Quantum dots with two electrons: Singlet-triplet transitions

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The magnetic character of the ground-state of two electrons on a double quantum dot, connected in series to left and right single-channel leads, is considered. By solving exactly for the spectrum of the two interacting electrons, it is found that the coupling to the continuum of propagating states on the leads, in conjunction with the electron-electron interactions, may result in a delocalization of the bound state of the two electrons. This, in turn, reduces significantly the range of the Coulomb interaction parameters over which singlet-triplet transitions can be realized. It is also found that the coupling to the leads favors the singlet ground-state.

I. INTRODUCTION

The spin state of the many electron ground-state is determined by the interplay between the kinetic and the electrostatic interactions. According to Hund’s law the ground-state of electrons in a partially filled shell of an atom has the maximal possible spin, in order to minimize the electrostatic repulsion. On the other hand, Anderson’s super-exchange antiferromagnetic interaction, which favors zero total spin, arises from the reduction in the ground-state energy brought about by the hopping of electrons between adjacent ions. Another example is realized in gases made of homonuclear diatomic molecules, in which the distance between the nuclei determines the magnetic properties of the gas; in other words, the ground-state of the molecule can be either a triplet or a singlet.

The possibility to study these rules as function of controlled parameters, and in particular to observe deviations from them, has been opened up in recent years with the intensive experimental and theoretical investigations of small confined systems. Devices based on quantum dots formed in GaAs heterostructures allow to probe the electronic states in situations where there are only a few electrons in the system, as function of, e.g., the number of electrons in the sample, the spacing of the single-electron energy levels, the internal structure of the dots (e. g. the distance and the coupling between internal parts) or the coupling of the system to external leads. Thus, modulating the single-particle spectrum by a magnetic field applied perpendicularly to the plane of the electron gas, has produced a structure of the conductance peaks that has been interpreted in terms of singlet-triplet transitions of the last two electrons in the dot (see also Ref. 7). This has been observed on vertical quantum dots and also in lateral ones. The ground-state spin of chaotic quantum dots has been studied by tracing the conductance peak spacing as function of a weak parallel magnetic field (which couples primarily to the spins).

A numerical investigation of such a configuration analyzed the influence of the exchange interaction on the peak structure. In a similar fashion, the singlet-triplet transitions in such dots have been attributed to avoided-crossings in the many-electron states, and the relation between those and kinks in the conductance pattern has been explored.

When the device consists of two or more coupled dots, another controllable parameter comes into the play: the interdot coupling. Experimentally, it has been found that this coupling shows up in the conductance peak positions. Theoretically, the effect of the interdot distance on the states of a few electrons confined in a parabolic potential has been analyzed using mean-field methods, the Kohn-Sham equation and numerical diagonalization. Different spin states of double-quantum-dots systems have been also studied using the numerical renormalization group method.

Here we present an exact analytical solution for the eigenvalues and eigenfunctions of two electrons on a double-dot system, which is coupled to two single-channel leads. The two electrons interact while they are on the dots, and we include in the calculation the direct Coulomb and the exchange interactions between the dots, and the on-site Hubbard interactions on the dots. We obtain the condition for the singlet-triplet transitions of the ground-state energies, and in particular examine the role of the delocalization effect of the interactions: It has been found in our previous works as well as in other studies that the interplay between the hybridization of the localized single-particle states on the dot with the propagating states on the leads, and the electron-electron interactions, may promote one of the electrons to the continuum. If the system had only one doubly bound state, then the above ‘promotion’ results in a ‘delocalization’ of...
the ground state. Once delocalized, the singlet and the triplet states become degenerate.

Our earlier work included only the case with on-site Coulomb repulsion. In that case, the bound ground state is a singlet. As the coupling of the dots with the leads was increased, the parameter range where this bound state exists was decreased, and the delocalization was accompanied by a transition from the singlet to a degenerate singlet-triplet ‘metallic’ state. It is usually assumed that the presence of the coupling to the leads, it is well known that the ground state changes from a singlet to a triplet upon increasing the exchange interaction. Here we investigate what happens to that singlet-triplet transition in the presence of the coupling to the leads. After discussing our model Hamiltonian in Sec. II, we present the exact two-electron solution in Sec. III. Section IV then discusses the singlet-triplet transition, mainly for a special choice of the parameters where it is easiest to explore the solution analytically. Our results are then summarized in Sec. V.

