On the formation of Wigner molecules in small quantum dots

S.M. Reimann, M. Koskinen, and M. Manninen
Department of Physics, University of Jyväskylä, FIN-40351 Jyväskylä, Finland

It was recently argued that in small quantum dots the electrons could crystallize at much higher densities than in the infinite two-dimensional electron gas. We compare predictions that the onset of spin polarization and the formation of Wigner molecules occurs at a density parameter \( r_s \approx 4 a_B^* \) to the results of a straight-forward diagonalization of the Hamiltonian matrix.
PACS 73.20.Dx, 71.45.Gm, 85.30.Vw

I. INTRODUCTION

If the number of electrons artificially confined on a quasi two-dimensional electron island (made for example in a semiconductor heterostructure) is very large, many properties of such a so-called “quantum dot” or “artificial atom” \([3]\) can be described from what is known about the limit of the infinite (two-dimensional) electron gas (2DEG). Until now, most experiments \([2]\) were performed at electron densities which are slightly below the equilibrium density of the 2DEG. The liquid-like properties then still dominate. For only a few trapped particles (as experimentally realized in vertical quantum dots \([3]\)), pronounced addition energy maxima as a consequence of shell structure and aligned spins in the mid-shell regions due to Hund’s rules were observed in close analogy to atomic physics \([3]\). Even the simplest picture of \( N \) non-interacting particles in a two-dimensional harmonic trap could explain many features of the conductance spectra. For larger systems, mean field approaches like Hartree-Fock \([13]\) or density functional methods \([14,15]\) were applied. In the small-\( N \) limit, much theoretical work focused on exact diagonalization techniques \([13,14]\). This approach was mostly used for dots in magnetic fields, where correlations become increasingly important with field strengths. It was particularly successful in the (integer and fractional) quantum Hall regime where one can restrict the basis set to the lowest spin-polarized Landau level \([15,16]\). Quantum Monte Carlo (QMC) \([17–20]\) methods provide alternative approaches yielding energies whose accuracy reaches that of exact diagonalizations.

When the electron density is lowered and the Coulomb energy increases relative to the kinetic energy, correlations begin to strongly dominate the electronic structure also in the absence of magnetic fields. For densities smaller than a certain critical value a Wigner crystallization is expected at very low densities. Monte-Carlo calculations indicate that in the 2D bulk a transition to a Wigner crystal-like state, preceded by a transition to a polarized phase \([22]\), occurs only at densities corresponding to Wigner-Seitz radii \( r_{s,2D} > 37 a_B^* \) \([23]\) (the density \( n_0 \) and \( r_{s,2D} \) are related by \( n_0 = 1/(\pi r_{s,2D}^2) \)), whereas in 3D the classical limit lies as high as \( r_{s,3D} = 100 a_B^* \) \([4]\). (In the following, for simplicity we write \( r_{s,2D} = r_s \).) Chui and Tanatar \([22]\) found that in 2D systems without translational invariance the critical density parameter for a fluid-solid transition is shifted to a considerably smaller value \( r_s \approx 7.5 a_B^* \). Could this be important for finite systems such as the above mentioned lateral or vertical semiconductor quantum dot structures? This question was recently posed \([21,23]\) and it was argued that in finite systems confining only a few particles localization would indeed occur at significantly higher densities than in the 2D bulk. In the Wigner limit the few electrons in the trap would distribute such that their electrostatic repulsion is minimized. The internal structure of the wave function of the many body system should then have the symmetry of the corresponding classical charge distribution. Wigner crystallization was found to be particularly pronounced in quantum dots with steep walls and polygonal geometry \([24]\). Egger et al. \([5]\) have performed quantum Monte Carlo studies using a multilevel blocking algorithm \([25]\). For parabolic quantum dots with azimuthal symmetry they reported that at a critical density of \( r_s = 4 a_B^* \) the formation of Wigner molecule-like ground states should become energetically favorable. Hartree-Fock (HF) methods by definition fail to accurately describe the correlated regime. When performed in an unrestricted scheme, however, spontaneous symmetry breaking and localization in the spatial distribution of the electronic densities of quantum dots and lateral quantum dot molecules at \( r_s \approx 3.5 a_B^* \) was attributed to the onset of Wigner crystallization \([3]\).

