

**Quenched dynamics of entangled states in correlated quantum dots**N. S. Maslova,<sup>1</sup> P. I. Arseyev,<sup>2</sup> and V. N. Mantsevich<sup>1,\*</sup><sup>1</sup>*Moscow State University, 119991 Moscow, Russia*<sup>2</sup>*P.N. Lebedev Physical Institute RAS, 119991 Moscow, Russia*

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The time evolution of an initially prepared entangled state in the system of coupled quantum dots has been analyzed by means of two different theoretical approaches: equations of motion for all orders localized electron correlation functions, considering interference effects, and kinetic equations for the pseudoparticle occupation numbers with constraint on the possible physical states. Results obtained by means of different approaches were carefully analyzed and compared to each other. Revealing a direct link between concurrence (degree of entanglement) and quantum dots pair correlation functions allowed us to follow the changes of entanglement during the time evolution of the coupled quantum dots system. It was demonstrated that the degree of entanglement can be controllably tuned during the time evolution of quantum dots system.

DOI: [10.1103/PhysRevA.96.042301](https://doi.org/10.1103/PhysRevA.96.042301)**I. INTRODUCTION**

One of the most interesting problems in present-day nanophysics is the controllable formation of entangled electronic states for use in quantum information processing and cryptography. Coupled quantum dots (QDs) systems recently seem to be promising candidates for quantum information applications, as single and two-electronic states can be well initialized, processed, and read out in such ultrasmall structures [1–8].

The properties of entangled states are usually analyzed in the stationary case. However, the time evolution of spin and charge configurations, initially prepared in coupled QDs, is also of great interest as nonstationary characteristics could reveal new information about the physical properties of nanoscale systems in addition to the stationary ones [9–16]. The kinetics of initially prepared charge and spin states in quantum dots systems is strongly governed by the high-order localized electrons correlation functions due to the presence of Coulomb interaction [17] and is also influenced by the interference effects between electrons traveling through different paths [18–20].

One of the challenges in the area of nonstationary electron transport through coupled QDs is to prepare interacting few-level systems with different initial states [21–24], from simple product states to complex entanglements. Various ideas for the entangling of spatially separated electrons were proposed, such as, by splitting Cooper pairs [25] or by spin manipulation in QDs [26,27]. In double correlated QDs the entangled state can appear as an eigenstate with a particular number of electrons [4,28] or by sending an electrical current through the nanoscale structure [29]. There are many possible applications of entangled states in nanoelectronics [30], including quantum information processing [31]. Most of the proposed schemes for quantum computation deal with the spin control due to the localized spins long decoherence times [32]. As it was recently shown, entangled states in correlated QDs could reveal long relaxation times due to the particular symmetry of the investigated system. Moreover, entangled states in

correlated quantum dots can be well controlled by applied bias voltage changing [33,34] or by external laser pulses [35,36].

Recently the potential of quantum information processing and quantum computation results in numerous proposals of specific material systems for creation and manipulation of entanglement in a solid state. The system based on the coupled quantum dots with Coulomb correlations has several appealing features: (1) single spin is a natural qubit, (2) presence of strong Coulomb interaction within the system creates entanglement even in the most easily experimentally obtained ground state, (3) entangled quantum states in the coupled QDs can be experimentally realized without such restrictions as for the two-impurity Kondo model. Moreover, the degree of entanglement can change during the relaxation of the initially prepared charge state in double QD coupled to a reservoir [8].

In the present paper we analyze the time evolution of an initially prepared entangled state in the correlated double QD due to the interaction with an electronic reservoir. Two different approaches were considered: the first one is based on the equations of motion for all orders localized electron correlation functions and the second one deals with the kinetic equations for pseudoparticle occupation numbers, considering constraint on the possible physical states. The results, obtained by means of these approaches were carefully analyzed and compared to each other. It was demonstrated that both approaches allow one to follow the changes of the system entanglement during time evolution due to the direct link between concurrence and quantum dots pair correlation functions. For different initial mixed states entanglement could reveal nonmonotonic behavior and even increase considerably during the relaxation processes in coupled quantum dots in the particular time interval. So, one can tune the degree of entanglement during the time evolution of correlated QDs. The proposed system is a good candidate for quantum information protocol (QIP) realization with the help of scanning tunneling microscopy and spectroscopy technique.

**II. THEORETICAL MODEL**

We consider a system of two coupled correlated QDs connected to an electronic reservoir. The Hamiltonian  $\hat{H}_D$ ,

\*vmantsev@gmail.com

describing interacting quantum dots reads

$$\hat{H}_D = \sum_{l=1,2,\sigma} \varepsilon_l c_{l\sigma}^+ c_{l\sigma} + \sum_{l=1,2} U_l n_{ll}^\sigma n_{ll}^{-\sigma} + \sum_{\sigma} T (c_{1\sigma}^+ c_{2\sigma} + c_{1\sigma} c_{2\sigma}^+), \quad (1)$$

where  $\varepsilon_l$  ( $l = 1, 2$ ) are the spin-degenerate single-electron energy levels and  $U_l$  is the on-site Coulomb repulsion for the quantum dots double occupation. The creation or annihilation of an electron with spin  $\sigma = \pm 1$  within the dot is denoted by operators  $c_{l\sigma}^+/c_{l\sigma}$  and  $n_{ll}^\sigma$  is the corresponding occupation number operator. The coupling between the dots is described by tunneling transfer amplitude  $T$  which is considered to be independent on momentum and spin.