II. HAMILTONIAN

The Hamiltonian of the model reads

\[ H = H_{\text{sp}} + H_{\text{c}}. \]  

(1)

The single-particle tight binding Hamiltonian is

\[ H_{\text{sp}} = \sum_{i} \varepsilon_{i} c_{i\sigma}^{\dagger} c_{i\sigma} - \sum_{ij} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma}, \]  

(2)

where \( c_{i\sigma} \) creates an electron with spin \( \sigma \) on site \( i \). The site energies \( \varepsilon_{i} \) are different from zero only on the ‘quantum dots’, which can be viewed as ‘impurities’ on the lattice. In what follows, we specifically consider two neighboring quantum dots, which will be denoted by \( \ell \) and \( r \), and study the symmetric case where \( \varepsilon_{\ell,r} = \varepsilon_{o} \). The hopping matrix elements \( t_{ij} \) are divided into three kinds: the hopping among the ‘dots’ is denoted by \( t_{D} \) (\( \equiv t_{\ell r} \)), the hopping between a dot and a neighboring site on the ‘lead’ is denoted by \( t_{0} \) (e.g. \( t_{\ell i} \) for \( i \neq r \)), and the hopping between neighboring ‘lead’ sites is denoted by \( t = 1 \), setting the units of energy.

The Coulomb interactions are assumed to exist only among electrons which sit on the dots. Generally, this interaction has the form

\[ H_{\text{c}} = \sum_{ijmn} \Gamma_{ij}^{mn} \sum_{\sigma\sigma'} c_{i\sigma}^{\dagger} c_{j\sigma'}^{\dagger} c_{m\sigma'} c_{n\sigma}, \]  

(3)

where

\[ \Gamma_{ij}^{mn} = \int d\mathbf{r} \int d\mathbf{r}' v(\mathbf{r} - \mathbf{r}') \varphi_{i}^{*}(\mathbf{r}) \varphi_{j}^{*}(\mathbf{r}') \varphi_{m}(\mathbf{r}') \varphi_{n}(\mathbf{r}), \]  

(4)

while \( \varphi_{i}(\mathbf{r}) \) is the Wannier wave function of an electron localized at site \( i \). It is usually assumed that the dominant terms will be those in which the indices are equal pair-wise. Here we follow the parametrization used in magnetic coupling studies, in which the case \( i = j = m = n \) is treated separately. Neglecting the coefficient with \( i = j \neq m = n \), which (when negative) leads to the superconductivity vertex, we are left with three possible parameters:

\[ 2\Gamma_{ii}^{ij} = U, \quad 2\Gamma_{ij}^{ij} = V, \quad 2\Gamma_{ij}^{ij} = K, \quad i \neq j, \]  

(5)

where \( U, V \) and \( K \) are the (intra-dot) Hubbard, (inter-dot) direct, and (inter-dot) exchange interactions, respectively. Then

\[ H_{\text{int}} = U \sum_{i} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + \frac{1}{4} (2V - K) \sum_{i \neq j} \hat{n}_{i\uparrow} \hat{n}_{j\downarrow} - K \sum_i \mathbf{S}_i \cdot \mathbf{S}_j, \]  

(6)

where

\[ \hat{n}_{i\sigma} = c_{i\sigma}^{\dagger} c_{i\sigma}, \quad \hat{n}_{i} = \sum_{\sigma} \hat{n}_{i\sigma}, \]  

(7)

and \( \sigma \) is the vector of Pauli’s spin matrices. It is important to note that with the above approximation for the Coulomb interaction vertex, the interaction Hamiltonian becomes spin-dependent.

A similar Hamiltonian has been used to study the effects of the spin states on the conducting properties of confined mesoscopic quantum dots. In these studies, \( i \) represents some orbital state on the dot. In Ref. it has been argued that when the Thouless conductance of the confined (mesoscopic) system is large, the Coulomb vertices can be parametrized in terms of two coupling constants, independent of the orbital indices. In terms of our parameters, these are the charging energy, \( E_{c} = (2V - K)/4 \), and the exchange energy, \( J = K \), while \( U = V + K \). It is straightforward to apply our results to that case.

In our special case of the double dot, the energies \( \Gamma \) in Eq. (3) differ from zero only for \( i, j = \ell, r \), and thus

\[ H_{\text{int}} = U (\hat{n}_{\ell\uparrow} \hat{n}_{r\downarrow} + \hat{n}_{\ell\downarrow} \hat{n}_{r\uparrow}) + (V - \frac{K}{2}) \hat{n}_{\ell\uparrow} \hat{n}_{r\downarrow} - 2KS_{\ell} \cdot S_{r}. \]  

(8)
III. THE TWO-ELECTRON ‘MOLECULE’