In the present article we report numerically exact configuration interaction calculations. This method has a long history in quantum chemistry, and was applied to quantum dots by many different groups \([4]\). Much of the previous work, however, concentrated on the electronic structure in large magnetic fields where the electron gas is polarized. Our purpose here is a comparison of the exact diagonalization results to the above mentioned recent predictions of localized states in the low-density limit and zero magnetic field. We first give a brief outline of the configuration interaction method and then turn to a discussion of the many-body spectra of a six-electron quantum dot at zero angular momentum as a function of the average electron density in the dot. Calculations for different \( r_s \)-values indicate that the ground state remains unpolarized. At values of \( r_s \) which are accessible to exact diagonalization techniques, for a dot confining six electrons clear signals of formation of a Wigner molecule.
II. METHOD AND CONVERGENCE

Consider \( N \) interacting electrons trapped in a circularly symmetric harmonic well \( V(r) = m^* \omega_0^2 r^2 / 2 \), where \( r^2 = x^2 + y^2 \). (In the quasi-two-dimensional limit one assumes that the confinement in \( z\)-direction is much stronger than in the \( x-y \)-plane. Then, only the lowest subband in \( z \)-direction is populated.) We write for the Hamiltonian

\[
H = \sum_{i=1}^{N} \left[ \frac{-\hbar^2}{2m^*} \nabla_i^2 + V(r_i) \right] + \sum_{\sigma} \frac{e^2}{4\pi\varepsilon_0 \varepsilon} \sum_{ij} \frac{1}{| \mathbf{r}_i - \mathbf{r}_j |}.
\]

Here, \( m^* \) and \( \varepsilon \) are the effective mass and the dielectric constant. The calculations are done for different values of the density parameter \( r_s \) which determines the average particle density in the dot, \( n_0 = 1/(\pi r_s^2) \). The latter is approximated by setting the oscillator parameter \( \omega_0^2 = e^2/(4\pi\varepsilon_0 \varepsilon m^* r_s^2) \). Throughout this paper we use effective atomic units in which the length unit \( \hbar \) is given in effective Hartree, \( \text{Ha}\). For GaAs these parameters are the closest in energy the larger the particle number

For a quantum dot confining \( N = 6 \) electrons at a density corresponding to \( r_s = 4 a_B^2 \) (the largest value of \( r_s \) we found accessible within the calculational scheme used here), Figure 2 shows the convergence of the many-body spectra as a function of the cut-off energy \( E_c/(\hbar \omega) \). Shown are the 6 lowest states with \( L = 0 \), together with the spin \( S \) of each state.

The ground-state energy for zero angular momentum is \( E_0 = 3.049 \text{ Ha}^* \) for \( 22\hbar \omega \) and \( E_0 = 3.045 \text{ Ha}^* \) for \( 24\hbar \omega \). An extrapolation to infinite cut-off energy can be made by plotting the total eigenvector is determined by calculating the expectation value of the \( S^2 \) operator.

As mentioned above, setting up the Hamiltonian matrix from chosen Fock states and subsequent diagonalization only in principle yields an exact solution of the many-body problem. For reasons of numerical feasibility it is necessary to truncate the set of basis functions to be used in the diagonalization. One then has to make sure that convergence of the spectra is reached with respect to the cut-off. As the required matrix size increases rapidly with \( N \), computational expenses severely restrict the calculations to only the smallest systems at not too large values of \( r_s \). Thus, with increasing electron number or \( r_s \), the results become less accurate due to the restricted number of basis states that can be included in the calculations. (The fact that the ground and excited states are the closer in energy the larger the particle number \( N \) imposes an additional difficulty.)

