Finite-thickness effects in ground-state transitions of two-electron quantum dots

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Using the exactly solvable excitation spectrum of two-electron quantum dots with parabolic potential, we show that the inclusion of the vertical extension of the quantum dot provides a consistent description of the experimental findings of Nishi et al. [Phys.Rev.B 75, 121301(R) (2007)]. We found that the second singlet-triplet transition in the ground state is a vanishing function of the lateral confinement in the three-dimensional case, while it always persists in the two-dimensional case. We show that a slight decrease of the lateral confinement leads to a formation of the Wigner molecule at low magnetic fields.

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Two-electron quantum dots (QDs) have drawn a great deal of experimental and theoretical attention in recent years [1, 2, 3]. Experimental data including transport measurements and spin oscillations in the ground state under a perpendicular magnetic field in two-electron QDs may be explained as a result of the interplay between electron correlations, a two-dimensional (2D) lateral confinement and magnetic field. A 2D interpretation of experiments, however, leads to inconsistencies [2, 4], providing, for example, too low values of the magnetic field for the first singlet-triplet (ST) transition. There is no consensus on origin of this disagreement, since various experiments are dealing with different QDs. Evidently, it is important to understand basic sources of such inconsistencies from view point of possible technological applications, since QDs may provide a natural realization of quantum bit. This problem is also related to fundamentals of strongly correlated finite systems, which are different from bulk and can be controlled experimentally.

It was predicted that the ground state of N-electron quantum dot becomes the spin polarized maximum density droplet (MDD) [5] at high magnetic field. For a two-electron QD it is expected that the MDD occurs after a first ST transition (see discussion in [2]). Theoretical calculations [6] assert that after the first ST transition the increase of the magnetic field induces several ground state transitions to higher orbital-angular and spin-angular momentum states. This issue was addressed in transport study of the correlated two-electron states up to 8T and 10 T in a lateral [7] and vertical [8, 9] QDs, respectively. It is quite difficult to detect the structure of ground states after the first ST transition in a lateral QD due to a strong suppression of the tunnel coupling between the QD and contacts. Altering the lateral confinement strengths, the transitions beyond the first ST transition are reported in vertical QDs [8]. In fact, the variation of the confining frequency with the same experimental setup opens a remarkable opportunity in the consistent study of effects of the magnetic field on electron correlations. One of the major aspects of the present paper is to demonstrate that the experimental results found in Refs [8] can be explained if one takes into account the 3D physical nature of the QD. We will discuss the additional criterion to distinguish the 2D and 3D nature experimentally and will analyze the formation of the Wigner molecule in the 3D two-electron QD.

Three vertical QDs with different lateral confinements have been studied in the experiment [8]. In all samples clear shell structure effects for an electron number $N = 2, 6, ...$ at $B = 0$ T have been observed, implying a high rotational symmetry. Although there is a sufficiently small deviation from this symmetry in sample C (from now on in accordance with the list of Refs [8]), a complete shell filling for two and six electrons was observed. Such a shell structure is generally associated with a 2D harmonic oscillator (x-y) confinement [2]. However, it is noteworthy that a similar shell structure is produced by a 3D axially-symmetric harmonic oscillator (HO) if the confinement in the z-direction $\omega_z = 1.5 \omega_0$ is only slightly larger than the lateral confinement ($\omega_x = \omega_y = \omega_0$). In this case six electrons fill the lowest two shells with $n_z = 0$. It was found also that the lateral confinement frequency for the axially-symmetric QD decreases with the increase of the electron number [10], since the screening in the lateral plane becomes stronger with large electron number. In turn, this effectively increases the ratio $\omega_z/\omega_0$ making the dot to be more ”two-dimensional”, since the vertical confinement is fixed by the sample thickness. Indeed, the N-dependence of the effective lateral frequency is observed in [8]. All these facts imply that the three-dimensional nature is a prerequisite of a consistent quantitative analysis of small QDs with a few electrons.