The reservoir is modeled by the Hamiltonian

$$\hat{H}_{\text{res}} = \sum_{k\sigma} \varepsilon_k c_{k\sigma}^+ c_{k\sigma}, \quad (2)$$

where the operator  $c_{k\sigma}^+/c_{k\sigma}$  creates or annihilates an electron with spin  $\sigma$  and momentum  $k$  in the lead. Coupling between both dots and reservoir is described by the Hamiltonian

$$\hat{H}_{\text{tun}} = \sum_{k\sigma} t (c_{k\sigma}^+ c_{l\sigma} + c_{l\sigma}^+ c_{k\sigma}). \quad (3)$$

The tunneling amplitude  $t$  is independent of momentum and spin. When coupling between QDs exceeds the value of interaction with the reservoir, one can use the basis of exact eigenfunctions and eigenvalues of coupled QDs without interaction with the leads. In this case all energies of single- and multielectron states are well known.

Two single electron states are present in the system and can be described by the wave function

$$\Psi_i^\sigma = \mu_i |0\uparrow\rangle|00\rangle + \nu_i |00\rangle|0\uparrow\rangle, \quad (4)$$

where basis functions  $|0\uparrow\rangle|00\rangle$  and  $|00\rangle|0\uparrow\rangle$  describe the existence of a single electron with a given spin in each quantum dot. Single electron energies

$$\varepsilon_{a(s)} = \frac{\varepsilon_1 + \varepsilon_2}{2} \pm \sqrt{\frac{(\varepsilon_1 - \varepsilon_2)^2}{4} + T^2} \quad (5)$$

and coefficients  $\mu_i$  and  $\nu_i$  are determined by the eigenstates of the matrix

$$\begin{pmatrix} \varepsilon_1 & -T \\ -T & \varepsilon_2 \end{pmatrix}. \quad (6)$$

Six two-electron states exist in the system: two states with the same electrons spin in each dot are given by the wave functions  $T^+ = |\uparrow 0\rangle|\uparrow 0\rangle$  and  $T^- = |\downarrow 0\rangle|\downarrow 0\rangle$ . Such states can be formed only by electrons localized in the different dots. Four states with the opposite spins can be described by the wave function

$$\Psi_j^{\sigma-\sigma} = \alpha_j |\uparrow\downarrow\rangle|00\rangle + \beta_j |\downarrow 0\rangle|0\uparrow\rangle + \gamma_j |0\uparrow\rangle|\downarrow 0\rangle + \delta_j |00\rangle|\uparrow\downarrow\rangle, \quad (7)$$

where basis wave functions  $|\uparrow\downarrow\rangle|00\rangle$ ;  $|00\rangle|\uparrow\downarrow\rangle$  correspond to electrons localized in the same quantum dot (the first one or the second one and functions  $|\downarrow 0\rangle|0\uparrow\rangle$ ;  $|0\uparrow\rangle|\downarrow 0\rangle$  describe the situation when electrons are localized in different dots.

Two electron states energies and coefficients  $\alpha_j, \beta_j, \gamma_j, \delta_j$  are determined by the eigenvalues and eigenvectors of the matrix

$$\begin{pmatrix} 2\varepsilon_1 + U_1 & -T & -T & 0 \\ -T & \varepsilon_1 + \varepsilon_2 & 0 & -T \\ -T & 0 & \varepsilon_1 + \varepsilon_2 & 0 \\ 0 & -T & -T & 2\varepsilon_2 + U_2 \end{pmatrix}. \quad (8)$$

These are low-energy singlet  $S^0$  and triplet  $T^0$  states and excited singlet ( $S^{0*}$ ) and triplet states ( $T^{0*}$ ). The low-energy triplet state  $T^0$  with energy  $\varepsilon_1 + \varepsilon_2$  exists for any values of QDs energy levels  $\varepsilon_l$  and Coulomb interaction  $U_l$ . The corresponding coefficients in Eq. (7) are  $\alpha = \delta = 0$  and  $\beta = -\gamma = \frac{1}{\sqrt{2}}$ .

Two three-electron states with the wave function

$$\Psi_m^{\sigma\sigma-\sigma} = p_m |\uparrow\downarrow\rangle|\uparrow 0\rangle + q_m |\uparrow 0\rangle|\uparrow\downarrow\rangle, \quad m = \pm 1 \quad (9)$$

are present in the system. In this case, basis functions  $|\uparrow\downarrow\rangle|\uparrow 0\rangle$  and  $|\uparrow 0\rangle|\uparrow\downarrow\rangle$  describe the situation, when one of the dots is fully occupied by two electrons with opposite spins and only a single electron with a given spin is present in another dot. Coefficients  $p_m, q_m$  and energies are determined by the eigenvectors and eigenvalues of the matrix

$$\begin{pmatrix} 2\varepsilon_1 + \varepsilon_2 + U_1 & -T \\ -T & 2\varepsilon_2 + \varepsilon_1 + U_2 \end{pmatrix}. \quad (10)$$

Finally, a single four-electron state exists in the system with the wave function

$$\Psi_n = |\uparrow\downarrow\rangle|\uparrow\downarrow\rangle. \quad (11)$$

In this case both quantum dots are fully occupied.