![FIG. 1. Convergence of the many-body spectra of \( N = 6 \) electrons in a harmonic trap at a density corresponding to \( r_s = 4 a_B^2 \) as a function of the cut-off energy \( E_c/(\hbar \omega) \). Shown are the 6 lowest states with \( L = 0 \), together with the spin \( S \) of each state.](image-url)
energy as a function of \((E_c - 10\hbar\omega)^{-3/2}\) \[3\]. This gives the estimate 3.043 Ha* for the fully converged results at \(r_s = 4 a_B^*\). For \(E_c = 14\hbar\omega\) a too small number of Slater determinants was included to build up the required correlations, such that the polarized \(S = 3\) state appeared as the ground state. (We identify a similar effect in unrestricted HF results mentioned above \[3\], where the single Slater determinant that is available incorrectly favors a spin-polarized ground state \[13\].) While for \(E_c = 22\hbar\omega\) the matrix dimension 44181 with 21448811 non-zero matrix elements is reasonably small, the value \(E_c = 24\hbar\omega\) already yields a matrix dimension 108375 with 67521121 non-zero matrix elements. As for larger densities the states are less correlated, a smaller number of Slater determinants is needed for an accurate description. Density parameters larger than \(r_s = 4 a_B^*\) or a higher number of particles than \(N = 6\) would go beyond the limits of numerical feasibility and accurate results could not be obtained.

III. MANY-BODY SPECTRA OF A SIX-ELECTRON QUANTUM DOT

We now analyze the many-body spectra and the sequence of spins for the low-lying states as the two-dimensional density parameter \(r_s\) is varied. We choose the particle number \(N = 6\) as it corresponds to the smallest dot size for which classically two stable crystalline structures co-exist: A pentagonal ring with one electron at the center, and a slightly distorted six-fold ring \[31,32\].

We fix the angular momentum to \(L = 0\) and show in Figure 2 the 50 lowest states for a quantum dot confining \(N = 6\) particles at density parameters between \(r_s = 1 a_B^*\) and \(r_s = 4 a_B^*\) (in steps of 0.5 \(a_B^*\)). To obtain a better resolution of the spectra the energies of the eigenstates \(\epsilon_i\) are scaled such that the energy difference between the ground state and 50th excited state equals one, i.e. plotted are the dimensionless quantities \(\tilde{\epsilon}_i = (\epsilon_i - \epsilon_1)/(\epsilon_{50} - \epsilon_1)\).

(The total energies of the ground state with \(S = 0\) and the excited state with \(S = 3\) are given in Table 1 below.) At a very large density corresponding to \(r_s = 1 a_B^*\), the ground state has spin \(S = 0\) and is separated from the lowest excited state with spin \(S = 2\) by a gap of 0.49 Ha*. This state is followed by a state with \(S = 1\) and again another (excited) spin singlet. The lowest fully polarized state is only found at a fairly high energy, the energy difference to the \(S = 0\) ground state being about 0.95 Ha*.

We now analyze the many-body spectra and the sequence of spins for the low-lying states as the two-dimensional density parameter \(r_s\) is varied. We chose for \(N = 6\) at \(r_s = 1.73 a_B^*\) we obtained excellent agreement of the \(S = 0\) ground state with the result of Pederva et al. \[19\]. As \(r_s\) is increased, the fully polarized \(S = 3\) state moves down in energy. At \(r_s = 2.5 a_B^*\) it has passed the excited singlet, but is still far from competing with the nonpolarized \(S = 0\) ground state. From the evolution of the energy difference of the \(S = 3\) state to the ground state, we do not expect any crossing of the polarized state and the ground state unless \(r_s\) becomes much larger than \(4 a_B^*\). Estimating the decrease in energy of the polarized state with respect to the ground state as \(r_s\) is increased, our data seem to support the result of Egger et al. \[18\] that the ground state of the 6 electron dot is not polarized for \(r_s\)-values smaller than about \(8 a_B^*\).

![Figure 2](image_url)

**Figure 2.** Spectra for angular momentum \(L = 0\) and six electrons in a harmonic trap at densities corresponding to Wigner-Seitz radii between \(r_s = 1 a_B^*\) and \(r_s = 4 a_B^*\) in steps of 0.5 \(a_B^*\). The square and the triangle show the first excited singlet state and the lowest fully polarized state, respectively. The lowest state has always \(S = 0\).

