Canted phase in double quantum dots

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We perform a Hartree-Fock calculation in order to describe the ground state of a vertical double quantum dot in the absence of magnetic fields parallel to the growth direction. Intradot and interdot exchange interactions determine the singlet or triplet character of the system as the tunneling is tuned. At finite Zeeman splittings due to in-plane magnetic fields, we observe a quantum phase transition from a ferromagnetic to a symmetric phase through a canted antiferromagnetic state. The latter is obtained even at zero Zeeman energy for an odd number of electrons.

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Interaction in two-dimensional (2D) electron gases leads to new quantum phases when more degrees of freedom (external fields, spin and layer indices) are supplied to the system. In bilayer $\nu=2$ quantum Hall (QH) structures, theoretical calculations have predicted[1] and experimental evidence has confirmed[2] the existence of a particularly exotic canted antiferromagnetic (C) phase, which continuously connects the naively expected ferromagnetic (F) and paramagnetic (P) ground states (GS's) as the layer separation is tuned. Interesting predictions regarding C states in few-electron double quantum dots (DQD's) in the QH regime have also been reported.[3] Thus high magnetic fields seem an unavoidable condition to observe this quantum transition given that for vanishingly small magnetic fields, F and C states in bilayer systems would cost a good deal of kinetic energy. Our goal is to challenge this idea by lowering the system dimensionality and benefitting from the atomiclike spectrum of a semiconductor quantum dot.[4] In DQD's (termed artificial molecules as well) Coulomb-blockade effects,[5] magnetization,[6] and the formation of a delocalized molecular GS (Ref. 7) are some of the exciting features observed. From the theoretical viewpoint exact diagonalization methods,[8] Hubbard-based models,[9] and spin-density-functional theories[10] have been developed to show the presence of magic-number, molecule-type, and Hund's-rule-violating states in vertically coupled dots.

Here, we present a Hartree-Fock (HF) theory for addressing many-body effects in two vertically coupled parabolic quantum dots separated by a distance a with a total electron number N. We study this system in the absence of magnetic fields perpendicular to the dots. Still, in order to add spin symmetry breakings we allow for a parallel magnetic field whose coupling to the electronic orbital motion is neglected (a is assumed to be much smaller than the corresponding magnetic length). We are interested in quantum dots whose atomiclike character results in half-filled shells formed by quasidegenerate eigenstates, thus having large-spin expected values acting as effective magnets. Our main findings are (i) the existence of a robust C phase (envisaged as tilted spin vectors) at finite Zeeman energies for even values of N linking the F (fully spin polarized or, equivalently, triplet) and P (fully isospin polarized or singlet) GS's via a second-order phase transition; (ii) the persistence of C states for N odd, even in the *absence* of Zeeman gaps; and (iii) the overture of a simple model that qualitatively explains our results, allowing to deal with more complex quantum-dot systems.

The electron spatial coordinates are denoted by $\vec{r}=(x,\vec{\rho})$, where $\vec{\rho}=(y,z)$ and x is the growth direction. The wave function of the ith electron may be expanded in terms of 2D harmonic-oscillator eigenstates, $\phi_{nl}(\vec{\rho})$, $n=0,1,2,\ldots$ being the radial quantum number and l is the angular momentum obeying $l=-n,-n+2,\ldots,n-2,n$. Accordingly, the Hamiltonian reads

$$\mathcal{H}=\sum \varepsilon_{n_i} c^{\dagger}_{n_i l_i \sigma_i \alpha_i} c_{n_i l_i \sigma_i \alpha_i} - 2tI_x - \Delta_Z S_z + \frac{1}{2}\sum \mathcal{V}_{n_i l_i \alpha_i, n_j l_j \alpha_j, n_k l_k \alpha_k, n_m l_m \alpha_m} c^{\dagger}_{n_i l_i \sigma_i \alpha_i} c^{\dagger}_{n_j l_j \sigma_j \alpha_j} \times c_{n_k l_k \sigma_j \alpha_k} c_{n_m l_m \sigma_i \alpha_m}, \quad (1)$$

