Exchange energy tuned by asymmetry in artificial molecules

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Laterally coupled asymmetric quantum dots occupied by two electrons are studied using the exact diagonalization approach. It is shown that the asymmetry enhances the exchange energy, i.e., the triplet-singlet energy difference for finite magnetic fields. At high magnetic field, electrons enter the deepest dot more easily if they have parallel spins.

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The spins of an electron pair in coupled quantum dots[1] were proposed[2] to serve as a basis of a quantum gate for a solid-state quantum computer. The coupling between the spins can be realized in the orbital[3] degrees of freedom exploiting the spatial symmetry of the singlet and triplet wave functions. The strength of the coupling is quantified by the exchange energy defined[3] as the energy difference of the lowest triplet and the lowest singlet states. The value of the exchange energy depends on interdot tunnel coupling. In vertically stacked dots[1,4–8] the coupling is fixed by the process of the growth which determines the composition and thickness of the interdot layer. The interdot barrier can be more conveniently controlled in laterally coupled dots[1,3,9–15] by potentials applied to gate electrodes. But there are possible difficulties with the control of the exchange energy resulting from the fact that electrostatic confinement potential in gated quantum dots is usually weak.[16] In such quantum dots with weak confinement the dominant electron Coulomb repulsion leads to the formation of Wigner molecules[1] for which the ground state becomes degenerate with respect to the spin. This is due to the vanishing overlap between the single-electron wave functions.[8,17] Therefore for a pair of large quantum dots the exchange energy may remain negligibly small even if the interdot barrier is totally removed. Consequently, the exchange energy risks to be too small to be of practical use for spin control.

The exchange energy can be controlled by an external magnetic field.[3,4,7,12–14] But its value is maximal in the absence of the field.[3,4,7,12–14] The application of a magnetic field diminishes the interdot tunnel coupling due to an increased localization[18] of the electrons in each of the dots. A high magnetic field results in a large effective interdot barrier leading to a vanishing exchange energy[3] due to the separation of the single-electron wave functions. This is similar to Wigner crystallization appearing in a single large quasi-one-dimensional quantum dot.[8,17] In a single circular quantum dot the magnetic field induces singlet-triplet oscillations[1] which when the Zeeman spin effect is neglected, continue to infinity. In coupled quantum dots the magnetic field usually induces at least one singlet-triplet transition,[3,4,7,12–14] reminiscent of the singlet-triplet oscillations in a single dot,[13] before the exchange energy is eventually reduced to zero.

In the present paper we show that at zero magnetic field the exchange energy can be strongly enhanced by an asymmetry introduced in the confinement potential of laterally coupled dots. At high magnetic fields a small asymmetry[19] turns out to be irrelevant for the exchange energy which vanishes (when the Zeeman spin effect is neglected) due to a complete localization of electrons in different dots.

Asymmetry of the confinement potential in existing devices containing laterally coupled dots[9,10] is the rule. In fact symmetric coupling appears only for voltages along the diagonal[20] connecting the triple points at the honeycomb stability diagram.[10] Although vertical coupling of asymmetric dots[4] has been considered in the context of the exchange energy, most[3,12,13] of the theoretical work on laterally coupled dots dealt with pairs of identical dots. Only recently was the effect of the asymmetry on the few-electron charging considered.[9] The influence of asymmetry on the exchange energy in laterally coupled dots was addressed in Ref. 3 where the effect of the electric field was studied in the Heitler-London approximation assuming single occupancy of the dots with a neglected dependence of the single-electron wave functions on the magnetic field. This neglect leads[3] to a magnetic-field-independent shift of the exchange energy, which is in disagreement with the exact diagonalization results presented below.

We consider a two-dimensional double quantum dot in a perpendicular magnetic field $\boldsymbol{B}=(0,0,B)$ and neglect the spin Zeeman effect, which does not influence the orbital wave functions, and which can be trivially accounted for as a shift linear in B to the exchange energy.[21] The Hamiltonian of the pair reads

$$H = H_1^s + H_2^s + e^2/4\pi\epsilon\epsilon_0 r_{12}, \tag{1}$$

where ϵ is the dielectric constant and H^s the single-electron Hamiltonian

$$H^s = (-i\hbar\nabla + e\boldsymbol{A})^2/2m^* + V(x,y), \tag{2}$$

with m^* the electron band mass and $V(x,y)$ the potential of two Gaussian[22,23] dots

$$V(x,y) = -V_l e^{-[(x+d/2)^2+y^2]/R^2} - V_r e^{-[(x-d/2)^2+y^2]/R^2}, \tag{3}$$