### A. Equations of motion for localized electron correlation functions

Coupling to reservoir leads to the changing of the dots' occupation due to the tunneling processes. We now derive kinetic equations for bilinear combinations of the Heisenberg operators  $c_{l\sigma}^+/c_{l\sigma}$ , which allow to analyze the dynamics of localized electron occupation numbers and high-order correlation functions due to the coupling to the reservoir

$$\begin{aligned} c_{1\sigma}^+ c_{1\sigma} &= \hat{n}_{11}^\sigma(t); & c_{2\sigma}^+ c_{2\sigma} &= \hat{n}_{22}^\sigma(t); \\ c_{1\sigma}^+ c_{2\sigma} &= \hat{n}_{12}^\sigma(t); & c_{2\sigma}^+ c_{1\sigma} &= \hat{n}_{21}^\sigma(t). \end{aligned} \quad (12)$$

We consider the time evolution of the initially prepared state in the case of an "empty" reservoir in a wide band limit approximation and for deep energy levels ( $\frac{|\varepsilon_i - \varepsilon_F|}{\Gamma} \gg 1$ , where  $\Gamma = \pi \nu_0 t^2$ ,  $\nu_0$  is the unperturbed density of states in the reservoir) when the applied bias is equal to zero.

By means of Heisenberg equations of motion one can get a closed system of equations for localized electrons occupation numbers by exactly taking into account correlations of all orders [12, 13] (for weak tunneling coupling between QDs and the reservoir). Kinetic equations describe time evolution of the electron occupation numbers in the proposed system:

$$\begin{aligned} \frac{\partial}{\partial t} \hat{n}_{11}^\sigma &= -\Gamma (\hat{n}_{21}^\sigma + \hat{n}_{12}^\sigma) + iT (\hat{n}_{21}^\sigma - \hat{n}_{12}^\sigma) - 2\Gamma \hat{n}_{11}^\sigma, \\ \frac{\partial}{\partial t} \hat{n}_{22}^\sigma &= -\Gamma (\hat{n}_{21}^\sigma + \hat{n}_{12}^\sigma) - iT (\hat{n}_{21}^\sigma - \hat{n}_{12}^\sigma) - 2\Gamma \hat{n}_{22}^\sigma, \end{aligned}$$

$$\begin{aligned}
 \frac{\partial}{\partial t} \hat{n}_{21}^{\sigma} &= -\Gamma(\hat{n}_{11}^{\sigma} + \hat{n}_{22}^{\sigma}) + iT(\hat{n}_{11}^{\sigma} - \hat{n}_{22}^{\sigma}) - i(\xi - 2i\Gamma)\hat{n}_{21}^{\sigma} \\
 &\quad - iU_{11}n_{21}^{\sigma}n_{11}^{-\sigma} + iU_{22}n_{21}^{\sigma}n_{22}^{-\sigma}, \\
 \frac{\partial}{\partial t} \hat{n}_{12}^{\sigma} &= -\Gamma(\hat{n}_{11}^{\sigma} + \hat{n}_{22}^{\sigma}) + iT(\hat{n}_{11}^{\sigma} - \hat{n}_{22}^{\sigma}) + i(\xi + 2i\Gamma)\hat{n}_{12}^{\sigma} \\
 &\quad + iU_{11}n_{12}^{\sigma}n_{11}^{-\sigma} - iU_{22}\hat{n}_{12}^{\sigma}n_{22}^{-\sigma}, \quad (13)
 \end{aligned}$$

where  $\xi = \varepsilon_1 - \varepsilon_2$  is the detuning between energy levels in the dots. The first term in each right-hand part of Eqs. (13) [ $\Gamma(\hat{n}_{21}^{\sigma} + \hat{n}_{12}^{\sigma})$  or  $\Gamma(\hat{n}_{11}^{\sigma} + \hat{n}_{22}^{\sigma})$ ] appears due to the interference effects caused by the charge relaxation to the reservoir through different possible channels, similar to the Fano effect. These terms are absent if only one quantum dot is coupled to the reservoir. The system of Eqs. (13) contains pair correlation operators  $\hat{K}_{lr'l'r'}^{\sigma\sigma'}$ , which also determine relaxation and, consequently, should be calculated. If one is interested in the relaxation dynamics of the two-electron initial state, only pair correlation functions should be retained as the situation of the “empty” reservoir is considered.

Let us introduce the correlations operators  $\hat{K}_{lr'l'r'}^{\sigma\sigma'}$ , averaged values  $K_{lr'l'r'}^{\sigma\sigma'} = \langle c_{l\sigma}^{\dagger} c_{r\sigma} c_{l'\sigma'}^{\dagger} c_{r'\sigma'} \rangle$ , which are elements of the  $\hat{\mathbf{K}}$   $4 \times 4$  matrix. The system of equations for the pair correlation functions can be written in the compact matrix form (the symbol  $[\ ]$  means commutation and the symbol  $\{ \}$  means anticommutation)

$$i \frac{\partial}{\partial t} \hat{\mathbf{K}} = [\hat{\mathbf{K}}, \hat{H}] + \{\hat{\mathbf{K}}, \hat{\Gamma}\}, \quad (14)$$

where matrix  $\hat{H}$  has the following form:

$$\hat{H} = \begin{pmatrix} 0 & T + i\Gamma & T - i\Gamma & 0 \\ T - i\Gamma & \xi + U_1 & 0 & T - i\Gamma \\ T + i\Gamma & 0 & -\xi + U_2 & T + i\Gamma \\ 0 & T + i\Gamma & T - i\Gamma & 0 \end{pmatrix} \quad (15)$$

and  $\hat{\Gamma}$  is the relaxation diagonal  $4 \times 4$  matrix with nonzero elements  $\Gamma_{nn} = -2i\Gamma$ .

