Transitions between electron-molecule states in electrostatic quantum dots

P. A. Maksym
Department of Physics and Astronomy, University of Leicester, Leicester LE1 7RH, UK

H. Aoki
Department of Physics, University of Tokyo, Hongo, Tokyo 113-0033, Japan

(Dated: December 21, 2021)

Intermediate spin states that occur in electrostatic dots in the magnetic field regime just beyond the maximum density droplet are investigated. The 5-electron system is studied with exact diagonalization and group theory. The results indicate that the intermediate spin states are mixed symmetry states with a superposition of 5- and 4-fold electron-molecule configurations. A superposition of 5- and 4-fold correlation functions is found to reproduce the exact mixed symmetry pair correlation function to around 2%.

PACS numbers: 73.63.Kv, 73.23.Hk

I. INTRODUCTION

Recently, Nishi et al [1] have identified the ground state quantum numbers of a few-electron quantum dot in the strong magnetic field regime where the filling factor, \( \nu \), is less than one. The device they studied is a vertical pillar dot [2] containing up to 5 electrons and they identified the quantum numbers by comparing experimental excitation spectra with those from an accurate theoretical model [3]. In the particular case of a 5-electron dot, the orbital angular momentum and spin, \( (L, S) \), for the maximum density droplet state [2] which occurs at \( \nu = 1 \) is \((10, 5/2)\). Beyond this, a sequence of transitions occurs as \( \nu \) decreases and the sequence of observed \((L, S)\) values is \((14, 3/2), (15, 5/2), (18, 3/2)\) and \((20, 5/2)\). However the work of Nishi et al only gives the ground state quantum numbers and further theoretical analysis is needed to gain insight into the form of the ground state. This is done in the present work.

The fact that the observed combinations of \( L \) and \( S \) are severely restricted suggests that the ground states have a molecular form [4]. That is, the electron ground state is localised around the classical equilibrium positions and corresponds to rotational and vibrational motion. This turns out to be the case for the fully spin-polarised states \((S = 5/2)\) but the intermediate spin states \((S = 3/2)\) are different. In this case, states centred on different classical minima are allowed to couple and the ground state is an unusual state of mixed symmetry. The evidence for this comes from the form of the ground state pair correlation functions which are calculated by exact diagonalization (section II) and from group theory which provides the link between the ground state quantum numbers and the point symmetry of the electron molecule (section III). In addition, it turns out that the pair correlation function of the mixed symmetry states is well-approximated by a superposition of pair correlation functions for each symmetry type (section III).

II. PAIR CORRELATION FUNCTIONS

The exact diagonalization calculations are performed with the same theoretical model that Nishi et al used to analyse their data [3]. The model includes the effect of electron-electron interactions, screening, finite dot thickness and image charges. Physically, this leads to a model in which electrons are confined in a parabolic potential, move in two dimensions and interact via a modified interaction. The confinement energy, \( \hbar \omega \), for the 5-electron dot is 4.88 meV. The electron ground states are found by diagonalization of the Hamiltonian in a Fock-Darwin basis that includes higher Landau levels and the numerical relative error in the ground state energies is about \( 3 \times 10^{-4} \). The calculations reproduce experimental addition energies to about 5% [1].

The pair correlation function is the probability of finding an electron of spin \( s \) at \( r \) given that there is one of spin \( s' \) at \( r_0 \):

\[
P_{ss'}(r, r_0) = \frac{(2\pi \lambda^2)^2}{N(N-1)} \sum_{i \neq j} \delta(r_i - r)\delta_{ss'}\delta(r_j - r_0)\delta_{ss'},
\]

where the angle brackets denote a quantum mechanical expectation value and \( N \) is the number of electrons [5]. \( \lambda \) is the length parameter for harmonic oscillator confinement in a magnetic field, \( \lambda^2 = \hbar/2\mu^*\Omega \) where \( \Omega^2 = \omega^2 + \omega_c^2 \) and \( \omega_c \) is the cyclotron frequency.

Spin summed pair correlation functions for the \((15, 5/2), (18, 3/2)\) and \((20, 5/2)\) states are shown in Fig. I. The white regions correspond to the largest values and each square corresponds to an area of \( 12\lambda \times 12\lambda \) centred on the origin. In each case the electron density has the form of a ring with a minimum in the centre of the dot. The fixed position \( r_0 \) is taken to be on \( x \)-axis, at the position of maximum density. The pair correlation functions for spin polarised states show pentagonal symmetry: 4 peaks on
FIG. 1: Spin summed pair correlation functions for \((L, S) = (15, 5/2)\) (left), \((18, 3/2)\) (centre) and \((20, 5/2)\) (right). The white spot indicates \(r_0\).