where V_l and V_r are the depths of the left and right dots, respectively, d is the distance between the dot centers, and R is the radius of each of the dots. It was recently found[24] that the confinement energy as generated electrostatically in a gated two-dimensional electron gas is largest when the po-

tential has a Gaussian shape. The single-electron eigenfunctions of Hamiltonian (2) are obtained in the Landau gauge $\boldsymbol{A}=(-By,0,0)$ using a basis $\Psi_\mu(\boldsymbol{r})=\Sigma_{i=1}^{M}c_i^\mu\psi_{\boldsymbol{R}_i}(\boldsymbol{r})$ of the lowest Landau level eigenfunctions centered around points $\boldsymbol{R}_i=(X_i,Y_i)$:[18,25–27]

$$\psi_{\boldsymbol{R}_i}(\boldsymbol{r}) = \sqrt{\alpha_i}\exp\{-\alpha_i(\boldsymbol{r}-\boldsymbol{R}_i)^2/4 + ieB(x-X_i)(y+Y_i)/2\hbar\}/\sqrt{2\pi}. \quad (4)$$

Two-electron eigenfunctions $\chi(\boldsymbol{r}_1,\boldsymbol{r}_2)$ of Hamiltonian (1) are subsequently calculated in a basis of symmetrized (antisymmetrized) products of single-electron wave functions $\Psi_\mu(\boldsymbol{r}_1)\Psi_\nu(\boldsymbol{r}_2)$ for the singlet (triplet) two-electron states. Extensive discussion of the exact diagonalization with wave functions (4) is given in Ref. 18. Here we just remind the reader that the displaced lowest Landau wave functions also reproduce higher Fock-Darwin[1] states. We use 14 centers $\boldsymbol{R}_i$, i.e., 7 centers per dot, one in the center of each dot and six on a circle surrounding it. Radii of both circles as well as parameters α_i responsible for the localization of the wave functions (4) are optimized variationally.[28] Comparing the results for the magnetic field dependence of the exchange energy presented in Fig. 4 of Ref. 13 with the results of the present method applied to the model potential used therein[13] we find a nearly exact agreement with differences that do not exceed 5 μeV. However, contrary to the present approach, the basis used in Ref. 13, consisting of wave functions localized around the origin, is bound to be slowly convergent for larger interdot distances and/or high magnetic fields.

We use the material data of GaAs, $\epsilon=12.9$, $m^*=0.067$, and take the potential parameters $R=30$ nm, $V_r=25$ meV, $d=52$ nm (unless stated otherwise), and the value of V_l is varied to induce the asymmetry. For a single quantum dot ($V_l=0$) the energy spacing between the ground and first excited single-electron energy levels is 6.6 meV and between the first and the second excited energy levels the spacing equals 5.2 meV.

For the explanation of the results presented below we find it useful to introduce the single-electron basis consisting of lowest-energy states s_l, s_r, p_l, p_r, where $s(p)$ stands for the $0(-\hbar)$ angular momentum states and the subscript $l(r)$ denote the localization of the state in the left (right) dot. Figure 1 shows the confinement potential for a symmetric ($V_l=25$ meV) pair of coupled dots and for a pair with a small[19] asymmetry ($V_l=32$ meV). For $B=0$ the asymmetry-induced shift of the charge density to the left (deeper) dot is visibly stronger for the singlet state [see Figs. 1(a) and 1(b)]. At $B=0$ the exchange energy for the asymmetric system of coupled dots presented in Fig. 1(b) equals 0.32 meV and is three times larger than in the symmetric case of Fig. 1(a) for which $\Delta E=0.1$ meV (see below).

At high B the singlet and the triplet charge densities become identical [Figs. 1(c) and 1(d)] for both the symmetric and asymmetric coupling. At high magnetic field for which the probability of double occupancy of each of the dots vanishes (see below) both lowest-energy singlet and the lowest-energy triplet wave functions can be described using the