The energy axis is scaled such that the energy difference between the ground state \(\epsilon_1\) and 50th excited state \(\epsilon_{50}\) equals one, i.e. plotted are the dimensionless quantities \(\tilde{\epsilon}_i = (\epsilon_i - \epsilon_1)/(\epsilon_{50} - \epsilon_1)\).

Table 1 compares the total energies of the ground state with spin zero and the lowest polarized state with spin \(S = 3\) with the corresponding result obtained from DFT.

| \(r_s [a_B^*]\) | paramagnetic \text{ exact} | LSDA \text{ exact} | ferromagnetic \text{ exact} | LSDA |
|-----------------|--------------------------|----------------|--------------------------|------|
| 1.0             | 14.27                    | 14.30          | 15.22                    | 15.30|
| 1.5             | 8.983                    | 8.988          | 9.363                    | 9.409|
| 2.0             | 6.508                    | 6.503          | 6.695                    | 6.724|
| 2.5             | 5.084                    | 5.073          | 5.188                    | 5.204|
| 3.0             | 4.162                    | 4.148          | 4.225                    | 4.233|
| 3.5             | 3.519                    | 3.502          | 3.559                    | 3.560|
| 4.0             | 3.045                    | 3.027          | 3.071                    | 3.068|

**TABLE 1:** Table of energies [Ha*] for the paramagnetic \((S = 0)\) and ferromagnetic \((S = 3)\) states in a 6-electron quantum dot for different densities. In the diagonalization we used \(E_c = 22\hbar\omega\) for \(r_s \leq 3.5 a_B^*\) and \(E_c = 24\hbar\omega\) for \(r_s = 4 a_B^*\). For comparison the energies obtained with the local spin density approximation (LSDA) are also shown.
where the exchange-correlation part of the electron-electron interactions is treated in LSDA. For the DFT results we used an interpolation formula for the Tanatar-Ceperley [23] exchange-correlation energy. We refer to [7] for the further details concerning the numerical method. The CI energies of the $S = 0$ ground states and $S = 3$ isomer compare well with the LSDA results. For the paramagnetic case, the LSDA gives lower energies than the exact results, when $r_s \geq 2a_B^*$. This might be mainly due to the fact that the Tanatar-Ceperley interpolation formula slightly overestimates the correlation energy. Nevertheless, the LSDA gives surprisingly accurately the energy difference between the fully polarized ($S = 3$) and the paramagnetic ($S = 0$) state, as seen in Fig. 3. (For comparison, we also show the results for the infinite electron gas.)

\[ E \left[ \text{[ Ha]} \right] \quad S \quad L \quad E \left[ \text{[ Ha]} \right] \quad S \quad L \]

| $r_s = 3a_B^*$ | $r_s = 4a_B^*$ |
|----------------|----------------|
| $4.162$ | $0$ | $0$ | $3.046$ | $0$ | $0$ |
| $4.183$ | $1$ | $1$ | $3.054$ | $1$ | $1$ |
| $4.194$ | $1$ | $3$ | $3.060$ | $2$ | $0$ |
| $4.196$ | $2$ | $0$ | $3.062$ | $1$ | $3$ |
| $4.201$ | $1$ | $1$ | $3.063$ | $1$ | $1$ |
| $4.205$ | $0$ | $1$ | $3.065$ | $0$ | $1$ |
| $4.209$ | $1$ | $0$ | $3.066$ | $1$ | $0$ |
| $4.209$ | $0$ | $2$ | $3.068$ | $2$ | $1$ |
| $4.209$ | $0$ | $3$ | $3.068$ | $0$ | $2$ |
| $4.213$ | $1$ | $2$ | $3.070$ | $1$ | $2$ |
| $4.216$ | $2$ | $1$ | $3.070$ | $0$ | $3$ |
| $4.216$ | $2$ | $2$ | $3.071$ | $3$ | $0$ |
| $4.225$ | $3$ | $0$ |