FIG. 2: Spin resolved pair correlation functions for \((L, S) = (18, 3/2)\): like spin (left), unlike spin (right).

a ring are clearly visible in the figure and these peaks together with \(r_0\) form a pentagon. In contrast, the structure around the ring in the intermediate spin case is significantly weaker than for the \((15, 5/2)\) and \((20, 5/2)\) cases. In addition, there is pronounced intensity in the centre and the value at the origin is about 3 times larger than for the \((20, 5/2)\) case.

Figure 2 shows spin resolved pair correlation functions for the \((18, 3/2)\) state. The like spin correlation function has a ring-like structure but the symmetry is not 5-fold because there is a broad ridge on the left side of the ring instead of two pronounced peaks. The unlike spin correlation function does have 4 distinct peaks on the ring but the peaks are superimposed on a plateau that extends into the centre of the correlation function, which suggests an antiferromagnetic correlation between outer and central sites.

In the strong magnetic field limit, the structure in the pair correlation functions tends to be associated with minima in the potential energy [5]. The global energy minimum configuration of 5 confined, interacting electrons has the form of a pentagon but there is also a local minimum about 0.5-1% higher in energy which has the form of a square with one electron at its centre. The form of the pair correlation functions for \((18, 3/2)\) is consistent with the idea that both configurations contribute to the quantum state. The broad ridge on the left side of the ring in the like spin correlation function is consistent with the overlap of peaks from 5- and 4-fold configurations and the relatively large value at the centre of the unlike spin correlation function is consistent with a contribution from the central electron in the 4-fold configuration.

III. MIXED SYMMETRY STATES

The allowed \((L, S)\) values for rotational-vibrational states localised around potential energy minima can be determined with group theory and this supports the idea that the \((18, 3/2)\) state is a mixture of symmetry types. The electron positions that minimise the energy have either 5- or 4-fold symmetry. When the confinement is harmonic, the centre of mass motion separates and the relative Hamiltonian can be expressed in the Eckart frame in which the rotational and vibrational motion is approximately decoupled [4]. The relative Hamiltonian is conveniently expressed in terms of \(2N - 3\) normal co-ordinates \(Q_i\) and one Euler angle \(\chi\). Before antisymmetrization, the eigenstates have the form

\[
\Psi = \psi_{CM} \exp(-iL_{RM}\chi) f_{L_{RM}, n_1...n_{2N-3}}(Q_1, ..., Q_{2N-3}) \psi_{spin}(S_z),
\]

(2)

where \(\psi_{CM}\) is the centre of mass eigenstate, the exponential factor is an eigenstate of relative angular momentum, \(f\) is a vibrational eigenstate, \(n_i\) is the number of quanta in the \(i\)th vibrational mode and \(\psi_{spin}\) is the spin state. \(L_{RM}\) is the relative angular momentum quantum number. For ground states the centre of mass angular momentum is 0 so \(L_{RM} = L\). The electron states are found by anti-symmetrizing \(\Psi\). Application of the antisymmetrization operator \(\hat{A}\) to \(\Psi\) either gives a valid electron state or zero, depending on whether \(L, S_z\) and \(n_i\) are compatible with the Pauli principle. For ground states \(n_i = 0\) so the antisymmetrization process gives the allowed values of \((L, S)\).
Although $\hat{A}$ generates a sum over $N!$ permutations, it is not necessary to consider the full permutation group to find the allowed $(L, S)$ values. It turns out that a cyclic permutation operating on $\Psi$ is equivalent to a rotation. This enables the $(L, S)$ values for a system with $m$-fold symmetry to be found by considering $C_m$, the subgroup of the permutation group that is isomorphic to the cyclic group of order $m$. The relevant procedure is outlined in references $4$ and $5$ and examples are given for the spin polarised case. A few additional points, relevant to the intermediate spin case, are summarised here. The $N$-electron spin functions used in the procedure are eigenfunctions of the cyclic permutations. For each value of $S_z$, the entire set of $N$-electron spin functions forms a basis for a reducible representation of $C_m$. Reduction of this representation gives the number of spin functions associated with each representation of $C_m$ at a given value of $S_z$. What is needed is the number of spin functions for each value of total spin $S$. This is found by an algorithm that depends on use of spin lowering operators. When $S_z$ takes its maximum value, $S_z^* = 5/2$ in the present case, the only possible value of $S$ is $S = S_z$. Application of the spin lowering operator to $|S, S_z\rangle = |S^*, S^*\rangle$ generates $|S^*, S^* - 1\rangle$. This enables all the states with $S = S^*, S_z = S^*$ and $S = S^*, S_z = S^* - 1$ to be eliminated. In the remaining states, all those that have $S_z = S^* - 1$ also have $S = S^* - 1$. Recursive application of the spin lowering operator together with consideration of rotational symmetry therefore enables the allowed $(L, S)$ values to be determined.