Our analysis is carried out by means of the exact diagonalization of the Hamiltonian for two 3D interacting electrons in a perpendicular magnetic field:

$$H = \sum_{j=1}^{2} \left[ \frac{1}{2m} \left( \mathbf{p}_j - \frac{e}{c} \mathbf{A}_j \right)^2 + U(r_j) \right] + \frac{\alpha}{|\mathbf{r}_1 - \mathbf{r}_2|} + H_{\text{spin}}.$$  \hspace{1cm} (1)

Here $\alpha = e^2/4\pi\varepsilon_0\varepsilon_r$ and $H_{\text{spin}} = g^* \mu_B (\mathbf{s}_1 + \mathbf{s}_2) \mathbf{B}$ describes the Zeeman energy, where $\mu_B = e\hbar/2m_e c$ is the...
Bohr magneton. The effective mass is \( m^* = 0.067 m_e \), the relative dielectric constant of a semiconductor is \( \varepsilon_r = 12 \) and \( |g^*| = 0.44 \) (bulk GaAs values). For the perpendicular magnetic field we choose the vector potential with gauge \( A = 4B \times r = \frac{1}{2}B(-y, x, 0) \). The confining potential is approximated by a 3D axially-symmetric HO

\[
U(r) = m^*\left[\omega_0^2(x^2+y^2)+\omega_z^2z^2\right]/2,
\]

where \( \hbar\omega_z \) and \( \hbar\omega_0 \) are the energy scales of confinement in the \( z \)-direction and in the \( xy \)-plane, respectively.

The evolution of the ground-state energy of a two-electron QD under the perpendicular magnetic field can be traced by means of the additional energy \( \Delta E = \mu(2, B) - \mu(1, B) \), where \( \mu(N, B) = E(N, B) - E(N - 1, B) \) and \( E(N, B) \) denotes the total energy of the QD with \( N \) electrons under a magnetic field of the strength \( B \). \( \Delta E \) fitting the \( B \)-field dependence of the first and second Coulomb oscillation peak positions to the lowest Fock-Darwin energy levels of the 2D HO with the potential \( m^*\omega_0^2/2 \), Nishi et al. \( \cite{8} \) estimated \( \omega_0 \) for all three samples A, B, C. Although the general trend in the experimental data is well reproduced by the 2D calculations, the experimental positions of the ST transition points are systematically higher (see Fig.3 of Ref. \( \cite{8} \)). Different lateral confinements in the above experiment are achieved by the variation of the electron density, without changing the sample thickness. Using the “experimental” values \( \omega_x \) for the lateral confinement and the confinement frequency \( \omega_z \) as a free parameter, we found that the value \( \hbar\omega_z = 8 \text{ meV} \) provides the best fit for the positions of kinks in the additional energy

\[
\Delta E = \hbar\omega_0\varepsilon - E(1, B) + E_Z \tag{2}
\]

in all three samples. Here, \( \hbar\omega_0\varepsilon \) is the relative energy, \( E(1, B) = \hbar\omega_0 + \hbar\omega_z/2 \) and the Zeeman energy \( E_Z \) is zero for the singlet states (we recall that the total Hamiltonian \( \hat{H} \) is separated onto the center-of-mass Hamiltonian, the Hamiltonian of the relative motion and the Zeeman term). For the sake of illustration, we display in Fig.1 the magnetic dependence of the experimental spacing between the first and the second Coulomb oscillation peaks \( \Delta V_y = V_y(2) - V_y(1) \) for samples A–C, which can be transformed to the additional energy \( \Delta\mu \) (see details in Refs. \( \cite{8, 9} \)). In the \( \Delta V_y - B \) plot, ground state transitions appear as upward kinks and shoulders \( \cite{9} \). It was found from the Zeeman splitting at high magnetic fields that \( |g^*| = 0.3 \) \( \cite{8} \) and we calculate the additional energy with this and the bulk values.

We nicely reproduce the experimental position of the first ST transitions at \( B = 4.2, 3, 2.3 \text{ T} \) in samples A, B, and C, respectively (see Fig.1). When the magnetic field is low, a difference between the calculations with different \( |g^*| \) factors is negligible. Upon decreasing the lateral confinement \( \hbar\omega_0 \) from sample A to sample C (the increase of the ratio \( \omega_z/\omega_0 \)), the Coulomb interaction becomes dominant in the interplay between electron correlations and the confinement \( \cite{11} \). In turn, the smaller the lateral confinement at fixed thickness is (the stronger is the electron correlations) the smaller the value of the magnetic field is at which the ST transitions or, in general, crossings between excited states and the ground state may occur.