\[ \begin{align*}
S &\quad L &\quad E \left[ \text{[ Ha]} \right] &\quad S &\quad L \\
4.162 & 0 & 0 & 3.046 & 0 & 0 \\
4.183 & 1 & 1 & 3.054 & 1 & 1 \\
4.194 & 1 & 3 & 3.060 & 2 & 0 \\
4.196 & 2 & 0 & 3.062 & 1 & 3 \\
4.201 & 1 & 1 & 3.063 & 1 & 1 \\
4.205 & 0 & 1 & 3.065 & 0 & 1 \\
4.209 & 1 & 0 & 3.066 & 1 & 0 \\
4.209 & 0 & 2 & 3.068 & 2 & 1 \\
4.209 & 0 & 3 & 3.068 & 0 & 2 \\
4.213 & 1 & 2 & 3.070 & 1 & 2 \\
4.216 & 2 & 1 & 3.070 & 0 & 3 \\
4.216 & 2 & 2 & 3.071 & 3 & 0 \\
4.225 & 3 & 0 \\
\end{align*} \]

**TABLE 2:** Table of energies [Ha*], spins $S$ and angular momenta $L$ of all levels up to the lowest ferromagnetic state for $r_s = 3a_B^*$ (left) and $r_s = 4a_B^*$ (right).

\[ \begin{align*}
S &\quad L &\quad E \left[ \text{[ Ha]} \right] &\quad S &\quad L \\
4.162 & 0 & 0 & 3.046 & 0 & 0 \\
4.183 & 1 & 1 & 3.054 & 1 & 1 \\
4.194 & 1 & 3 & 3.060 & 2 & 0 \\
4.196 & 2 & 0 & 3.062 & 1 & 3 \\
4.201 & 1 & 1 & 3.063 & 1 & 1 \\
4.205 & 0 & 1 & 3.065 & 0 & 1 \\
4.209 & 1 & 0 & 3.066 & 1 & 0 \\
4.209 & 0 & 2 & 3.068 & 2 & 1 \\
4.209 & 0 & 3 & 3.068 & 0 & 2 \\
4.213 & 1 & 2 & 3.070 & 1 & 2 \\
4.216 & 2 & 1 & 3.070 & 0 & 3 \\
4.216 & 2 & 2 & 3.071 & 3 & 0 \\
4.225 & 3 & 0 \\
\end{align*} \]

**IV. CHARGE DENSITIES AND PAIR CORRELATION**

For $r_s = 4a_B^*$ the radial densities of the $S = 0$ ground state and the 3rd excited state of $L = 0$, which is the lowest fully polarized state with spin $S = 3$, are shown in Figure 4. The azimuthally symmetric charge density for the polarized case shows a clear maximum at the center surrounded by an outer ring of lower density. In the paramagnetic case, the density profile is more smooth, the maximum density being at about $r \approx 6a_B^*$. The LSDA result shows a clear minimum at the origin, while the exact result has a larger density at the center. For comparison with the results of Egger et al. [18] we also show the density $\rho(r)$ multiplied by a factor $2\pi r$ (cf. lower panel of Fig. 4). For the polarized state, the maximum at the center is now seen as a clear shoulder in the density profile. Note that this is missing in the paramagnetic ground state density. (This is in disagreement with the results of Egger et al. [18] who found for the ground state a density profile with a clear shoulder as in the polarized case.) The azimuthal averages of the density profiles qualitatively have similarities with the broken symmetry solutions of the unrestricted HF [9] which for the paramagnetic case results in a ring of six electrons, and for the ferromagnetic case (ground state in HF) a ring of five electrons with one electron in the center. However, the localization of the electrons is largely exaggerated in HF. Opposite to HF, the LSDA correctly gives the paramagnetic state as the ground state, and its density profile resembles the exact result. LSDA does not break the azimuthal symmetry until $r_s > 8a_B^*$ when spin- or charge density wave-like states can occur [7]. Purely classical Monte Carlo [32,31] computations have shown that for $N < 6$ the charges are distributed on the perimeter of the dot, and none of
the particles occupies the dot center. This changes for \( N = 6 \), where the charge distribution with lowest energy consists of five electrons sitting on a ring, with the remaining electron occupying the center of the dot. This configuration is labeled by \((5,1)\). If all 6 particles are arranged on the dot perimeter (labeled by \((6,0)\)), the classical state is stable but has a higher energy than the \((5,1)\)-configuration.