where the sums are extended to all indices, σ and α are the spin and layer indices, and $\varepsilon_n=\hbar\omega(n+1)$ ($\hbar\omega$ is the confinement strength). The second and third terms describe the coupling of the isospin and the spin of the system with external perturbations, namely, the tunneling t and the Zeeman splitting Δ_Z. The isospin points along $+(-)z$ when the electron is at the top (bottom) layer.[11] Notice that the tunneling term only switches layer indices, thus conserving the rest of quantum numbers. In the following $t>0$ is chosen so that bonding (antibonding) states have the lowest (highest) energy. S_z is the third component of the total spin and $\Delta_Z=g\mu_B B$, where g is the Landé factor, μ_B the Bohr magneton, and B the applied magnetic field in the z direction. $\mathcal{V}$ is the matrix element of the Coulomb potential $V(|\vec{r}-\vec{r}'|)$.

Because we seek to identify spin and particle-number broken symmetries that are reflected in the interdot coherence, we restrict our study to radial-symmetry-conserving solutions within an isolated dot despite the fact that spontaneously radial-symmetry-breaking states might take place.[12] For $N\gg 1$ we expect the electronic distribution to be radial. The resulting states ψ can thus be labeled with *individual* angular momenta: $\psi_{l_i}(\vec{r})=\Sigma_{n_i\sigma_i\alpha_i} d_{n_i\sigma_i\alpha_i}\phi_{n_i l_i}(\vec{\rho}) f_{\alpha_i}(x)\chi_{\sigma_i}$, so that the expected value of total angular momentum of the system is simply $\langle L\rangle=\Sigma_{i=1}^{N} l_i$. Here d are the coefficients of the expansion to be self-consistently calculated, $f_\alpha(x)$ is the

vertical component of the wave function, and χ_σ is the spin part. Hence the Hamiltonian is numerically diagonalized in separate l subspaces (though the matrix elements do depend on the total system configuration).

We consider that only the lowest energy states in the vertical structure are populated, and approximate the form of $f(x)$ as follows: $f_{\alpha=T}=\sqrt{\delta(x)}$ for the top layer, and $f_{\alpha=B}=\sqrt{\delta(x-a)}$ for the bottom layer. More precise expressions for $f(x)$ would involve accurate form factors entering the final results[13] but qualitatively all would remain the same. Then taking a close look at the x part of $\mathcal{V}$,

$$\mathcal{V}\propto\int dx\int dx' f_{\alpha_i}(x)f_{\alpha_j}(x')V(|\vec{r}-\vec{r}'|)f_{\alpha_k}(x')f_{\alpha_m}(x),$$

we observe that the only terms different from zero are either $\alpha_i=\alpha_j=\alpha_k=\alpha_m$ (*intra*dot interaction) or $\alpha_i=\alpha_m\neq\alpha_j=\alpha_k$ (*inter*dot interaction) since crossed terms (e.g., $\alpha_i=\alpha_k\neq\alpha_j=\alpha_m$) would be null because top and bottom wave functions have zero overlap.

In the HF approach the electron-electron interaction part of the Hamiltonian of Eq. (1) can be arranged in two parts: the Hartree operator ϑ^H,

$$\vartheta^H=\sum_{n_in_ml_il_m\sigma_i\alpha}\mathcal{H}_{n_in_ml_il_m}c^\dagger_{n_il_i\sigma_i\alpha}c_{n_ml_m\sigma_i\alpha}, \tag{2}$$

where

$$\mathcal{H}_{n_in_ml_il_m}\equiv\sum_{n_jn_kl_jl_k\sigma_j\alpha'}\mathcal{V}\langle c^\dagger_{n_jl_j\sigma_j\alpha'}c_{n_kl_k\sigma_j\alpha'}\rangle \tag{3}$$