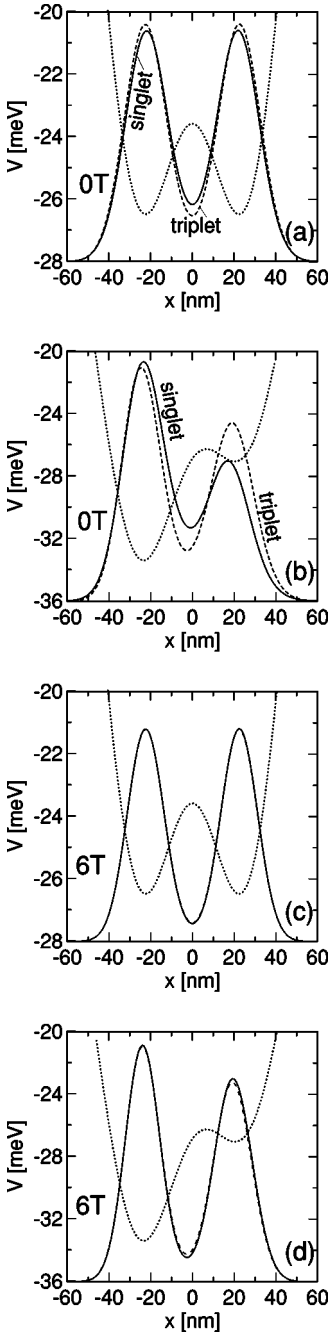

FIG. 1. Confinement potential (dotted line) and singlet (solid line) and triplet (dashed line) charge density plotted in arbitrary units at $y=0$ axis for $V_r=25$ meV and (a) $V_l=25$ meV and $B=0$, (b) $V_l=32$ meV and $B=0$, (c) $V_l=25$ meV and $B=10$ T, (d) $V_l=32$ meV and $B=6$ T.

lowest-energy wave functions localized in the left and right dot

$$\chi(\boldsymbol{r}_1,\boldsymbol{r}_2) = s_l(\boldsymbol{r}_1)s_r(\boldsymbol{r}_2) \pm s_r(\boldsymbol{r}_1)s_l(\boldsymbol{r}_2), \quad (5)$$

with a + sign for the singlet state and − for the triplet state (this function is not normalized). The charge density is obtained by integrating the two-electron probability density over coordinates of one of the electrons

$$\rho(\boldsymbol{r}) = \int d\boldsymbol{r}_2 |\chi(\boldsymbol{r},\boldsymbol{r}_2)|^2$$

$$= |s_l(\boldsymbol{r})|^2 + |s_r(\boldsymbol{r})|^2 \pm 2\mathrm{Re}[s_r^*(\boldsymbol{r}) s_l(\boldsymbol{r}) \int d\boldsymbol{r}_2 s_l^*(\boldsymbol{r}_2) s_r(\boldsymbol{r}_2)]. \quad (6)$$

As the magnetic field increases s_l and s_r wave functions become more strongly localized.[18] Finally, the overlap integral between these functions appearing in Eq. (6) vanishes leading to identical charge densities for the singlet and triplet states (see Fig. 1) and to the singlet-triplet degeneracy (see below). Similarly the singlet and triplet charge densities become indistinguishable in large quantum dots without the external magnetic field.[17] Note that the external magnetic field reduces the effect of the asymmetry on the charge densities [see Figs. 1(b) and 1(d)] due to an increased depth of the effective potential well at the electron localization positions.

Figure 2(a) shows the charge accumulated at the left of the origin $2\int_{-\infty}^{0} dx_1 \int_{-\infty}^{\infty} dy_1 \int d\boldsymbol{r}_2 |\chi(\boldsymbol{r}_1,\boldsymbol{r}_2)|^2$ as a function of the depth of the left quantum dot for $B=0$. For the symmetric system ($V_l=25$ meV) the charge is equally distributed between the dots. For $d=52$ nm near the symmetric point the charge in the left dot for both the triplet and the singlet is approximately a linear function of V_l, but the slope of the straight line for the singlet is more than twice steeper indicating that the triplet state is more robust against the inbalance (V_l/V_r). For larger barrier thickness ($d=60$ nm) the curves acquire a more stepwise character.

The probability of finding both electrons at the same side of the origin $(\int_{-\infty}^{0} dx_1 dx_2 \int_{-\infty}^{\infty} dy_1 dy_2 |\chi(\boldsymbol{r}_1,\boldsymbol{r}_2)|^2 + \int_{0}^{\infty} dx_1 dx_2 \int_{-\infty}^{\infty} dy_1 dy_2 |\chi(\boldsymbol{r}_1,\boldsymbol{r}_2)|^2)$, quantifying the double occupancy of the dots, is plotted as function of V_l in Fig. 2(b) for $d=52$ nm and $B=0$. For the symmetric system the double occupancy probability in the singlet state is almost twice as large as the one in the triplet state. When the left dot is 1/3rd deeper or shallower than the right dot the probability that the deepest dot is double occupied is roughly 50% in the singlet but only 10% in the triplet state.