The system of equations (13) and (14) for the two-electron pure state  $|\Psi_j^{\sigma\sigma'}\rangle$  time evolution can be solved with the following initial conditions:  $n_{11}^{\sigma}(0) = \alpha^2 + \beta^2$ ;  $n_{12}^{\sigma}(0) = n_{21}^{\sigma}(0) = \alpha\gamma + \beta\delta$ ;  $n_{22}^{\sigma}(0) = \delta^2 + \gamma^2$ ;  $K_{1111}^{\sigma-\sigma} = \alpha^2$ ;  $K_{2222}^{\sigma-\sigma} = \delta^2$ ;  $K_{1122}^{\sigma-\sigma} = \beta^2$ ;  $K_{2211}^{\sigma-\sigma} = \gamma^2$ ;  $K_{1221}^{\sigma-\sigma} = K_{2112}^{\sigma-\sigma} = \beta\gamma$ ;  $K_{2121}^{\sigma-\sigma} = K_{1212}^{\sigma-\sigma} = \alpha\delta$ ;  $K_{1211}^{\sigma-\sigma} = K_{2111}^{\sigma-\sigma} = \gamma\alpha$ ;  $K_{1112}^{\sigma-\sigma} = K_{1121}^{\sigma-\sigma} = \alpha\beta$ ;  $K_{1222}^{\sigma-\sigma} = K_{2122}^{\sigma-\sigma} = \beta\delta$ ;  $K_{2221}^{\sigma-\sigma} = K_{2212}^{\sigma-\sigma} = \gamma\delta$ , where coefficients  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $\delta$  are given by the eigenvectors of matrix (8).

For an initial mixed two-electron state with density matrix  $\rho(0) = \sum_{j,\sigma,\sigma'} N_j^{\sigma\sigma'}(0) |\Psi_j^{\sigma\sigma'}\rangle \langle \Psi_j^{\sigma\sigma'}|$ , where  $N_j^{\sigma\sigma'}(0)$  ( $j = S^0, T^0, S^{0*}, T^{0*}, T^{\pm}$ ) is the occupation number of  $j$  two-electron state at  $t = 0$ , the initial conditions for second-order correlation functions and for first-order correlators are

$$K_{lr'l'r'}^{\sigma-\sigma}(0) = Sp[\hat{\rho}(0)\hat{K}_{lr'l'r'}^{\sigma-\sigma}] \quad (16)$$

and

$$n_{lr}^{\sigma}(0) = Sp[\hat{\rho}(0)\hat{n}_{lr}^{\sigma}]. \quad (17)$$

We will consider the time evolution of singlet  $S^0$  and triplet  $T^0$  initial states because excited  $S^{0*}$  and  $T^{0*}$  states are separated by a Coulomb gap. One can also exclude states  $T^{\pm}$  at low temperature by introducing weak exchange interaction with exchange constant  $J_z > 0$ :

$$\hat{H}_{\text{ex}} = J_z(n_{11}^{\sigma} - n_{11}^{-\sigma})(n_{22}^{\sigma} - n_{22}^{-\sigma}). \quad (18)$$

Consequently, the initial two-electron density matrix can be written as

$$\rho(0) = N_{S^0}(0)|S^0\rangle\langle S^0| + N_{T^0}(0)|T^0\rangle\langle T^0|. \quad (19)$$

For the singlet initial state  $S^0$  coefficients  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $\delta$  are determined as an eigenvector of matrix (8) corresponding to its minimal eigenvalue,  $N_{S^0}(0) = 1$  and  $N_{T^0}(0) = 0$ . For the triplet initial state  $T^0$  coefficients  $\alpha = \delta = 0$  and  $\beta = -\gamma = \frac{1}{\sqrt{2}}$ ,  $N_{S^0}(0) = 0$  and  $N_{T^0}(0) = 1$ .

## B. Entangled states in correlated quantum dots

Electron states in the correlated quantum dots can be entangled. An entangled state is characterized by the nonzero value of concurrence  $C$  [37]. The concurrence for pure state  $|\Psi_j^{\sigma\sigma'}\rangle$  is determined as  $C = |\langle \Psi_j^{\sigma\sigma'} | \tilde{\Psi}_j^{\sigma\sigma'} \rangle|$ , where  $|\tilde{\Psi}_j^{\sigma\sigma'}\rangle$  is the “spin flipped” state  $|\Psi_j^{\sigma'\sigma}\rangle$ . For mixed state concurrence  $C = \max\{0, \lambda_1 - \sum_i \lambda_i\}$ , where  $\{\lambda_i\}$  are square roots of matrix  $\tilde{\rho}\rho$  ( $\tilde{\rho}$  is the “spin flipped” matrix  $\rho$ ) eigenvalues arranged in the decreasing order. For the initial two-electron entangled pure state  $|\Psi_j^{\sigma\sigma'}\rangle$  with opposite spins [8]

$$C = |\alpha^2 + \delta^2 + 2\beta\gamma|. \quad (20)$$

During the time evolution of the initial state system entanglement changes. To follow these changes concurrence could be expressed through the time-dependent correlation functions. We will demonstrate that for an arbitrary mixed state of two correlated quantum dots the concurrence  $C$  can be determined through the mean value of a particular combination of pair correlation functions  $\hat{K}_{lr'l'r'}^{\sigma-\sigma}$ :

$$C = \langle \hat{K}_{1111}^{\sigma-\sigma} + \hat{K}_{1221}^{\sigma-\sigma} + \hat{K}_{2112}^{\sigma-\sigma} + \hat{K}_{2222}^{\sigma-\sigma} \rangle. \quad (21)$$

Let us introduce operator  $\hat{K}'$ , which can be written as a combination of pair correlation functions operators

$$\hat{K}' = \hat{K}_{1111}^{\sigma-\sigma} + \hat{K}_{1221}^{\sigma-\sigma} + \hat{K}_{2112}^{\sigma-\sigma} + \hat{K}_{2222}^{\sigma-\sigma}. \quad (22)$$