![Figure 1](https://example.com/figure1.png)

**FIG. 1:** (Color online) The magnetic dependence of the additional energy \( \Delta E \) in two-electron QDs with lateral confinements \( \hbar\omega_0 = 4.2, 3.7, 3.5, 2.9 \text{ meV} \) (the first, second and fourth values are experimental values for sample A, B, and C, respectively \( \cite{8} \)). The confinement in the third \( (z) \) direction \( \hbar\omega_z = 8 \text{ meV} \) is fixed for all samples. The results for \( |g^*| = 0.3(0.44) \) are connected by solid (dotted) line for \( \hbar\omega_0 = 4.2, 3.5, 2.9 \text{ meV} \) and by dashed (dotted) line for \( \hbar\omega_0 = 3.7 \text{ meV} \). The solid grey lines display the experimental spacing \( \Delta V_y \) as a function of \( B \). The arrows identify the position of experimental ground state transitions \( \cite{8} \).

There is no signature of the second crossing in the ground state for sample A at large \( B \) (up to 10 T). Here, the ratio \( \omega_z/\omega_0 \approx 1.9 \) and the effect of the third dimension is most visible: the confinement has a dominant role in the electron dynamics and very high magnetic field is required to observe the next transition in the ground state due to electron correlations. Thus, the MDD phase survives until very high magnetic fields (\( B \approx 10 \text{ T} \)).

A second kink is observed at \( B = 7 \text{ T} \) in sample B \( \cite{8} \). Our calculations with the “experimental” lateral confinement \( \hbar\omega_0 = 3.7 \text{ meV} \) produces the second kink at \( B = 9.5 \text{ T} \), which is located higher than the experimental value. The slight decrease of the lateral frequency until \( \hbar\omega_0 = 3.5 \text{ meV} \) shifts the second kink to \( B = 8.7 \text{ T} \), improving the agreement with the experimental position of the first ST transition as well. In addition, the use of \( |g^*| = 0.3 \) (instead of the bulk value) with the latter frequency creates a plateau, which bears resemblance to the experimental spacing \( \Delta V_y \). However, there is no detailed information on this sample and we lack a full understanding of this kink. It seems there is an additional mechanism responsible for the second kink in sample B.

The most complete experimental information is related to sample C and we also study this sample in detail. In sample C the first experimental ST transition occurs at \( B = 2.3 \text{ T} \), while the signatures of the second and the third ones are observed at \( B \approx 5.8, 7.1 \text{ T} \), respectively (see Fig.1). The 2D calculations (with the “experimental” values \( \hbar\omega_0 = 2.9 \text{ meV}, |g^*| = 0.44 \)) predict the first, second and third ST crossings at lower magnetic fields:
$B = 1.7, 4.8, 5.8 \, T$, respectively (see Fig.2). The results can be improved to some degree with $|g^*| = 0.3$. To reproduce the data for $\Delta \mu$ Nishi et al. \cite{Nishi} have increased the lateral confinement ($\hbar \omega_0 = 3.5 \, meV$, $|g^*| = 0.44$). As a result, the first, second and third ST transitions occur at $B = 2, 6.3, 7.5 \, T$, respectively. Evidently, 2D calculations overestimate the importance of the Coulomb interaction. The increase of the lateral confinement weakens simply the electron correlations in such calculations. In contrast, the 3D calculations reproduce quite well the positions of all crossings with the “experimental” lateral confinement ($\hbar \omega_0 = 2.9 \, meV$ at $B = 2.3, 5.8, 7.1 \, T$ (see Fig.2).

\begin{figure*}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig2.png}
\caption{(Color online) Magnetic field dependence of the addition energy $\Delta \mu$ for the 2D model with $\hbar \omega_0 = 2.9, 3.5 \, meV$ and for the 3D model ($\hbar \omega_0 = 2.9 \, meV$, $\hbar \omega_z = 8 \, meV$). Ground states are labeled by $(M, S)$, where $M$ and $S$ are the total orbital momentum and the total spin, respectively. Grey vertical lines indicate the position of the experimental crossings between different ground states.}
\end{figure*}