The dependence of the exchange energy on the asymmetry for $B=0$ is presented in Fig. 2(c). The exchange energy is minimal for the symmetric system (i.e., $V_r=V_l=25$ meV). For a thick interdot barrier ($d=60$ nm) between symmetric dots the exchange energy is 0 due to the negligibly small tunnel coupling and complete charge separation. The exchange energy becomes nonzero only when the charge of both electrons in the singlet state starts to occupy the deepest dot [see Fig. 2(a)]. For thinner barriers [see plots for $d=52$ and 48 nm in Fig. 2(c)] even a small asymmetry increases the exchange energy.

The results of Figs. 2(a)–2(c) can be explained in the following way. In symmetric systems and in systems with a small[19] asymmetry the two-electron singlet (triplet) states consist mainly of the symmetrized (antisymmetrized) products given in Eq. (5). In the singlet state the double occupancy is introduced mainly by symmetric combinations $s_l(\boldsymbol{r}_1)s_l(\boldsymbol{r}_2)$ and $s_r(\boldsymbol{r}_1)s_r(\boldsymbol{r}_2)$, which, however, do not contribute to the triplet state due to the Pauli exclusion. The double occupancy in the triplet state can be realized by admixtures of states $\phi_r = \boldsymbol{A}\{s_r(\boldsymbol{r}_1)p_r(\boldsymbol{r}_2)\}$ and $\phi_l = \boldsymbol{A}\{s_l(\boldsymbol{r}_1)p_l(\boldsymbol{r}_2)\}$ ($\boldsymbol{A}$ stands for the antisymmetrizer). Triplets ϕ_r and ϕ_l correspond to maximum density droplets[1] confined in the right and left dots, respectively. In absence of the magnetic field these states have larger energies than $s_l(\boldsymbol{r}_1)s_l(\boldsymbol{r}_2)$ and $s_r(\boldsymbol{r}_1)s_r(\boldsymbol{r}_2)$ combinations resulting in a smaller double occupancy probability for the triplet state. In the presence of the asymmetry the singlet combination with a doubly occupied s energy level of the deepest dot has the lowest single-electron energy, which increases the double occupancy probability in the singlet state. The probability of the double occupancy in the triplet state also increases with asymmetry [see Fig. 2(b)],

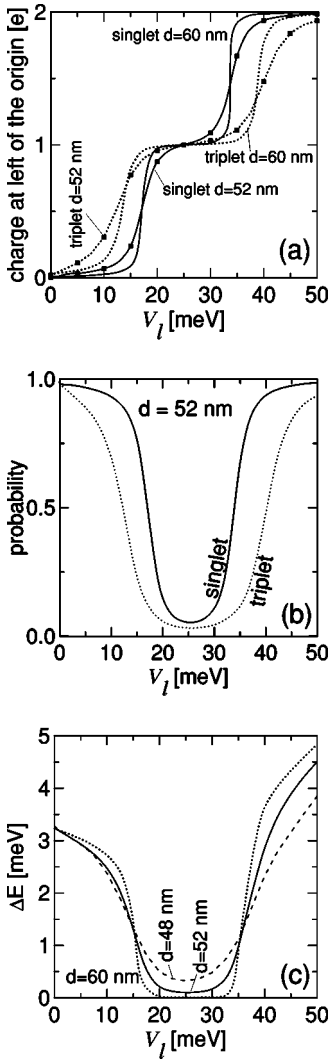

FIG. 2. (a) Charge accumulated at left of the origin for $d=52$ nm and $d=60$ nm in the singlet (solid lines) and triplet (dotted lines) states as function of the depth of the left quantum well V_l. Data for $d=52$ nm are marked with squares. (b) Probabilities that both electrons are on the same side of the origin for the singlet (solid lines) and triplet (dotted lines) states for $d=52$ nm. (c) Singlet-triplet energy difference as function of V_l. The values for $d=48$, 52 and 60 nm are plotted with dashed, solid and dotted lines, respectively.

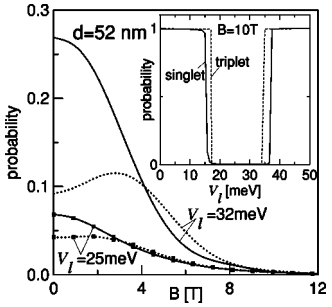

FIG. 3. Probability that both electrons are on the same side of the origin for singlets (solid lines) and triplets (dashed lines) for symmetric (V_l=25 meV) and asymmetric (V_l=32 meV) quantum dots as function of the magnetic field. Curves for the symmetric case are marked by squares. Inset: Same but for B=10 T as function of V_l.