We will confine ourselves to a double-dot system (‘molecule’), containing two electrons. The system is modeled by two identical single-level impurities, each having an on-site single-particle energy level \( \epsilon_t = \epsilon_r = \epsilon_0 \). The two impurities are coupled to one another by the interdot matrix element \( t_D \). When the molecule is isolated \((t_0 = 0)\), there are three degenerate triplet states, of energy \( 2\epsilon_0 + V - K \), and three singlet states. One of the latter has the energy \( 2\epsilon_0 + U \), while the other two energies are

\[
E = 2\epsilon_0 + \frac{U + V + K}{2} \pm \sqrt{4t_D^2 + \left(\frac{U - V - K}{2}\right)^2}, \tag{9}
\]

Examination of the interaction matrix elements, Eqs. (1) and (3), shows that \( U > V > 0 \) and \( U > K \). Moreover, the exchange interaction \( K \) involves the square of the overlap matrix element of the two impurities, and hence is of the order of \( t_D^2 \). Therefore one concludes that the lowest singlet energy is given by the minus sign in (9). The triplet states energy is below the lowest singlet one provided that

\[
2t_D^2 < K(U - V + K). \tag{10}
\]

One notes that when the direct and exchange Coulomb vertices \( V \) and \( K \) are disregarded, the energy of the singlet state is lowered by the interdot kinetic energy, producing \( t_D^2 \sim 4t_D^2/U \). The triplet state becomes the ground-state once the exchange energy \( K \) wins over this kinetic energy. Finally, with the choice of parameters in which \( U = V + K \) (see above), the condition becomes \( t_D^2 < K^2 \) (see also Ref. 3). Both \( t_D \) and \( K \) decay exponentially with the interdot distance. However, Eq. (10) indicates that \( K \) is roughly of order \( t_D^2 \), and thus decays faster. Therefore, one might expect a transition from the triplet to the singlet ground state as this distance increases.

The main purpose of the present paper is to study how the criterion for the singlet-triplet transitions, Eq. (10), is modified when the double quantum dot is connected to external leads, on which it is assumed that there are no electron-electron interactions. In other words, we study the changes in these transitions due to coupling the dot to a continuum of propagating states. A similar problem was discussed in Ref. 24 in the framework of the mean-field approximation, within a single-lead geometry. Here we first derive exactly the ground-state energy of two electrons, and then compare the singlet and the triplet ground-state energies.

For further simplification, we describe the external leads by single channel one dimensional chains, with nearest neighbor hopping \( t = 1 \). In our previous work, we have shown that the spectrum and the wave functions of two interacting electrons can be obtained in terms of the energy spectrum and the wave functions of the single-particle Hamiltonian. For our model, cf. Eq. (3), the latter can be divided into even and odd solutions, denoted by “e” and “o”. Of particular interest here will be the regions in the parameter plane \((\epsilon_0, \gamma)\), where \( \gamma = t_D^2 \), where the spectrum has bound states, see Fig. 1. For \( \epsilon_0 < \gamma - 2 + t_D \), there appears one (even) bound state below the band of propagating states, of energy \( \epsilon_B^{(e)} < -2 \). For even smaller values of \( \epsilon_0 \), such that \( \epsilon_0 < \gamma - 2 - t_D \) there appears a second (odd) bound state below the band, of energy \( \epsilon_B^{(o)} \), with

\[
\epsilon_B^{(o)} = \frac{2 - \gamma}{1 - \gamma} \left(\frac{\epsilon_0}{2}\right) + \frac{\gamma}{1 - \gamma} \sqrt{\left(\frac{\epsilon_0}{2}\right)^2 + \gamma - 1}, \tag{11}
\]

Similarly, for \( \epsilon_0 > 2 - \gamma - t_D \) there appears the first bound state above the band, while for \( \epsilon_0 > 2 - \gamma + t_D \) there are two bound states above the band. Clearly, a necessary condition to have the two electrons bound in a triplet state is the existence of two distinct single-electron bound states, so that each electron occupies a different ‘state’. For simplicity, we shall confine ourselves to the case in which both occur below the continuum, i.e., to the lowest region II in Fig. 1.

![Fig. 1. The single-particle bound energies. The roman numbers denote the number of bound states. Here \( t_D = 0.4 \).](image-url)

It has been shown in Refs. 15 and 16 that when interactions between the two electrons are allowed, they may lead to the delocalization of one (or both) of the electrons from the doubly bound states. Thus, for example, when \( V = 0 \), in part of region I in Fig. 1 one finds that the doubly bound ground state is replaced by a ground state in which one of the electrons is shifted to the band. In these parameter regions there are no two-electron bound states, and the system may be called ‘metallic’. Indeed, such interaction-induced delocalization effects have been
recently observed in quantum dots containing two potential minima. This effect has also been found for two interacting electrons moving in a one-dimensional periodic structure and for spinless fermions on strongly disordered chains. In contrast to region I, in region II there potentially exist two doubly bound states, i.e., the singlet and the triplet, and our goal is to study which of these states is the ground state in the presence of this interaction-induced delocalization.