Acting by the operator  $\hat{K}'$  on the wave function  $|\Psi_j^{\sigma\sigma'}\rangle$  one obtains the “spin flipped” wave function  $|\tilde{\Psi}_j^{\sigma\sigma'}\rangle$ :

$$\hat{K}' |\Psi_j^{\sigma\sigma'}\rangle = |\tilde{\Psi}_j^{\sigma\sigma'}\rangle. \quad (23)$$

For any wave function  $|\Psi_j^{\sigma\sigma'}\rangle$ :

$$|\langle \Psi_j^{\sigma\sigma'} | \hat{K}' | \Psi_j^{\sigma\sigma'} \rangle| = |\langle \Psi_j^{\sigma\sigma'} | \tilde{\Psi}_j^{\sigma\sigma'} \rangle| = C. \quad (24)$$

If  $\{|\Psi_j^{\sigma\sigma'}\rangle\}$  are the two-electron eigenfunctions of the Hamiltonian  $\hat{H}$ , the two particle density matrix can be written as  $\rho = \sum_j |\Psi_j^{\sigma\sigma'}\rangle \langle \Psi_j^{\sigma\sigma'}| N_j^{\sigma\sigma'}$ . For simplicity we will further omit spin indexes in  $|\Psi_j^{\sigma\sigma'}\rangle$ . The following relations take place:  $|\langle \Psi_j | \hat{K}' | \tilde{\Psi}_i \rangle| = \delta_{ij}$  and  $|\langle \Psi_{i'} | \hat{K}'^2 | \Psi_i \rangle| = \delta_{i'i} = \sum_j \langle \Psi_{i'} | \hat{K}' | \tilde{\Psi}_j \rangle \langle \tilde{\Psi}_j | \hat{K}' | \Psi_i \rangle$ .

Let us prove that

$$\langle \Psi_j | \tilde{\rho} \rho | \Psi_i \rangle = \langle \Psi_j | \hat{K}' \rho \hat{K}' \rho | \Psi_i \rangle. \quad (25)$$

Really

$$\begin{aligned} \langle \Psi_j | (\hat{K}' \rho)^2 | \Psi_i \rangle &= \sum_{i_1} \langle \Psi_j | \hat{K}' | \Psi_{i_1} \rangle \langle \Psi_{i_1} | \hat{K}' | \Psi_i \rangle N_i N_{i_1} \\ &= \sum_{i_1} \langle \Psi_j | \tilde{\Psi}_{i_1} \rangle \langle \Psi_{i_1} | \tilde{\Psi}_{i_1} \rangle N_i N_{i_1} \end{aligned} \quad (26)$$

and

$$\begin{aligned} \langle \Psi_j | \tilde{\rho} \rho | \Psi_i \rangle &= \sum_{i_1} N_i N_{i_1} \langle \Psi_j | \tilde{\Psi}_{i_1} \rangle \langle \tilde{\Psi}_{i_1} | \Psi_i \rangle \\ &= \delta_{ij} \sum_{i_1} N_i N_{i_1} |\langle \Psi_j | \tilde{\Psi}_{i_1} \rangle|^2. \end{aligned} \quad (27)$$

Comparing expressions (26) and (27), one can find that statement (25) is valid and matrices  $\|\tilde{\rho}\rho\|_{ji}$  and  $\|(\hat{K}'\rho)^2\|_{ji'}$  have the same eigenvalues. If  $\tilde{\lambda}_p$  are the eigenvalues of matrix  $\|\tilde{\rho}\rho\|_{ji}$  and  $\lambda_i$  are the eigenvalues of matrix  $\|\hat{K}'\rho\|_{ji'}$ , then  $\lambda_i^2 = \tilde{\lambda}_p$ . So,

$$\langle \hat{K}' \rangle = \text{Tr}(\hat{K}' \rho) = \sum_{ij} \langle j | \hat{K}' | i \rangle \langle i | j \rangle \lambda_i = \sum_i \lambda_i \langle i | i \rangle. \quad (28)$$

The relative sign of  $\lambda_i = \pm \sqrt{\tilde{\lambda}_p}$  is determined by the sign of  $\langle i | i \rangle = \pm 1$  for singlet and triplet states.

Moreover, for the eigenstates

$$|\langle S^0 | \tilde{S}^0 \rangle| = |\langle T^0 | \tilde{T}^0 \rangle| = |\langle T^\mp | \tilde{T}^\pm \rangle| = 1. \quad (29)$$

Thus  $\langle \hat{K}' \rangle$  can be expressed through  $\tilde{\lambda}_p$ , arranged in decreasing order:  $\langle \hat{K}' \rangle = (\sqrt{\tilde{\lambda}_1} - \sum_{i>1} \sqrt{\tilde{\lambda}_i})$ . So, the common definition of concurrence  $C$  (see Ref. [37]) can be rewritten as

$$C = \max\{0, \langle \hat{K}' \rangle\}. \quad (30)$$

The behavior of the time-dependent quantum-dot system concurrence calculated by means of Eqs. (13), (21), and (30) for different initial conditions is demonstrated in Figs. 1 to 3. Figures 1 and 2 demonstrate an important fact, that