(the indices of $\mathcal{V}$ are omitted for the sake of simplifying notation) and the exchange part ϑ^F,

$$\vartheta^F=-\sum_{n_in_kl_il_k\sigma_i\sigma_j\alpha\alpha'}\mathcal{X}_{n_in_kl_il_k\sigma_i\sigma_j\alpha\alpha'}c^\dagger_{n_il_i\sigma_i\alpha}c_{n_kl_k\sigma_j\alpha'}, \tag{4}$$

where

$$\mathcal{X}_{n_in_kl_il_k\sigma_i\sigma_j\alpha\alpha'}\equiv\sum_{n_jn_ml_jl_m}\mathcal{V}\langle c^\dagger_{n_jl_j\sigma_j\alpha'}c_{n_ml_m\sigma_i\alpha}\rangle. \tag{5}$$

Throughout our calculation, we make $\langle c^\dagger_{l_i}c_{l_j}\rangle\propto\delta_{l_i,l_j}$ according to the aforementioned radial-symmetry approximation. In Eq. (3) $\alpha'=\alpha$ gives rise to intradot Hartree interaction. The effect of this term is to make electrons repel each other within the parabolic well. The total energy of the dot is thus augmented. Interdot Hartree interaction is naturally included for $\alpha'\neq\alpha$, notwithstanding it does not have a strong influence in the final magnetic configurations, for it merely involves a rigid shift of the energy levels. However, we keep it for having the same number of electrons within each dot when N is even and for obtaining a more reliable value of the total energy of the system. Intradot exchange interaction favors spin alignment within each dot as expected. For $\alpha'\neq\alpha$ we are left with interdot exchange interaction. We stress that although the barrier separating the double well in the x direction is wide enough and consequently the vertical parts of ψ have negligible overlap (in our case the overlap is strictly zero owing to the Dirac-delta functions), the interdot exchange interaction is *not* zero[14] and cannot be disregarded as it plays a crucial role in the final DQD magnetic order. In fact, it is the competition between the intradot exchange part and the interdot exchange interaction plus the tunneling term and the Zeeman energy that drives the system from a GS dominated by intradot contributions (large values of the interdot distance or small tunneling parameter) to a state in which interdot effects prevail (small values of a or large t). In between, nontrivial quantum phases can occur. Our allowance of significant nonzero order parameters, $\langle c^\dagger_{\uparrow T}c_{\downarrow B}\rangle\neq0$, $\langle c^\dagger_{\uparrow T}c_{\downarrow T}\rangle\neq0$, etc., eventually leads to spontaneous spin symmetry breakings, spin rotations, canted phases, and the fact that the particle number at each dot is not a good quantum number.

At this point a small digression about the trustworthiness of the HF model is needed. Having studied in detail the differences between (un)restricted HF theories and exact diagonalization methods in quantum dots, previous works[15] demonstrate that HF are well suitable within a broad range of N. It seems clear that a large number of electrons would result in a negligible amount of quantum fluctuations that are unlikely to destroy mean-field-based predictions. Furthermore, we can take advantage of large values of $\langle S_z\rangle$ (highest half-filled shells) to ease the appearance of C phases. Incidentally the existence of lower closed shells (hereafter designated as *core*) is a crucial difference between QH systems and DQD's in the absence of magnetic fields. In the former case, only the lowest Landau level is occupied and the kinetic energy plays a minor role. In the latter, the dot fills its levels following an Aufbau rule, thereby closing shells as N is increased.

Now, large values of N tend to contract the (renormalized) energy level interspacing in order to build a semiclassical radial density. This may involve the drop of valence electrons below the core levels and a subsequent reduction of $\langle S_z\rangle$. A more favorable situation can be accomplished in part by enhancing the confinement. Hence one should reach a compromise between these competing factors.[16]

The expansion of ψ is enlarged enough, in such a way that the highest n state contributes less than 0.01% to a typical density. We present data for $N=32$ (though similar results are found for $N=18$), setting $\hbar\omega=30$ meV and $a=l_0/2$, $l_0=\sqrt{\hbar/(m^*\omega)}$ being the harmonic-oscillator typical length (m^* is the GaAs effective mass). As a result, the single-particle value of the S_z projection onto the layer α is $\langle S_{z\alpha}\rangle=2$.

