) = \Theta(-t) H_0 + \Theta(t) H. \tag{6}$$

Here, the Hamiltonian H_0 describes the system before quantum quench in initial equilibrium for $t < 0$. Then, the final Hamiltonian H models the system for $t \geqslant 0$, when a sudden change in H is performed with respect to H_0. In the considered case, it is switching on the hopping between the quantum dots in a steplike fashion at time $t = 0$. In tdNRG both Hamiltonians are diagonalized in the Wilson chain geometry by using the numerical renormalization group method [69,77–80]. The discarded states determined in this iterative procedure, including all states found in the last iteration, are used to build a full many-body eigenbases of the Hamiltonians. In the next step, we determine the dynamical quantities. In order to obtain the relevant time dependencies, the discrete data in form of Dirac delta peaks is collected and then Fourier transformed into the time domain [73,79]. It is important to mention that tdNRG is a fully nonperturbative method, and, as such, it allows us to obtain high quality data where all correlations, also the ones driving the Kondo effect, are taken into account on an equal footing.

III. STATIC PROPERTIES

To begin our discussion, in Fig. 2 we present the static expectation value of S_{LR} calculated as a function of the hopping between the dots for different values of the coupling to external contacts. One can see that S_{LR} depends in a nonmonotonic manner on v. To understand this behavior, one needs to realize that there are two competing energy scales in the system. The first one is associated with the Kondo effect, which, in the absence of hopping between the dots, develops in each quantum dot separately. The Kondo temperature depends in an exponential way on the ratio of U/Γ as predicted by Haldane's formula [81], $T_K = \sqrt{U\Gamma/2} \exp(-\pi U/8\Gamma)$. On the

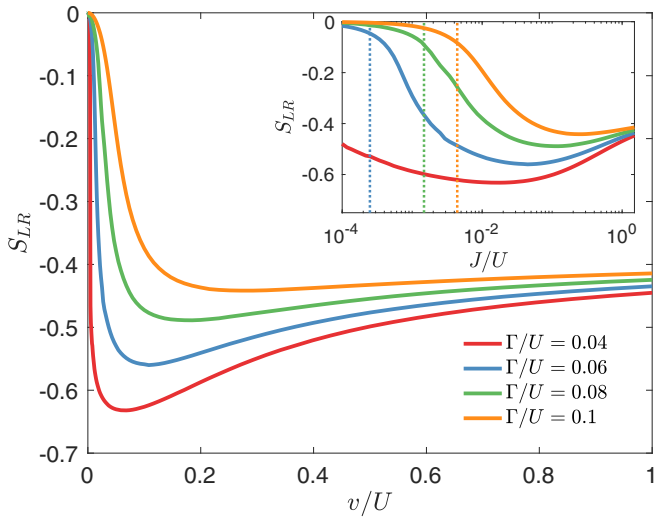

FIG. 2. The static expectation value of S_{LR} calculated as a function of the hopping between the quantum dots v for different values of the coupling to the contacts Γ, as indicated. The inset presents the dependence of S_{LR} on the exchange interaction J, where the dashed lines indicate the Kondo temperature T_K for corresponding values of Γ, as estimated from Haldane's formula [81]. Note the logarithmic x-axis scale in the inset. The parameters are $\varepsilon_L = \varepsilon_R = -U/2$ and $U = 0.1$ in units of band half-width.