FIG. 4. Charge density of a dot confining \( N = 6 \) electrons in a harmonic trap at \( r_s = 4 \, a_B^2 \); the exact result (solid line) is compared to the LSDA result (dashed line). Shown is the density \( n(r) \) (upper panel) and \( 2\pi n(r) \) multiplied by \( 2\pi r \) (lower panel) for the paramagnetic \( S = 0 \) state (left, \( ↑↓ \)) and the ferromagnetic \( S = 3 \) state (right, \( ↑↑ \)). The \( S = 3 \) state is separated from the \( S = 0 \) ground state by 0.026 \( \text{Ha}^* \).

The classical charge distribution can be arbitrarily oriented. The density from the CI solution, however, must be circularly symmetric. For an azimuthal average of the \((5,1)\)-pentagon structure, one would expect a pronounced maximum of the electron density in the center, and a less pronounced maximum at the dot radius. Correspondingly, the \((6,0)\) configuration should correspond to a minimum of charge density in the center and a maximum at finite radius. A first comparison of exact diagonalization calculations with the results of the mean-field approximation was given by Pfannkuche et al. \[13\] for "quantum dot helium", i.e., quantum dots containing only two electrons. They found from a comparison of exact diagonalizations with Hartree- and Hartree-Fock results that the exchange and correlation contributions are crucial. While the triplet state showed a reasonable agreement between the exact result and HF, the singlet could not be well reproduced. As mentioned above, Yannouleas and Landman \[7\] reported that in geometrically unrestricted HF at a density corresponding to \( r_s \approx 3.5 \, a_B^2 \) the \( N = 6 \) ground state is polarized and shows enhanced localization in the charge density. This \( S = 3 \) state exhibits the same geometry than the classical distribution of 6 electrons in a harmonic well: 5 particles are equidistantly localized on the perimeter of the dot, and the 6th particle is trapped in the center of the harmonic well. The non-polarized \( S = 0 \) state corresponding to the \((6,0)\)-configuration is about 0.034 \( \text{Ha}^* \) higher in energy. The exact diagonalization results described above do not support these HF results. Although being limited to small \( r_s \) values due to the necessary restrictions of the basis set, the systematic evolution and energy sequence of CI energies and densities shown in Figs. 3 and 5 seems to indicate that polarization as well as formation of Wigner molecules in circularly symmetric, parabolic wells would be impossible at densities as large as predicted by HF \[8\]. The geometrically unrestricted solution of the Kohn-Sham equations of \[7\] tend to overestimate the \( r_s \)-value at which spontaneously broken spin- or charge-symmetries can occur in the internal structure of the wave function \[33\]. Although calculated in a geometrically unrestricted DFT scheme, the fully converged LSDA densities for \( r_s \leq 4a_B^2 \) shown in Fig. 4 are azimuthally symmetric. Although LSDA suffers from the self-interaction problem, at the densities in question the results are in better agreement with CI studies than the unrestricted HF results.

FIG. 5. Pair correlation functions \( g_{\uparrow\uparrow}(\phi) \) calculated at the outer maxima of the density distribution (cf. upper panel in Fig. 4) as a function of the angle \( \phi \) between the electrons for the ground state (lower panel) and the excited polarized state with \( S = 3 \) (upper panel). The insets show schematic pictures of the electron configurations.