Figure 1(a) depicts the total energy of the system, $E=\langle\mathcal{H}\rangle$, as a function of the tunneling parameter. At low t our calculations show for the parameters chosen above that the GS is ferromagnetic with $\langle L\rangle=0$ and $\langle S_z\rangle=4$. In this phase the intradot interaction is more important than the interdot one plus the tunneling term. Because this is a fully spin-polarized state, its energy does not depend on t and remains constant until the system undergoes a continuous quantum phase transition[17] to the C phase. In this case, the system lowers its energy by increasing the tunneling contribution. This favors the formation of singlets as well as the increase

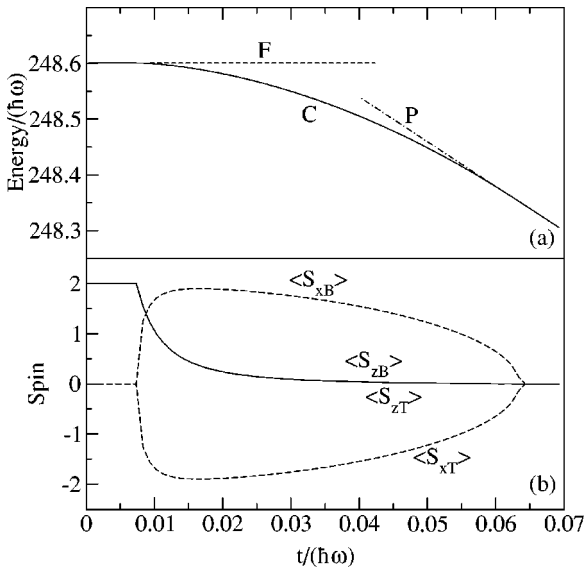

FIG. 1. (a) Total energy of a DQD with $n=32$, $\hbar\omega=30$ meV, $a=l_0/2$, and $\Delta_Z=0.017\hbar\omega$. A dashed (dot-dashed) line shows the behavior of the $F(P)$ state provided spontaneous spin symmetry breaking had not been taken place. (b) Expectation values of the total spin for each dot. z (x) components are drawn in full (dashed) lines.

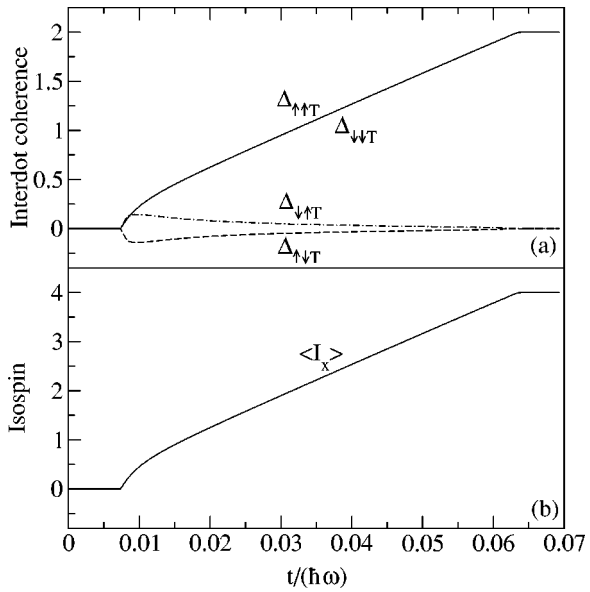

FIG. 2. (a) Up-up and down-down spin interdot coherence (full lines) for the top layer. Up-down (dashed line) and down-up (dot-dashed line) spin interdot coherence is also shown. They are different from zero only in the C phase. (b) x component of the total isospin. $\langle I_z\rangle=0$ throughout the tunneling range.