To solve for the pair of the two interacting electrons, we proceed as follows. Let us denote the eigenstates and the eigenvalues of the single-particle Hamiltonian by $\phi_a(n)$ and $\epsilon_a$, respectively, where $n$ is the site index. Consider first the two-electron states with spin $S_z = \pm 1$,

$$|\Psi^\sigma\rangle = \sqrt{\frac{1}{2}} \sum_{ab} X_{ab}^\sigma \phi_a^{\dag}(n) \phi_b^{\dag}(n') \phi_a(n') \phi_b(n) \sum_{\sigma' \sigma} c_{\sigma a}^{\dag} c_{\sigma' b}^{\dag} |0\rangle$$

$$= \sqrt{\frac{1}{2}} \sum_{ab} X_{ab}^\sigma \phi_a^{\dag}(n) \phi_b^{\dag}(n') \phi_a(n') \phi_b(n) \sum_{\sigma' \sigma} c_{\sigma a}^{\dag} c_{\sigma' b}^{\dag} |0\rangle$$

$$= \sqrt{\frac{1}{2}} \sum_{nn'} X^{\sigma}_{nn'} \phi_a^{\dag}(n) \phi_b^{\dag}(n') \phi_a(n') \phi_b(n) \sum_{\sigma' \sigma} c_{\sigma a}^{\dag} c_{\sigma' b}^{\dag} |0\rangle, \quad \sigma = \pm 1, \quad (12)$$

where the amplitudes $X_{nn'}^\sigma = -X_{n'n}^\sigma$ are antisymmetric in the coordinates (the origin is halfway between the dots), and $\sum_{nn'} |X_{nn'}^\sigma|^2 = 1$. The Schrödinger equation for the two electrons then yields

$$X_{nn'}^\sigma = X_{\ell l}(V - K) \times \sum_{ab} \frac{\phi_a(r) \phi_b(r) - \phi_a(r') \phi_b(r)}{E - \epsilon_a - \epsilon_b} \phi_a(n) \phi_b(n'). \quad (13)$$

Hence, the two-particle energies, $E$, are given by the solutions of the equation

$$\frac{1}{V - K} = \frac{1}{2} \sum_{ab} \frac{\phi_a(r) \phi_b(r) - \phi_a(r') \phi_b(r)}{E - \epsilon_a - \epsilon_b} \phi_a(n) \phi_b(n'). \quad (14)$$

Making use of the symmetry properties of $\phi_a$, i.e., $\phi_a^{\sigma_2}(r) = \phi_a^{\sigma_2}(r)$, and $\phi_a^{\sigma_2}(r) = -\phi_a^{\sigma_2}(r)$, we see that the only contributions to the sum come from the cases in which $a$ is even, and $b$ is odd, and vice versa. For the sake of brevity, in the following expressions we replace $|\phi_a^{\sigma}(r)| = |\phi_a^{\sigma}(r)| (i = e, o)$ by $|\phi_a^{\sigma}|$. Consequently, we may write the result in the form

$$\frac{1}{V - K} = G_{ee}(E), \quad \quad (15)$$

where $G_{ee}$ is a noninteracting two-electron Green’s function,

$$G_{ee}(E) = 4 \sum_{\sigma} \left| \phi_{\nu}^{\sigma}(r) \phi_{\nu'}^{\sigma}(r) \right| \left( E - \epsilon_{\nu}^{\sigma} - \epsilon_{\nu'}^{\sigma} \right) + 2 \sum_{i,j=1,2} \sum_{k} \left| \phi_{ik}^{\sigma} \right|^2 \left( E - \epsilon_{ik}^{\sigma} - \epsilon_k \right)$$

$$+ \sum_{kk'} \left| \phi_{kk'}^{\sigma} \right|^2 \left( E - \epsilon_k - \epsilon_{kk'} \right). \quad (16)$$

(In writing down this equation, we have assumed the existence of two single-particle bound states, as mentioned above.) Here the subscript $B$ denotes a single-particle bound state, and $k$ refers to a band state, with $\epsilon_k = -2 \cos k$. In the continuum of the band energies, there is no need to distinguish between $\epsilon_k^{\sigma}$ and $\epsilon_k^{\sigma'}$. Also, the sum over all states $k$ is divided into the sum over the even propagating states, and the sum over the odd ones. We give in the Appendix the explicit expressions for the eigenstates required for the calculation of $G$.