One of the questions addressed in the experiment \cite{Nishi} is related to a shoulder-like structure observed in a small range of values of the magnetic field (see our Fig.4 and Fig.4 of \cite{Nishi}). This structure is identified as the second singlet state $(2, 0)$ that persists till the next crossing with the triplet state $(3, 1)$. According to Ref.\cite{Nishi}, the ground state transition from the triplet $(1, 1)$ state to the singlet $(2, 0)$ is associated with the collapse of MDD state for $N = 2$. Therefore, a question arises: at which conditions it would be possible to avoid the collapse of the MDD phase (in general, to preserve the spin-polarized state); i.e., at which conditions the singlet $(2, 0)$ state never will show up in the ground state. In fact, the collapse of the MDD depends crucially on the value of the lateral confinement and the dimension of the system. We found that in the 2D consideration the $(2, 0)$ state always exists for experimentally available lateral confinement (see Fig.3). Moreover, in this range of $\omega_0$ the 2D approach predicts the monotonic increase of the interval of values of the magnetic field $\Delta B$, at which the second singlet state survives, with the increase of the lateral confinement. In contrast, in the 3D calculations, the size of the interval is a vanishing function of the lateral confinement for a fixed thickness ($\hbar \omega_z = 8 \, meV$). It is quite desirable, however, to measure this interval to draw a definite conclusion and we hope it will done in future.

\begin{figure*}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig3.png}
\caption{(Color online) The interval $\Delta B$ in which the singlet state $(2, 0)$ survives as a function of the lateral confinement for 2D and 3D calculations. The confinement in the third (z) direction $\hbar \omega_z = 8 \, meV$ is fixed for the 3D calculations.}
\end{figure*}

As discussed above, the decrease of the confinement at fixed thickness increases the dominance of the electron correlations in the electron dynamics. Furthermore, this decrease, related to the decrease of the electron density \cite{Nishi, Kohn}, creates the favorable conditions for onset of electron localization. This localization (crystallization) in QDs is associated with the formation of the so-called Wigner molecule \cite{Wigner}. In the 2D approach the crystallization is controlled by the ratio of Coulomb and confinement strengths $R_W = (\alpha/\bar{\alpha})/\hbar \omega_0$ ($\bar{\alpha} = (\hbar/m^* \omega_0)^{1/2}$) (cf.\cite{Kohn}), which is about $R_W \sim 3$ for the QDs considered in experiments \cite{Nishi}. For a 2D two-electron QD, it is predicted that the Wigner molecule can be formed for $R_W \sim 200$ at zero magnetic field \cite{Saito}, or at very high magnetic field \cite{Bedell} (for $\hbar \omega_0 \sim 3 \, meV$ and small $R_W$ such as in the experiments \cite{Nishi}). In the 3D axially-symmetric QDs the ratio between vertical and lateral confinements (anisotropy) may, however, affect the formation of the Wigner molecule. This problem can be analyzed by dint of the electron density

$$n(r) = \int \left[ |\Psi(r, r')|^2 + |\Psi(r', r)|^2 \right] \, dr',$$

when one electron is at the position $\mathbf{r}$ if the other one is located at a position $\mathbf{r}'$. A criterion for the onset of the crystallization in QDs can be the appearance of a local electron density minimum at the center of the dot \cite{Zaikin}. For 2D QDs this leads to a radial modulation in the electron density, resulting in the formation of rings and roto-vibrational spectra \cite{Shastry}.

Our analysis of the conditions realized in the experiments \cite{Nishi} predicts very high magnetic fields ($B > 12 \, T$) for the formation of the Wigner molecule. However, with a slight decrease of the lateral confinement, at $\hbar \omega_0 = 2 \, meV$ we obtain the desired result. The 3D analysis of
The magnetic dependence of the ground state in 2D and 3D approaches for a lateral confinement $h\omega_0 = 2 \text{ meV}$ is depicted in the 2D (left) and 3D (right) electron densities. The density grows from a central small core over a ring to a torus with the increase of the magnetic field.

**FIG. 4** (Color online) Top: the magnetic dependence of the ground state in 2D and 3D approaches for a lateral confinement $h\omega_0 = 2 \text{ meV}$. The 2D (left) and 3D (right) electron densities are displayed for different ground states $(M, S)$ at corresponding magnetic fields. The largest 3D density grows from a central small core over a ring to a torus with the increase of the magnetic field.

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