of interdot coherence (see below). To make the loss of Zeeman energy as small as possible the spin configuration is then canted. By further enhancement of t, the numerical simulations prove that a P phase fulfilling $\langle L\rangle=0$ and $\langle S_z\rangle=0$ is achieved and a linear dependence of E on the tunneling is obtained. The C phase is thus a linear combination of the wave functions associated with F and P states. In Fig. 1(b) we have plotted $\langle S_{z\alpha}\rangle$ for both dots. Its maximum is reached when the DQD is F, then it is depressed while entering the C phase, and eventually we are left with the singlet state. Notice that the entire conversion has been realized in a continuous manner. Indeed, that $\langle S_z\rangle$ varies continuously with tunneling is an artifact of the (semiclassical) mean-field approach. Exact diagonalizations show[3] that quantum-mechanically $\langle S_z\rangle$ changes in a discrete way, since S_z is a good quantum number. However, for large values of S_z the continuous variation is a reasonable approximation, as in the thermodynamic limit the U(1) spin symmetry is spontaneously broken.[1] From Fig. 1(b) we see that $\langle S_{xT}\rangle=-\langle S_{xB}\rangle$. This is the key feature of the appearance of a C phase—total spin components in the plane perpendicular to the field that are antiferromagnetically correlated. The governing physics is analogous to what is commonly found in QH bilayers and disks[1,3] but the origin is quite dissimilar. Here the dots behave as tiny magnets due to the Hund's rule (a feature stemming uniquely from the atomiclike character of the dots) and the role of the magnetic field applied in the plane of the dots is only to break the SU(2) spin symmetry by marking a privileged direction for the spin.

These conclusions may be reinforced by examining the interdot coherence of the top (bottom) quantum dot: $\Delta_{\sigma\sigma' T(B)}=\Sigma_{n_i,l_i}\langle c^\dagger_{n_i l_i \sigma T(B)} c_{n_i l_i \sigma' B(T)}\rangle$. It can be shown[18] that all the $\Delta_{\sigma\sigma'\alpha}$ components are zero in the triplet phase [see Fig. 2(a)]. As t increases, the system acquires interdot coherence until it is completely coherent in the symmetric state (for which $\Delta_{\uparrow\downarrow\alpha}=\Delta_{\downarrow\uparrow\alpha}=0$ and $\Delta_{\uparrow\uparrow\alpha}=\Delta_{\downarrow\downarrow\alpha}\neq 0$). As C phases involve a spin symmetry breaking, $\Delta_{\uparrow\downarrow\alpha}=-\Delta_{\downarrow\uparrow\alpha}$. Moreover, since $\langle I_x\rangle$ yields half the difference between the number of electrons in symmetric states and those in antisymmetric states, $\langle I_x\rangle$ is zero in the F case and reaches its maximal value in the P case. As Fig. 2(b) shows, the C phase develops intermediate quantities. The z isospin component would be different from zero, provided there is more charge in one of the wells, e.g., by applying an electric bias; but this is not the present case.

In Fig. 3(a) we draw the entire phase diagram that characterizes the distinct GS's as a function of the Zeeman energy and the tunneling. For large Δ_Z and small t the DQD is in the spin-polarized phase. In the opposite limit, the singlet-state energy is lower. The C phase lies between them. In the case of $\Delta_Z=0$ we obtain a purely antiferromagnetic or Néel GS with the spins pointing across from each other.

A more striking feature is observed when a hole is introduced into the system. For N odd the highest-lying shells are not closed and the remaining hole is shared by the two dots. From Fig. 3(b) we see that the region covered by the C phase is reduced at large t because the system takes advantage more easily of the possibility of tunneling by forming singlets. Likewise the F state is more energetically favored at low transmissions. There is a range in the tunneling parameter at $\Delta_Z=0$, where the lack of charge spontaneously induces ferromagnetism. But now the C state is extended even for $\Delta_Z=0$ since a Néel phase cannot exist for an odd number of electrons.

A simple model may be aimed to shed light on this phenomena. When the highest shell is occupied with an even