Equation (15) is an explicit equation for the exact two-electrons eigenenergies $E$. We will postpone the discussion of these solutions, and consider now the two-electron states with $S_z = 0$,

$$|\Psi^0\rangle = \sum_{ab} X_{ab}^0 \phi_a^{\dag}(n) \phi_b^{\dag}(n') \phi_a(n') \phi_b(n) \sum_{\sigma' \sigma} c_{\sigma a}^{\dag} c_{\sigma' b}^{\dag} |0\rangle$$

$$= \sum_{ab} X_{ab}^0 \phi_a^{\dag}(n) \phi_b^{\dag}(n') \phi_a(n') \phi_b(n) \sum_{\sigma' \sigma} c_{\sigma a}^{\dag} c_{\sigma' b}^{\dag} |0\rangle$$

$$= \sum_{nn'} X_{nn'}^0 \phi_a^{\dag}(n) \phi_b^{\dag}(n') \phi_a(n') \phi_b(n) \sum_{\sigma' \sigma} c_{\sigma a}^{\dag} c_{\sigma' b}^{\dag} |0\rangle, \quad (17)$$

with $\sum_{nn'} |X_{nn'}^0|^2 = 1$. For the triplet state, the amplitudes $X_{nn'}^0$ are antisymmetric in the site indices. Then (inserting Eq. (17) in the Schrödinger equation) the energies are again given by Eqs. (15) and (16). For the singlet state, $X_{nn'}^{0} = X_{0}^{0}$, and the Schrödinger equation yields

$$X_{nn'}^{0} = \sum_{ab} \left( U \sum_{i=\ell,r} \phi_a(i) \phi_b(i) + (V + K) X_{\ell r}^{0} \right) \phi_a^{\dag}(n) \phi_b^{\dag}(n') \frac{\phi_a(r) \phi_b(r) - \phi_a(r') \phi_b(r)}{E - \epsilon_a - \epsilon_b} \phi_a^{\dag}(n) \phi_b^{\dag}(n'). \quad (18)$$

We use this equation for $n, n' = \ell, \ell, n, n' = r, r$, and $n, n' = \ell, r, r$, and find two families of singlet solutions: (i) $X_{\ell r}^{0} = -X_{r\ell}^{0}$, $X_0^{0} = 0$, for which

$$1 \over U = G_{eo}(E); \quad \quad (19)$$

(ii) $X_{\ell r}^{0} = X_{r\ell}^{0}$, with

$$1 - (U + V + K)(G_{eo}(E) + G_{oo}(E)) + 4(U + V + K)G_{eo}(E)G_{oo}(E) = 0. \quad (20)$$

[Note that the last equation includes twice the number of solutions as Eqs. (15) and (16).] Here, $G_{ee, oo}$ are noninteracting, two-particle Green’s functions, which consist of the even and odd (with respect to interchanging the dots) solutions of the single-particle spectrum, respectively, with

$$G_{ee, oo}(E) = 2 \sum_{k} \left| \phi_{\nu}^{\sigma}(r) \right|^2 \left( E - \epsilon_{\nu}^{\sigma} - \epsilon_k \right)$$

$$+ \sum_{kk'} \left| \phi_{kk'}^{\sigma} \right|^2 \left( E - \epsilon_k - \epsilon_{kk'} \right). \quad (21)$$
We next determine the ground-state energy, starting with the triplet states, whose energies are given by Eq. (13). We assume that we are in the lower region II of Fig. 1, where there are two bound states below the band. (Note that in the regions marked 0 and I in Fig. 1, where there is at most one single-particle bound state, the triplet ‘bound’ state will always lie in the continuum.) The function $G_{ee}(E)$, Eq. (13), has the following behavior. As $E$ approaches $-\infty$, it goes to zero from below. At $E = \epsilon_B^e + \epsilon_B^o$, it diverges to $-\infty$, jumps to $+\infty$ as $E$ crosses that value, and then decreases, as $E$ approaches the bottom of the two-electron continuum states located at $-2 + \epsilon_B$. As discussed in our earlier work, in the thermodynamic limit of infinite ‘leads’ $G_{ee}$ has a finite value at this band threshold, due to the $k$-dependence of $\phi_{k}^{e(o)}$ at the impurities. The triplet ground-state energy is where $G_{oo}$ crosses $(V-K)^{-1}$. Hence, there will be a two-electron bound state only when $(V-K)^{-1} < G_{oo}(-2 + \epsilon_B^o)$. It follows that there are values of the direct and exchange Coulomb couplings such that the ground triplet state is not bound, but lies in the continuum.