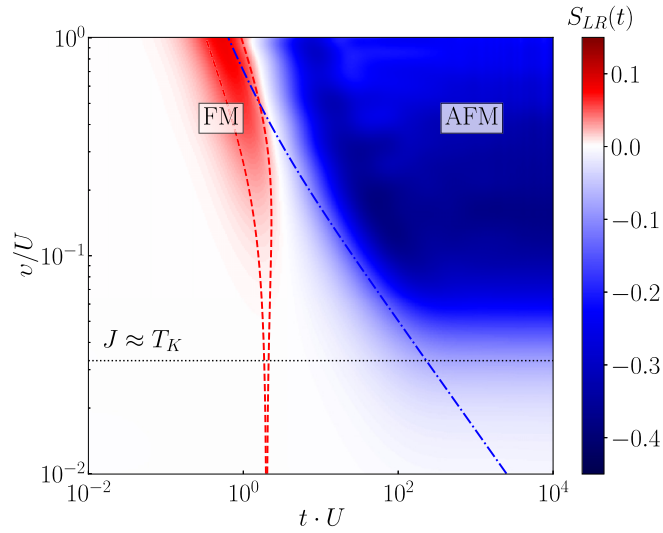

FIG. 3. The expectation value $S_{LR}(t)$ calculated as a function of time t and the hopping between the dots v. The system is prepared in the initial state with $v = 0$ and then time evolved according to the full Hamiltonian with a finite value of v. The dotted line presents the value of v for which $J \approx T_K$; see the main text for details. In short timescales the two spins on the dots exhibit temporary ferromagnetic (FM) order, $S_{LR}(t) > 0$, while for longer times the system shows considerable spin-singlet correlations, i.e., antiferromagnetic (AFM) order with $S_{LR}(t) < 0$. The dotted-dashed (dashed) line presents the timescale associated with excitation to the triplet (doublet) state. The other parameters are the same as in Fig. 2 and $\Gamma/U = 0.1$.

other hand, the hopping v generates an antiferromagnetic exchange interaction between the dots, which can be estimated from the singlet-triplet splitting

$$J = \tfrac{1}{2}[\sqrt{16v^2 + U^2} - U]. \tag{7}$$

For small values of v it can be approximated by $J \approx 4v^2/U$. When each quantum dot is singly occupied, as assumed in our considerations, the physics is governed by the ratio of J/T_K, as can be seen in the inset of Fig. 2, which presents S_{LR} plotted as a function of J on a logarithmic scale, while the vertical dotted lines indicate the corresponding Kondo temperatures. Clearly, the correlations S_{LR} become considerable when $J \gtrsim T_K$. Then, at some even larger value of J, S_{LR} exhibits a local minimum, with $S_{LR} \to -3/4$ for $\Gamma \ll U$, to rise again with further increase of J. This further increase (decrease in magnitude) can be explained by considering all the double quantum dot states, the energies of which strongly depend on v, such that for larger v, other states become populated and S_{LR} increases; see Fig. 2.

IV. BUILDUP OF SPIN-SINGLET CORRELATIONS

Let us now analyze the dynamical behavior of the spin-spin expectation value $S_{LR}(t)$, following the quench in the hopping between the dots. In the initial state the system consists of two identical, disconnected copies of a quantum dot attached to a metallic electrode, each displaying the Kondo effect; see Fig. 1(b). Then, we turn on the hopping between the dots and study how the entanglement builds up in the system, quantified by the spin-spin expectation value. The time evolution of $S_{LR}(t)$ for different values of the hopping between the dots is shown in Fig. 3. First of all, one can note that a finite value of $S_{LR}(t)$ develops only when the hopping v is sufficiently large. The dotted line in the figure indicates the value of v for which the effective exchange interaction J becomes of the order of the Kondo temperature, $J \approx T_K$. For assumed parameters, this happens when $v/U \approx 0.033$. Indeed, when $J < T_K$, the Kondo singlet state formed on each dot dominates and the entangled singlet state between the two dots hardly develops. However, once $J \gtrsim T_K$ (see $v/U \gtrsim 0.033$ in the figure), $S_{LR}(t)$ exhibits considerable values in the long-time limit, indicating spin-singlet (antiferromagnetic) correlations between the quantum dots. This is schematically displayed in Fig. 1(c). For intermediate values of v, the timescale at which the AFM correlations develop can be related to the singlet-triplet excitation energy, $t \sim 1/J$; see the dotted-dashed line in Fig. 3.