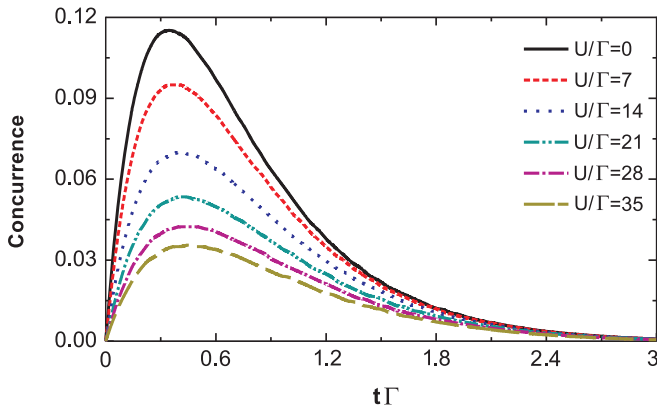


FIG. 1. Concurrence time evolution for different values of Coulomb interaction  $U_1/\Gamma = U_2/\Gamma = U/\Gamma$ .  $\varepsilon_1/\Gamma = \varepsilon_1/\Gamma = 7$ ,  $T/\Gamma = 2$ , and  $\Gamma = 1$ . Initial conditions are as follows:  $N_S(0) = 0.5$ ,  $N_T(0) = 0.5$ .

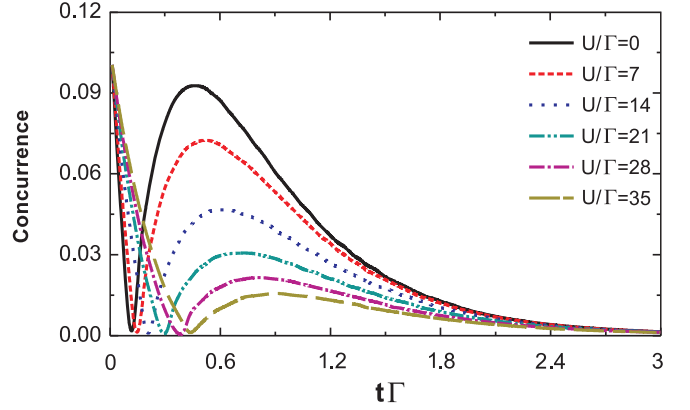


FIG. 2. Concurrence time evolution for different values of Coulomb interaction  $U_1/\Gamma = U_2/\Gamma = U/\Gamma$ .  $\varepsilon_1/\Gamma = \varepsilon_1/\Gamma = 7$ ,  $T/\Gamma = 2$ , and  $\Gamma = 1$ . Initial conditions are as follows:  $N_S(0) = 0.55$ ,  $N_T(0) = 0.45$ .

concurrence (the degree of entanglement) can increase during the relaxation processes in the system of coupled QDs, caused by the presence of on-site Coulomb correlations and interaction with the reservoir. The results depicted in Fig. 3 reveal the possibility of system switching between entangled and unentangled (concurrence is equal to zero) states during the relaxation process.

### C. Kinetic equations in pseudoparticle formalism

Another method of quantum-dot system dynamics analysis is based on the pseudoparticles' formalism [38]. Each pseudoparticle corresponds to a particular eigenstate of the system. Transitions between the states with different numbers of electrons caused by coupling to the reservoir can be analyzed in terms of pseudoparticle operators with constraint on the possible physical states (the number of pseudoparticles). Consequently, the electron operator  $c_{i\sigma}^+$  ( $l = 1, 2$ ) can be

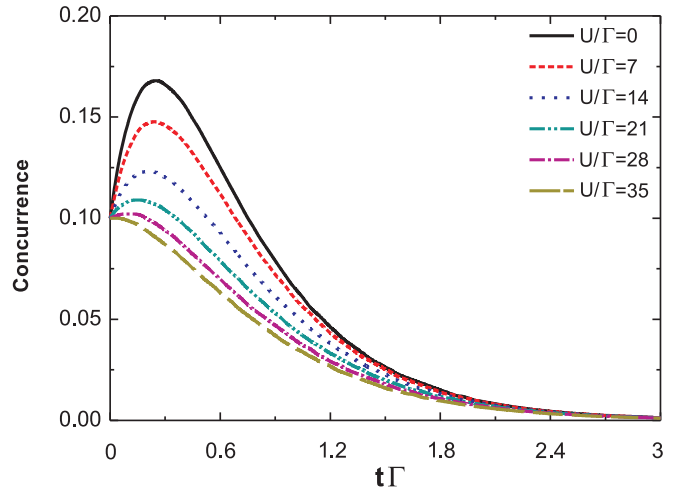


FIG. 3. Concurrence time evolution for different values of Coulomb interaction  $U_1/\Gamma = U_2/\Gamma = U/\Gamma$ .  $\varepsilon_1/\Gamma = \varepsilon_1/\Gamma = 7$ ,  $T/\Gamma = 2$ , and  $\Gamma = 1$ . Initial conditions are as follows:  $N_S(0) = 0.45$ ,  $N_T(0) = 0.55$ .

written in terms of pseudoparticle operators [8]:

$$\begin{aligned}
 c_{l\sigma}^+ &= \sum_i X_i^{\sigma l} f_{i\sigma}^+ b + \sum_{j,i} Y_{ji}^{\sigma-\sigma l} d_j^{+\sigma-\sigma} f_{i-\sigma} \\
 &+ \sum_i Y_i^{\sigma\sigma l} d^{+\sigma\sigma} f_{i\sigma} + \sum_{m,j} Z_{mj}^{\sigma\sigma-\sigma l} \psi_{m-\sigma}^+ d_j^{\sigma-\sigma} \\
 &+ \sum_m Z_m^{\sigma-\sigma-\sigma l} \psi_{m\sigma}^+ d^{-\sigma-\sigma} + \sum_m W_m^{\sigma-\sigma-\sigma l} \varphi^+ \psi_{m\sigma},
 \end{aligned} \quad (31)$$