We now turn to the singlet states, again assuming the existence of both $\epsilon_B^e$ and $\epsilon_B^o$. Consider first the solutions given by Eq. (13). Since $U>V$ (and $V-K$), the lowest solution of this equation lies above the lowest solution of the triplet state, (which is given by the same function $G_{oo}$). Hence, we need not consider anymore the states given by (13). To explore the other family of singlet solutions, it is convenient to re-write Eq. (20) in the form

$$\frac{1}{4} \left( \frac{1}{G_{ee}(E)} + \frac{1}{G_{oo}(E)} \right) = \frac{U + V + K}{2}$$

$$\pm \sqrt{\frac{1}{16} \left( \frac{1}{G_{ee}(E)} - \frac{1}{G_{oo}(E)} \right)^2 + \left( \frac{U - V - K}{2} \right)^2}. \quad (22)$$

Since the behavior of $G_{ee}$ and $G_{oo}$ as function of $E$ is similar to that of $G_{oo}$ described above, it follows from Eq. (22) that the lowest singlet state energy obeys that equation with the minus sign.

IV. SINGLET-TRIPLET TRANSITIONS

In order to decide when the lowest bound state of the two electrons is a singlet or a triplet, we need to (i) determine for which values of the Coulomb parameters Eqs. (13) and (22) have bound solutions; and (ii) to compare these two solutions, when they exist. Consider as an example the case in which there is only the on-site Hubbard interaction, that is, $V=K=0$. Then the triplet bound state has the energy $E_T = \epsilon_B^e + \epsilon_B^o$, cf. Eq. (13). The singlet energy, $E_S$, in that case is given by the lowest solution of

$$\frac{1}{U} = G_{ee}(E) + G_{oo}(E). \quad (23)$$

Similarly to the behavior of $G_{oo}(E)$, the right-hand-side of this equation starts at very small negative values when $E$ tends to $-\infty$. It then diverges to $-\infty$ as $E$ approaches $2\epsilon_B^o$ jumps to $+\infty$ as $E$ crosses that value, diverges again to $-\infty$ as $E \rightarrow 2\epsilon_B^o$, then jumps to $+\infty$, and finally decreases towards a finite value as $E$ approaches the bottom of the two-electron continuum. It follows that Eq. (23) has always a bound energy solution. Moreover, if $G_{ee} + G_{oo}$ is negative at $E = E_T \leq \epsilon_B^e + \epsilon_B^o$, then that solution $E_S$ lies below $E_T$, i.e., the ground-state is a singlet. This is indeed the case, as is shown in the Appendix [Eq. (A13)]. This is in accordance with the general rule, which states that in order for the ground-state to be a triplet, the exchange Coulomb energy has to overcome the kinetic energy.

For the sake of clarity of the presentation, we will carry the rest of the analysis to lowest order in the coupling to the leads, $\gamma$. In that case, it is possible to derive simple expressions for the two-particle Green’s functions, see Eqs. (A13). Using those equations, we find that the singlet energies are given by

$$E - 2\epsilon_0 + \gamma(e^{-\alpha^o} + e^{-\alpha^e}) = \frac{U + V + K}{2}$$

$$- \sqrt{4t_D^2 + \left( \frac{U - V - K}{2} \right)^2} + 4t_D\gamma(e^{-\alpha^o} - e^{-\alpha^e}), \quad (24)$$

and the triplet energies are given by

$$E - 2\epsilon_0 + \gamma(e^{-\alpha^o} + e^{-\alpha^e}) = V - K, \quad (25)$$

where $\alpha^{e,o}$ is related to the corresponding $E$ (with indices $S$ or $T$) via

$$E - \epsilon_B^{e,o} = -2\cosh e^{\alpha^{e,o}}. \quad (26)$$

Let us examine the case in which in the absence of the coupling to the leads, the singlet and the triplet ground-state energies are equal, i.e., $4t_D^2 = 2K(U+V+K)$, see Eq. (17). Then

$$E_S - E_T = \gamma(e^{-\alpha_T^e} + e^{-\alpha_T^o} - e^{-\alpha_S^e} - e^{-\alpha_S^o})$$

$$- \frac{4t_D}{U - V + 3K}\gamma(e^{\alpha_S^e} - e^{\alpha_S^o}). \quad (27)$$

The last term on the right-hand-side of this equation is negative. This follows from Eq. (23) and the fact that $\epsilon_B^e < \epsilon_B^o$. As for the first term, we use again Eq. (23), to write it in the form $(E_T - E_S)\gamma/(e^{2\alpha_S^o} - 1) + (e^{2\alpha_S^e} - 1)^{-1})$. Hence, $E_S < E_T$ and the singlet is preferred.