Interestingly, in the intermediate time regime, $t \approx 1/U$, we observe a region when the spin-spin expectation value is positive. This implies ferromagnetic correlations between the quantum dots. In other words, before the singlet state forms in the double dot system, the calculated system dynamics reveals that in short timescales the effective exchange interaction changes sign, favoring triplet alignment of quantum dot spins. The boundary of this dynamical regime, indicated by the red dashed lines in Fig. 3, is established by the two timescales corresponding to the excitations to the doublet states. Note that the energy of doublet states splits with increasing v, encompassing the region where $S_{LR}(t) > 0$; see Fig. 3. If one considers realistic, semiconducting quantum dot parameters, e.g., $U \sim 1$ meV, this would result in a timescale on the order of $t \sim 0.01$–0.1 ns [1,21,24]. As far as the mechanism of

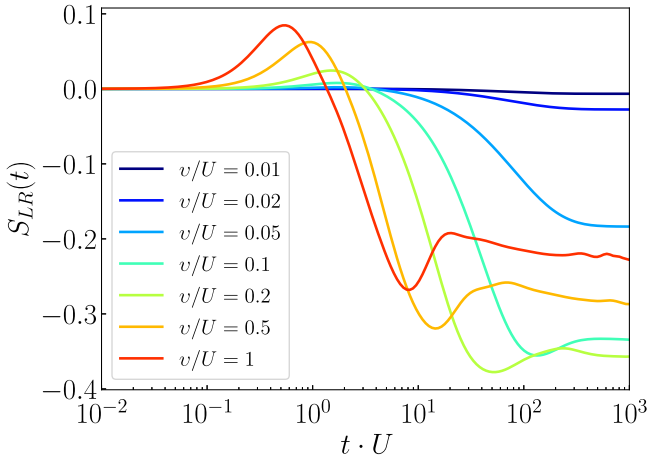

FIG. 4. The time-evolution of the expectation value $S_{LR}(t)$ calculated as a function of time for selected values of the hopping between the dots. The other parameters are the same as in Fig. 2 and $\Gamma/U = 0.1$.

this sign change is concerned, by comparing with the corresponding calculations for the two-impurity Kondo model (not shown), we can infer that it is due to the charge fluctuations to the doublet states (such sign change is absent in the Kondo model). As the system evolves toward the long-time limit, the timescale associated with the excitation to the triplet state (indicated by the blue dotted-dashed line) is surpassed and followed by the buildup of spin-singlet correlations.

The detailed behavior of $S_{LR}(t)$ is shown in Fig. 4, which presents the cross-sections of Fig. 3 for selected values of the hopping v, as indicated. One can now explicitly see that for small hoppings, such that $J \lesssim T_K$, $S_{LR}(t)$ is generally suppressed with a relatively low value in the long-time limit. However, when $J \gtrsim T_K$ (see the case of $v/U \gtrsim 0.05$ in Fig. 4), the absolute value in the long-time limit increases. Moreover, the timescale when considerable antiferromagnetic correlations develop decreases as v rises. As already mentioned, in the short timescale, temporary ferromagnetic correlations develop between the quantum dots. These are present when $v \gtrsim \Gamma$; see Fig. 4.

V. CONCLUSIONS

In this work we have studied the buildup of entanglement between the two quantum dots quantified by the spin-spin expectation value $S_{LR}(t)$. By employing the numerical renormalization group in time domain, we were able to include all correlations effects in a fully nonperturbative manner. In particular, we have examined the time evolution of $S_{LR}(t)$ upon turning on the hopping between the two quantum dots. It was found that, at small timescales, $t \sim 1/U$, $S_{LR}(t)$ becomes first positive, indicating ferromagnetic exchange interactions, while only for larger times does the effective exchange interaction become antiferromagnetic and an entangled spin singlet state is formed between the dots. The formation of such a singlet state is conditioned by the value of hopping between the quantum dots; it develops for such v that the effective exchange interaction $J \approx 4v^2/U$ becomes larger than the corresponding Kondo temperature for each quantum dot. Then, in the course of time evolution, the singlet state forms between the dots, winning over the two separate Kondo singlet states. The timescale when this happens is approximately given by $t \sim 1/J$, i.e., the inverse of the strength of the exchange interaction between the quantum dots.

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