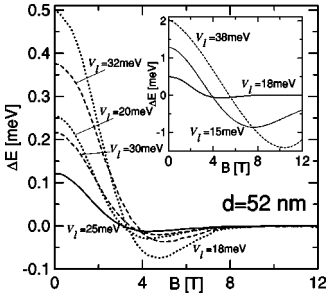

FIG. 4. Exchange energy as function of the magnetic field for V_r=25 meV and various values of V_l for which the electrons at high B occupy different dots. Inset: Exchange energy for V_l=18 meV (same as in the main figure) and for V_l=15 and 38 meV. For the two latter values the deeper dot is doubly occupied at high B.

but more slowly, since in the triplet state the p-excited state of the deepest dot has to be occupied. Therefore, the asymmetry lowers the energy of the singlet with respect to the triplet which explains the exchange energy enhancement by the asymmetry observed in Fig. 2(c).

Figure 3 shows that the double occupancy probability changes as a function of the magnetic field for a symmetric double dot and a double dot with a small[19] asymmetry. In both the symmetric and asymmetric dots the probability for the singlet decreases monotonically to zero with increasing field. A similar high-B limit behavior is observed for the triplet state. However, surprisingly, as the magnetic field is switched on the probability for the triplet initially increases. This is because in the subspace of states with both electrons in the deepest dot the lowest-energy-state undergoes singlet-triplet oscillations[1] as in the single-dot problem. Above a critical value of the magnetic field[29] the maximum density droplet states ϕ_r and ϕ_l acquire lower energy than the singlets built of products of s single-electron states confined in the same dot. This explains why beyond a certain value of the external field the probability of finding both electrons in the same dot is larger in the triplet than in the singlet state.

The discussed singlet-triplet energy crossings for the states confined in the single dot has a striking effect on the capacity of the deepest dot to bind both electrons for larger[19] asymmetry. The inset of Fig. 3 shows the probability that both electrons are in the same dot for B=10 T. Usually in the triplet state the electrons avoid each another more efficiently than in the singlet state due to the Pauli exclusion principle. However, contrary to the case of B=0 [see Fig. 2(b)] for B =10 T (see the inset of Fig. 3) counterintuitively, the electrons in the triplet state occupy the same dot more easily (for smaller asymmetry) than in the singlet state. At high magnetic field the lowest singlet and triplet energy levels corresponding to electrons occupying separate dots are degenerate [see the discussion after Eqs. (5) and (6)] but the lowest-energy state with both electrons in the deepest dot is the triplet maximum density droplet. As a consequence, the lowest triplet becomes localized in the deepest dot for smaller asymmetry than the lowest singlet.

The magnetic-field dependence of the exchange energy is displayed in Fig. 4. The magnetic field inducing singlet-triplet transition increases with the asymmetry, which is more effective for $V_l > V_r$ because of the increased strength of the confinement in the left (deeper) dot which weakens the relative effect of the external field. For large B the exchange energy tends to zero as long as the carriers are localized in separate dots at the high-magnetic field limit (see inset to Fig. 3). This is not the case for strongly asymmetric potentials for which both electrons stay in the same dot and for which the singlet-triplet oscillations continue to higher magnetic fields (see plots for V_l=15 meV and V_l=38 meV in the inset of Fig. 4).

In summary, we have studied the effect of the asymmetry on the lateral coupling of quantum dots in a perpendicular magnetic field using a numerically exact method. We have shown that at B=0 the exchange energy is minimal for a symmetric system of laterally coupled dots, and that the asymmetry promoting double occupancy of the deepest dot in the singlet state can enhance this by a factor of 4. If for practical reasons a stronger coupling between the dots was needed and the dots could not be made any smaller the solution is to make one of them even larger. We showed that for high magnetic fields localization of both electrons in the deepest potential minimum is easier if the electrons have parallel spins and explained this effect in terms of singlet-triplet oscillations in the lowest-energy state with both electrons in the deepest dot.

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[19] We consider the asymmetry to be *small* if at high magnetic field each of the dots is singly occupied in the ground state in contrast to the *large* asymmetry case for which only the deepest dot is occupied.
[20] For two electrons those in which $\mu_{11}=\mu_{10}=\mu_{01}$ and $\mu_{11}=\mu_{21}=\mu_{12}$, where μ_{ij} is the chemical potential of $i+j$ electrons, with i electrons in one dot and j in the other, see Ref. 10.
[21] Zeeman singlet-triplet splitting is $B\hbar g^*\mu_B$, where g^* is the effective Landé factor and μ_B the Bohr magneton. For GaAs this splitting is $0.025B$ meV/T.
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[28] We use two different variational parameters α_r and α_l for the seven centers associated with the right and the left dot, respectively.
[29] In a *single* dot the first spin-polarized ground-state appears between $B=5$ T and 15 T for $V=25$ meV and between $B=6.5$ T and 19.5 T for $V=35$ meV.