The above discussion shows that the coupling to the continuum of propagating states enhances the tendency of the two electrons to form a singlet state, in the situation where in the absence of that coupling, the singlet and the triplet states are degenerate. In order to investigate whether this tendency persists for other choices of parameters (and at the same time to keep the calculations tractable) we will now confine ourselves to the
choice $U=V+K$. In this case, again to leading order in $\gamma$, the equation for the singlet energies (24) reads
\[
f_S(E) = V + K, \\
f_S(E) = E - 2\epsilon_0 + 2t_D + 2\gamma e^{-\alpha^*}. \tag{28}
\]
Similarly, Eq. (25) for the triplet energies can be written as
\[
f_T(E) = V - K, \\
f_T(E) = E - 2\epsilon_0 + \gamma(e^{-\alpha^*} + e^{-\alpha^*}). \tag{29}
\]

Let us first determine for which parameters these equations yield bound, two-electron energies. To this end, we consider Eqs. (24) and (29) at the bottom of the two-electron continuum, $E=-2+e_B^\alpha$. The first of these equations will have a bound state for $f_S(-2+e_B^\alpha) > V + K$; the second will have such a solution when $f_T(-2+e_B^\alpha) > V - K$. These conditions are plotted in Fig. 2 as the thick lines there. A bound triplet state exists in regions I+III, below the heavy line of positive slope. A singlet bound state exists in I+II, below the heavy line of negative slope. In region IV, there are no bound states; both the triplet and the singlet states are in the continuum, and their energy is about the same. Crossing the line between regions II and IV (or III and IV) thus corresponds to the delocalization transition discussed above, from a singlet (triplet) bound ground state to a degenerate ‘metallic’ state. This transition is the most striking effect of the coupling to the leads, and we expect it to appear irrespective of the quantitative approximation used in Fig. 2.

In region I one has to compare the singlet energy with the triplet one. These two become degenerate along the diamond curve, whose equation is derived from (26), (28), and (29)
\[
V - K = -\frac{e_B^\alpha}{2}(B-1) + \frac{e_B^\alpha}{2}(B+1) - 2\epsilon_0 - (1-\gamma)AB, \\
A = \frac{2}{\gamma}(K - t_D), \\
B = \sqrt{1+4/[A(A+e_B^\alpha-e_B^\beta)]}. \tag{30}
\]

This line is almost vertical, with $E_S < E_T$ to its left, and $E_S > E_T$ to its right. The conclusion is that, as long as there exists a bound state of the two electrons, then this line moves slightly to the right as $\gamma$ is increased from zero (when the line was at $K=t_D$). However, the coupling to the continuum states delocalizes the electrons, making the two states degenerate over a large part of the parameter plane \{V \& K\}, i.e. region IV in Fig. 2. With all other parameters fixed, one might expect that increasing the distance between the dots causes a decrease in $t_D$, in V and in K, thus causing a shift towards to lower left side of Fig. 2, towards a bound singlet ground state.

We have derived analytical expressions for the spectrum of two interacting electrons on a simplified model for a double quantum-dot. When the dot is decoupled from the external leads, it is straightforward to obtain this spectrum, and discuss the criterion for its ground-state to be a singlet or a triplet. The question we have addressed is how this criterion is modified when the single-particle states which are localized on the dot are coupled to the continuum of extended states on the leads. A typical example of our results is shown in Fig. 2 as long as the electron-electron interactions do not delocalize the ground-state, then the location for the singlet-triplet transition shifts continuously with the coupling to the leads, $\gamma$. In that case, one can still say that the singlet state is the ground one provided that the kinetic energy dominates over the exchange energy. In a way, the coupling to the leads enhances the kinetic energy, and therefore it slightly favors the singlet ground state. However, the coupling to the leads has a much more drastic effect: over a significant part of the parameter space (which consists of the Coulomb couplings, the single-particle energies on the dots, and the coupling to the leads), e.g. region IV in Fig. 2, the interplay between the coupling to the leads and the electron-electron interactions delocalizes one or both electrons. Then, the singlet and the triplet states are degenerate. In such a situation, the bound state (which may be either a singlet or a triplet) disappears and the ground state becomes a degenerate singlet-triplet ‘metallic’ state. We believe that this delocalization effect should be taken into consideration in the analyzes of experimental data related to this question.
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APPENDIX A: THE TWO-ELECTRON GREEN’S FUNCTIONS

We first list the eigenfunctions required for the calculation of the noninteracting, two-particle Green’s functions. These are needed only on the double quantum dot, that is, on the sites $r$ and $\ell$. Writing the even and odd bound-state energies, Eqs. (11), in the form

$$\epsilon^e_o = -2\cosh\alpha^e_o,$$

(A1)

the wave functions on the dot sites is

$$|\phi^e_o|^2 = \frac{1}{2} \left( 1 + \frac{\gamma}{e^{2\epsilon^e_o} - 1} \right)^{-1}.$$  

(A2)

The band states have been calculated assuming periodic boundary conditions for a system of $N$ sites (these include the leads). Then

$$|\phi^0_k|^2 = \frac{2}{N} \sum_{k} \frac{\gamma \sin^2 k}{\gamma^2 \sin^2 k + (\epsilon_k - \epsilon^0_o + \gamma \cos k)^2},$$

(A3)

with $\epsilon_k = -2 \cos k$, for both the even and odd states. In calculating the sums over $k$ in the two-particle Green’s functions, we shall use the continuum limit, dividing the $k$-integrations on the even (odd) functions by 2.