It is finally of interest to study how the spin and spatial symmetry in the internal structure of the wave function can be recognized in the pair correlation function

\[
g_{\uparrow\uparrow}(\phi) = \langle \hat{n}_{\uparrow}(r,0)\hat{n}_{\uparrow}(r,\phi) \rangle ,
\]

(2)
which describes the probability to find another (spin up or down) particle if a particle with spin up is placed at \((r, 0)\). Here \(r\) is the radius of maximum density and \(\varphi\) the angle between the electrons. Figure 3 shows \(g_{\uparrow\uparrow}(\varphi)\) and \(g_{\uparrow\uparrow}(\varphi)\) for the ground state (lower panel) and \(g_{\uparrow\uparrow}(\varphi)\) for the excited polarized state with \(S = 3\) (upper panel). From the \(\varphi\)-values of the maxima in \(g_{\uparrow\uparrow}(\varphi)\) one clearly concludes that the \(S = 0\) state has 6-fold symmetry with antiferromagnetic spin ordering, whereas the fully polarized case shows four maxima, corresponding to a five-fold symmetry. These intrinsic symmetries are in qualitative agreement with the unrestricted HF results, although the crystallization predicted by HF does not yet occur.

V. CONCLUSIONS

We commented on the recent conjecture that Wigner molecules would form in quantum dots at rather large electron densities \(r_s \gtrsim 3.5 a_B^*\) [13]. Our results are essentially exact up to \(r_s \leq 4a_B^*\) for six confined particles. The many-body spectra, densities and pair correlations obtained for \(N = 6\) clearly illustrate that the onset of formation of Wigner molecules and in particular, polarization of the ground state should be expected at much higher \(r_s\)-values than anticipated from unrestricted HF [3]. The critical density at which such a transition occurs does, in fact, strongly depend also on geometry [20] and on the number of confined particles at fixed average electron density in the dot. For the six-electron dot at \(r_s \approx 4a_B^*\) in question [3], the “exact” ground-state clearly prefers \(S = 0\) and shows antiferromagnetic order in the pair correlation. The polarized state with \(C_{5v}\)-symmetry is clearly higher in energy than the \(S = 1\) state, which would also be a candidate for the (5, 1) ground state configuration [23]. In addition, for values below \(r_s = 4 a_B^*\) we did not find clear signals of rotational structure in the spectra for non-zero angular momenta that would indicate a crystallized ground state. We note that the situation is different for \(N < 5\), where for densities as large as \(r_s = 2a_B^*\) the low-lying states could be well understood by assuming a square-shaped (4, 0) Wigner molecule for the internal structure of the wave function and analyzing its rotational structure. This also became clear when comparing the low-energy spectrum of a Heisenberg model with four electrons on a square [23]. For \(N > 5\), however, this simple picture does seem to not hold, as our results for \(N = 6\) clearly point out. The ground-state energies and densities obtained by density functional calculations in the local spin density approximation agree rather well with the results of exact diagonalizations, even though this comparison was restricted to a small particle number where the accuracy of the local density approximation is questionable. This gives some confidence that the method is well suited for describing the ground state electronic structures for larger sizes.

This work was supported by the Academy of Finland, the “Bayerische Staatsministerium für Wissenschaft, Forschung und Kunst” and the TMR programme of the European Community under contract ERBFM-BICT972405.