with constraint on the possible physical states

$$\hat{n}_b + \sum_{i\sigma} \hat{n}_{fi\sigma} + \sum_{j\sigma\sigma'} \hat{n}_{dj}^{\sigma\sigma'} + \sum_{m\sigma} \hat{n}_{\psi m\sigma} + \hat{n}_\varphi = 1, \quad (32)$$

where  $f_\sigma^+$  ( $f_\sigma$ ) and  $\psi_\sigma^+$  ( $\psi_\sigma$ ) are pseudofermion creation (annihilation) operators for the electronic states with one and three electrons correspondingly.  $b^+$  ( $b$ ),  $d_\sigma^+$  ( $d_\sigma$ ), and  $\varphi^+$  ( $\varphi$ ) are slave boson operators, which correspond to the states without any electrons, with two electrons or four electrons. Operators  $\psi_{m-\sigma}^+$  describe a system configuration with two spin up electrons  $\sigma$  and one spin down electron  $-\sigma$  in the symmetric and asymmetric states.

Further we will consider only single- and low-energy double-occupied states because the excited double-occupied states, three-, and four- particle states are separated by the Coulomb gap. Consequently, all the terms containing  $\varphi^+$  and  $\psi_{m-\sigma}^+$  in expression (32) are omitted. Matrix elements  $X_i^{\sigma l}$ ,  $Y_{ji}^{\sigma-\sigma l}$ , and  $Y_{ji}^{\sigma\sigma l}$  can be defined as

$$\begin{aligned}
 X_i^{\sigma l} &= \langle \Psi_i^\sigma | c_{l\sigma}^+ | 0 \rangle, \\
 Y_{ji}^{\sigma-\sigma l} &= \langle \Psi_j^{\sigma-\sigma} | c_{l\sigma}^+ | \Psi_i^{\sigma-\sigma} \rangle, \\
 Y_{ji}^{\sigma\sigma l} &= \langle \Psi_j^{\sigma\sigma} | c_{l\sigma}^+ | \Psi_i^{\sigma\sigma} \rangle.
 \end{aligned} \quad (33)$$

So, taking into account constraint on the possible physical states the following nonstationary system of equations can be obtained for the pseudoparticle occupation numbers  $N_i^\sigma$ ,  $N_j^{\sigma-\sigma}$ ,  $N_j^{\sigma\sigma}$ , and  $N_b$  by means of Heisenberg equations:

$$\begin{aligned}
 \frac{\partial N_j^{\sigma-\sigma}}{\partial t} &= -2\Gamma \sum_{i\sigma} |Y_{ji}^{\sigma-\sigma}|^2 N_j^{\sigma-\sigma}, \\
 \frac{\partial N_i^\sigma}{\partial t} &= 2\Gamma \sum_j |Y_{ji}^{\sigma-\sigma}|^2 N_j^{\sigma-\sigma} \\
 &- 2\Gamma |X_i^\sigma|^2 N_i^\sigma + 2\Gamma \sum_j |Y_{ji}^{\sigma\sigma}|^2 N_j^{\sigma\sigma}, \\
 \frac{\partial N_b}{\partial t} &= 2\Gamma \sum_{i\sigma} |X_i^\sigma|^2 N_i^\sigma, \\
 \frac{\partial N_j^{\sigma\sigma}}{\partial t} &= -2\Gamma \sum_i |Y_{ji}^{\sigma\sigma}|^2 N_j^{\sigma\sigma},
 \end{aligned} \quad (34)$$

where matrix elements can be easily expressed through the elements of matrices (6), (8), and (10) eigenvectors

$$\begin{aligned}
 |X_i^\sigma|^2 &= |v_i + \mu_i|^2, \\
 |Y_{ji}^{\sigma-\sigma}|^2 &= |\alpha_j \mu_i + \beta_j v_i + \gamma_j \mu_i + \delta_j v_i|^2, \\
 |Y_{ji}^{\sigma\sigma}|^2 &= |v_i + \mu_i|^2.
 \end{aligned} \quad (35)$$

Depending on the tunneling barrier width and height typical tunneling rate  $\Gamma$  can vary from  $10 \mu\text{eV}$  [39] to  $1 \div 5 \text{meV}$  [40]. These equations conserve the total number of pseudoparticles

$$N_b + \sum_{i\sigma} N_i^\sigma + \sum_{j\sigma\sigma'} N_j^{\sigma\sigma'} = \text{const}. \quad (36)$$

So, Eqs. (34) provide the fulfillment of constraint

$$N_b + \sum_{i\sigma} N_i^\sigma + \sum_{j\sigma\sigma'} N_j^{\sigma\sigma'} = 1 \quad (37)$$

during time evolution, if it occurs at the initial time moment.

The system of Eqs. (34) can be solved analytically with initial conditions  $N_j^{\sigma\sigma'}(0) = N_j$ ,  $N_a^\sigma(0) = 0$ ,  $N_s^\sigma(0) = 0$ , and  $N_b(0) = 0$  ( $\sum_j N_j = 1$ ). For initial generally mixed singlet-triplet state (19)

$$\begin{aligned}
 N_j^{\sigma-\sigma}(t) &= N_j e^{-2\lambda_j t}, \\
 N_a^\sigma(t) &= \sum_j \left[ \frac{\lambda_{ja}}{2\lambda_j - \lambda_a} (e^{-\lambda_a t} - e^{-2\lambda_j t}) \right] N_j, \\
 N_s^\sigma(t) &= \sum_j \left[ \frac{\lambda_{js}}{2\lambda_j - \lambda_s} (e^{-\lambda_s t} - e^{-2\lambda_j t}) \right] N_j, \\
 N_b(t) &= 1 - N_{dj}^{\sigma-\sigma}(t) - \sum_\sigma N_a^\sigma(t) - \sum_\sigma N_s^\sigma(t),
 \end{aligned} \quad (38)$$

where

$$\begin{aligned}
 \lambda_{a(s)} &= 2\Gamma |\mu_{a(s)} + \nu_{a(s)}|^2, \\
 \lambda_{ja(s)} &= 2\Gamma |\alpha_j \mu_{a(s)} + \beta_j \nu_{a(s)} + \delta_j \nu_{a(s)} + \gamma_j \mu_{a(s)}|^2, \\
 \lambda_j &= \sum_{i=a,s} \lambda_{ji}.
 \end{aligned} \quad (39)$$