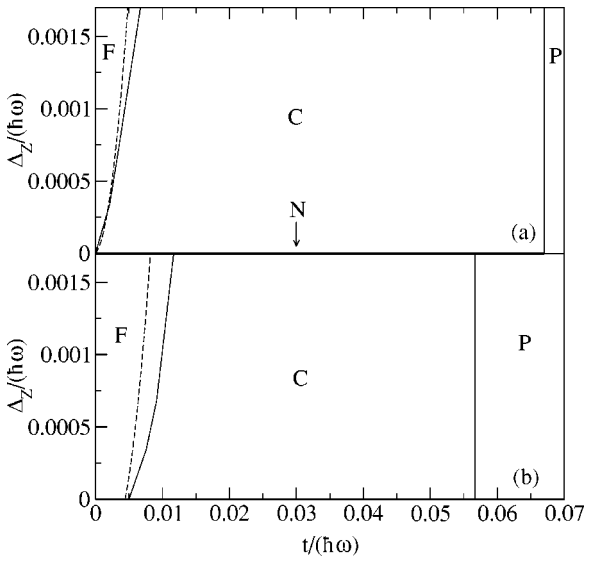

FIG. 3. (a) Phase diagram for 32 electrons. Full lines correspond to the numerical calculation. The dashed line is obtained from a simple model. The infinite slope of the *C*-*P* boundary seems to be correct in the thermodynamic limit (Ref. 1). (b) Same as (a) for $N=31$.

number of electrons, it is reasonable that a Heisenberg term accounts for the antiferromagnetic phase. In addition, the total energy must include a contribution stemming from the Zeeman energy that favors a parallel spin alignment. Close to the *F* phase, we propose the following energy functional:

$$E_{t,\Delta_Z}(\theta) = -J(t)\vec{S}_T\cdot\vec{S}_B - g\mu_B\vec{B}\cdot(\vec{S}_T+\vec{S}_B)$$
$$= -J(t)S^2\cos\theta - 2\Delta_Z S\cos(\theta/2). \qquad (6)$$

Here, we consider the total spins as classical vectorial entities centered at each dot and assume $|\vec{S}_T|=|\vec{S}_B|\equiv S$, θ being the angle spanned by both vectors. $J(<0)$ is a parameter fitted from the dependence of the total energy on t at $\Delta_Z=0$ [one curve analogous to Fig. 1(a)]. As a result, $J(t)$ is roughly quadratic for small t. Then the critical line marking the transition to the canted phase from the *F* state is achieved by setting $E''(\theta=0)=0$ that yields $\Delta_Z(t)=-4J(t)S/2$. This gives the piece of parabola shown in Fig. 3(a). The curve crosses the abscissa axis at $t=0$ proving that no *F* state can exist with $\Delta_Z=0$ at finite t and the DQD takes on a *N* phase.

When an electron is removed from the DQD, the remaining hole prefers to keep its spin parallel to the rest while hopping from dot to dot. Therefore, the system is sensitive to the particular spin orientation of the hole and Eq. (6) must include a term accounting for this fact: $E\rightarrow E-t\cos(\theta/2)$ (this is as if we had done a unitary rotation and kept only the diagonal terms) resulting in $\Delta_Z(t)=[t-4J(t)S^2]/2S$. Unlike the $N=32$ case, for $N=31$, $\theta=0$ is a minimum of E at small values of t (in the interval of physical interest, i.e., $[0,\pi]$) and the system remains spin polarized [see Fig. 3(b)]. Larger t means that $\theta=0$ corresponds to a relative maximum of $E(\theta)$ and one minimum at $\theta\neq 0$ shows up, fulfilling that the *C* phase is now the lowest energy state. Despite the simplicity of the model, the curves agree remarkably well with the self-consistent numerical solutions.

In summary, our analysis of the GS of a vertical DQD based upon a mean-field framework predicts the existence of a canted phase for intermediate tunneling and not too high Zeeman energies. For a sufficiently high even electron number (for which quantum-correlation effects are not expected to qualitatively alter the conclusions) the *C* phase continuously connects numerically found *F* and *P* states as the tunneling parameter is varied. When a hole is created within the highest half-filled shells, the kinetic energy of the remaining electron promotes the *F* phase at small t and the *C* phase arises even at *zero* (arbitrarily small) Zeeman splitting. A simple model is addressed to interpret these phenomena.

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[16] We have carefully checked that our HF results for an isolated dot showed an excellent agreement with the existing literature.

[17] Although rigorously speaking the term ''phase transition'' should be only employed in physical problems related to extended systems, we utilize it to denote the change between GS's with distinct symmetries.

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