[1] R. Ashoori, Nature 379, 413 (1996).
[2] L. P. Kouwenhoven et al., Electron transport in quantum dots, in Proceedings of the Advanced Study Institute on Mesoscopic Electron Transport, ed. L. L. Sohn, L. P. Kouwenhoven and G. Schön (1997).
[3] S. Tarucha, D. G. Austing, T. Honda, R. J. van der Hage, and L. P. Kouwenhoven, Phys. Rev. Lett. 77, 3613 (1996).
[4] H.-M. Müller and S.E. Koonin, Phys. Rev. B 54, 14532 (1996).
[5] C. Yannouleas and U. Landman, Phys. Rev. Lett. 82, 5325 (1999).
[6] M. Stopa, Phys. Rev. B 54, 13767 (1996).
[7] M. Koskinen, M. Manninen and S.M. Reimann, Phys. Rev. Lett. 79, 1817 (1997); S.M. Reimann, M. Koskinen and M. Manninen, Phys. Rev. B 59, 1613 (1999).
[8] O. Steffens, U. Rössler and M. Sührke, Europhys. Lett. 42, 529 (1998).
[9] I.-H. Lee, V. Rao, R.M. Martin and J.-P. Leburton, Phys. Rev. B 57, 9035 (1998); I.-H. Lee, H.-H. Ahn, Y.-H. Kim and R.M. Martin, Phys. Rev. B 56, 13720 (1999).
[10] S.M. Reimann, M. Koskinen, M. Manninen and B. Mottelson, Phys. Rev. Lett. 83, 3270 (1999).
[11] D.G. Austing, S.Sasaki, S. Tarucha, S.M. Reimann, M. Koskinen and M. Manninen, Phys. Rev. B 60, 11514 (1999); S.M. Reimann, J. Kolehmainen, M. Koskinen, M. Manninen, D.G. Austing and S. Tarucha, Europ. J. Phys. D 9, 105 (1999).
[12] K. Hirose and N. Wingreen, Phys. Rev. B59, 4604 (1999).
[13] D. Pfannkuche, V. Gudmundsson, and P.A. Maksym, Phys. Rev. B 47, 2244 (1993).
[14] see for example P.A. Maksym and T. Chakraborty, Phys. Rev. Lett. 65, 108 (1990); U. Merkt, J. Huser and M. Wagner, Phys. Rev. B 43, 7320 (1991); P. Hawrylak, Phys. Rev. Lett. 71, 3347 (1993); A. H. MacDonald and M. D. Johnson, Phys. Rev. Lett. 70, 3107 (1993); S.-R. E. Yang, A.H. MacDonald, and M. D. Johnson, Phys. Rev. Lett. 71, 3194 (1993); J.J. Palacios et al., Phys. Rev. B 50, 5760 (1994); P.A. Maksym, Phys. Rev. B 53, 10871 (1996) and T. Ezaki, N. Mori and C. Hamaguchi, Phys. Rev. B 56, 6428 (1997), to mention only a few.
[15] R. B. Laughlin, Phys. Rev. B 27, 3383 (1983).
[16] S. M. Girvin and T. Jach, Phys. Rev. B 28, 4506 (1983).
[17] F. Bolton, Phys. Rev. B 54, 4780 (1996).
[18] R. Egger, W. Häsler, C.H. Mak and H. Grabert, Phys. Rev. Lett. 82, 3320 (1999).
[19] F. Pederiva, C.J. Umrigar and E. Lipparini, cond-mat/9912166.
[20] A. Harju, V.A. Sverdlov, R.M. Nieminen and V. Halonen, Phys. Rev. B59, 5622 (1999).
[21] E.P. Wigner, Phys. Rev. 46, 1002 (1934).
[22] F. Rapisarda and G. Senatore, Aust. J. Phys. 49, 161 (1996).
[23] B. Tanatar and D. M. Ceperley, Phys. Rev. B 39, 5005 (1989).
[24] D.M. Ceperley and B. J Alder, Phys. Rev. Lett. 45, 566 (1980).
[25] S. T. Chui and B. Tanatar, Phys. Rev. Lett. 74, 458 (1995).
[26] C.E. Creffield, W. Häusler, J.H. Jefferson, S. Sarkar, Phys. Rev. B 59, 10719 (1999).
[27] C.H. Mak, R. Egger and H. Weber-Gottschick, Phys. Rev. Lett. 81, 4533 (1998); C.H. Mak and R. Egger, J. Chem. Phys. 110, 12 (1999).
[28] M. Weissbluth, Atoms and Molecules, Academic Press, N.Y. (1978).
[29] R. B. Lehoucq, D.C. Sørensen and Y. Yang, ARPACK User’s guide: Solution to large scale eigenvalue problems with implicitly restarted Arnoldi methods. See http://www.caam.rice.edu/software/ARPACK
[30] M. Koskinen, M. Manninen and P.O. Lipas, Phys. Rev. B 49, 8418 (1994).
[31] F. Bolton and U. Rössler, Superlatt. Microstruct. 13, 139 (1992).
[32] V.M. Bedanov and F.M. Peeters, Phys. Rev. B 49, 2667 (1994).
[33] M. Koskinen, M. Manninen, B. Mottelson and S.M. Reimann, to be published.