Electron occupation numbers  $N_{el}$  can be determined through the pseudoparticle occupation numbers considering spin degrees of freedom by the following expression:

$$N_{el}(t) = \sum_j N_j \left[ 2e^{-2\lambda_j t} + 2 \sum_{i=a,s} \frac{\lambda_{ij}}{2\lambda_j - \lambda_i} (e^{-\lambda_i t} - e^{2\lambda_j t}) \right]. \quad (40)$$

According to the concurrence definition through the eigenvalues of matrix  $\tilde{\rho}$  for initial state (19):

$$C(t) = \max(0, |N_{S^0}(t) - N_{T^0}(t)|), \quad (41)$$

where

$$\begin{aligned}
 N_{S^0}(t) &= N_{S^0}(0) e^{-2\lambda_{S^0} t}, \\
 N_{T^0}(t) &= N_{T^0}(0) e^{-2\lambda_{T^0} t}, \\
 \lambda_{S^0} &= |\alpha + \beta|^2 (\lambda_s + \lambda_a), \\
 \lambda_{T^0} &= \frac{1}{2} (\lambda_s + \lambda_a).
 \end{aligned} \quad (42)$$

The concurrence time evolution for different initial conditions and values of Coulomb correlations is shown in Figs. 1 to 3. The results obtained by both approaches exactly coincide for the same system parameters.

If at the initial time moment concurrence is not equal to zero [ $C(0) \neq 0$ ] there can exist a time moment  $t = t_0$ , when

concurrence turns to zero [ $C(t_0) = 0$ ] (see Fig. 2)

$$t_0 = \frac{1}{2(\lambda_{S^0} - \lambda_{T^0})} \ln\left(\frac{N_{S^0}}{N_{T^0}}\right). \quad (43)$$

A further system time evolution leads to the concurrence increasing reaching its maximum value

$$C(t_{\max}) = N_{T^0} e^{\frac{-\lambda_{T^0}}{\lambda_{S^0} - \lambda_{T^0}} \ln\left(\frac{N_{S^0} \lambda_{S^0}}{N_{T^0} \lambda_{T^0}}\right)} - N_{S^0} e^{\frac{-\lambda_{S^0}}{\lambda_{S^0} - \lambda_{T^0}} \ln\left(\frac{N_{S^0} \lambda_{S^0}}{N_{T^0} \lambda_{T^0}}\right)} \quad (44)$$

at time moment

$$t_{\max} = t_0 + \frac{1}{2(\lambda_{S^0} - \lambda_{T^0})} \ln\left(\frac{\lambda_{S^0}}{\lambda_{T^0}}\right). \quad (45)$$

So, concurrence could reveal nonmonotonic behavior for mixed two-electronic initial state with opposite spins. We would like to mention that despite the fact that both theoretical approaches give the same result for the considered system of two coupled quantum dots with Coulomb correlations there is some difference between them. The theoretical approach based on the equations of motion for localized electrons occupation numbers (see Sec. II A) provides the possibility for the analysis of concurrence and spin correlations time-dependent behavior in the complicated systems of many correlated coupled quantum dots taking into account all orders localized electron correlation functions and considering interference effects. A closed system of equations can be obtained for an arbitrary number of quantum dots in the situation of weak coupling to reservoir, but these equations will have a rather cumbersome form and can be hardly solved analytically. The theoretical approach based on the pseudoparticles formalism (see Sec. II C) is more straightforward and provides the possibility to analyze analytically time evolution of the degree of entanglement in the system of quantum dots with a small number of available electronic states in the case of weak interaction between correlated quantum dots and reservoir.

To conclude, both methods can be applied for the localized charge and spin kinetics analysis in coupled quantum dots with Coulomb correlations, but for the complicated systems method based on the equations of motion for localized electrons occupation numbers is more preferable. However, systems with a small number of available electronic states could be better analyzed by means of the pseudoparticles formalism as it provides the possibility to obtain explicit expressions for the time evolution of system characteristics.

### III. CONCLUSION

The time evolution of an initially prepared entangled state in the system of correlated coupled quantum dots has been analyzed by means of two different approaches. The first one is based on the equations of motion for all orders localized electron correlation functions taking into account interference effects. The second approach deals with the kinetic equations for pseudoparticle occupation numbers considering constraint on the possible physical states. Both approaches allow us to follow the changes of the entanglement during time evolution of the two coupled quantum dots system due to the concurrence direct link with quantum dots pair correlation functions. For different initial mixed states the concurrence (degree of entanglement) could reveal nonmonotonic behavior and even considerably increase during the time evolution of quantum dots system. The obtained results reveal the possibility of system switching between entangled and unentangled (concurrence is equal to zero) states during the relaxation process. This fact provides the method of controllable tuning of the degree of entanglement for the electronic quantum dots systems based on the analysis of its nonstationary characteristics.

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