We next derive $G_{ee}$, $G_{oo}$, and $G_{eo}$. It is convenient first to calculate the function

$$Q^{e,o}(\omega) = \sum_{k} \frac{|\phi^e_o|^2}{\omega - \epsilon_k}, \quad \omega < -2.$$  

(A4)

Writing

$$\omega = -2\cosh\alpha,$$

(A5)

and using Eq. (A3), we find

$$Q^{e,o} = -\gamma \frac{e^{\alpha + \kappa^e_o}}{e^{2\alpha + \kappa^e_o} - 1 + \gamma} \times \frac{\sin^2 k}{\sin^2 k + (\epsilon_k - \epsilon^0_o + \gamma \cos k)^2},$$

(A6)

where we have also used

$$\sum_{k} |\phi^e_o|^2 = 1 - 2|\phi^e_o|^2.$$  

(A7)

Exploiting this result, we now introduce the function $F$,

$$F^{e,o}(\omega) = |\phi^e_o|^2 + (\omega - \epsilon^e_o)\frac{1}{2}Q^{e,o}(\omega)$$

$$= \frac{1}{2} \left( 1 + \gamma/(e^{\alpha + \kappa^e_o} - 1) \right).$$  

(A8)

Note that

$$F^{e}(\epsilon^e_o) = |\phi^e_o|^2, \quad F^{e}(\epsilon^e_o) = |\phi^e_o|^2,$$

$$F^{o}(\epsilon^e_o) = F^{o}(\epsilon^e_o) = \frac{1}{2} \left( 1 + \gamma/(e^{\kappa^e_o} - 1) \right).$$  

(A9)

It is now straightforward to show, using Eqs. (21), (16), and (A8) that

$$G_{ee}(E) = \frac{2}{E - 2\epsilon^e_o} \left( \frac{F^{e}(E - \epsilon^e_o)^2}{E - 2\epsilon^e_o} + \sum_{k,k'} |\phi^e_k|^2 |\phi^e_{k'}|^2 (\epsilon^e_k - \epsilon^e_{k'})/2 \right)/(E - \epsilon^e_k - \epsilon^e_{k'})(E - \epsilon^e_k - \epsilon^e_{k'}),$$

(A10)

with an analogous result for $G_{oo}$, with $e$ replaced by $o$, and

$$G_{eo}(E) = \frac{4}{E - \epsilon^e_o} \left( \frac{F^{e}(E - \epsilon^e_o)F^{o}(E - \epsilon^o_o)}{E - \epsilon^e_o} + \sum_{k,k'} |\phi^e_k|^2 |\phi^o_{k'}|^2 (\epsilon^o_{k'} - \epsilon^e_{k'})/2 \right)/(E - \epsilon^o_{k'} - \epsilon^e_{k'})(E - \epsilon^e_k - \epsilon^o_{k'}).$$

(A11)

For energies $E$ below the bottom of the two-electron continuum, i.e., $E < -2 + \epsilon^e_o$, the double sum on $k$ and $k'$ in (A10) is negative. Using Eq. (A9) for the first terms in the equations for $G_{ee}$ and $G_{oo}$, it follows that

$$G_{ee}(E^e + \epsilon^e_o) + G_{oo}(E^o + \epsilon^o_o) < 0.$$  

(A12)

This result is used to show that when the only Coulomb coupling is the Hubbard $U$, the singlet is always the ground state.

Up to this point, the results were given for general $\gamma$. To lowest order in the coupling to the leads, $\gamma$, we may discard the double sums in (A10) and (A11). Then, using (A8), we find

$$\frac{1}{G_{eo}(E)} \sim E - 2\epsilon_o + \gamma(e^{-\alpha^e} + e^{-\alpha^o}),$$

$$\frac{1}{2G_{ee}(E)} \sim E - 2\epsilon_o + 2t_D + 2\gamma e^{-\alpha^e},$$

$$\frac{1}{2G_{oo}(E)} \sim E - 2\epsilon_o - 2t_D + 2\gamma e^{-\alpha^o},$$

(A13)

where we have used Eq. (24).

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