The group theoretical analysis shows that for states with $(L, S) = (5k, 5/2), (k:\text{ an integer})$, 5-fold ground states are allowed and 4-fold states are forbidden. Hence a ground state localised around the 5-fold global potential minimum is allowed. In contrast, both 5-fold and 4-fold states are allowed at $(4k + 2, 3/2)$. The 5-fold energy minimum still favours a 5-fold state but if the global minimum is close in energy to the local 4-fold minimum and the coupling between the 5- and 4-fold states is sufficiently strong the ground state is expected to be a mixture of the two symmetry types. The form of the pair correlation functions suggests this is indeed happening.

To test this idea quantitatively, the pair correlation function for a superposition of 5- and 4-fold states is computed. The $(18, 3/2)$ state is taken to have the form $\alpha|5\rangle + \beta|4\rangle$ where $|m\rangle$ is a $m$-fold state. The pair correlation function is $\langle\alpha^2|\langle 5|\hat{P}|5\rangle + \beta|^2\langle 4|\hat{P}|4\rangle + 29\alpha^*\beta\langle 5|\hat{P}|4\rangle\rangle$, where $\hat{P}$ is the operator in Eq. (1). If the cross term is assumed to be small this simplifies to $p|\langle 5\rangle\hat{P}|5\rangle + (1-p)|\langle 4\rangle\hat{P}|4\rangle$ where $p$ is the probability of finding the system in the 5-fold state. The 5- and 4-fold states are taken to be $(20, 5/2)$ and $(18, 5/2)$, the states of pure symmetry that are closest in energy to $(18, 3/2)$. Although the pure symmetry states have $S = 5/2$, the spatial factors in these states will be similar to those in $(18, 3/2)$ if Eq. (1) is a good approximation. $p$ is found by fitting to the exact $(18, 3/2)$ correlation function. $p = 0.72 \pm 0.01$ fits the exact function with an RMS error of $4.9 \times 10^{-4}$, about 2% of the typical peak height. The fact that the 5-fold component occurs with the largest probability is consistent with the 5-fold global minimum. Figure 3 shows the exact correlation function, together with the fitted one and the two components used for the fit. In each case $r_0$ is on the density maximum for $(18, 3/2)$. This differs from the density maximum position for $(18, 5/2)$ by about 10%. Consequently, the 4-fold component pair function has a small deviation from 4-fold symmetry. This contributes to the 2% error but it is clear that the main features of the exact correlation function are well reproduced in the fit.

The present analysis depends on the localised states, Eq. (1), being a good approximation. In other words, the electron molecule theory should describe physics. The theory is known to reproduce ground state energies to about 1 part in $10^4$ in the very strong field regime where the ground state angular momentum is large $4$ $5$ $6$. When $\nu \sim 1$, it is still accurate to a few %, sufficient for the present analysis.

IV. DISCUSSION

Both group theoretical analysis and the form of exact pair correlation functions suggest that the intermediate spin states that occur just beyond the maximum density droplet are mixed symmetry states in which two different point symmetries coexist. For systems with large numbers of electrons, the regime beyond the maximum density droplet has previously been described.
in terms of spin textured states [7]. Large systems can be divided into a core region and an edge region but in contrast, the present few-electron system is too small to have a core. Hence its states resemble those of a molecule rather than those of a bulk system and have the unique feature of mixed symmetry. The 5-electron system is the smallest for which competing potential minima occur, hence the smallest in which mixed symmetry states can occur. However competing minima are a common feature of potential landscapes so it is likely that similar states will occur just beyond the maximum density droplet in larger but still molecular systems.

We thank Y. Nishi for suggesting the fitting procedure.

[1] Y. Nishi, P. A. Maksym, D. G. Austing, T. Hatano, L. P. Kouwenhoven, H. Aoki and S. Tarucha, Phys. Rev. B, in press, cond-mat/0512239.
[2] L. P. Kouwenhoven, D. G. Austing and S. Tarucha, Repts. Prog. Phys. 64, 701 (2001).
[3] P. A. Maksym et al, in preparation.
[4] P. A. Maksym, H. Imamura, G. P. Mallon and H. Aoki, J. Phys. Condens. Mat., 12, R299 (2000).
[5] P. A. Maksym, Phys. Rev. B 53, 10871 (1996).
[6] H. Imamura, P. A. Maksym and H. Aoki, Physica B 249, 214 (1998).
[7] J. H. Oaknin, L. Martin-Moreno and C. Tejedor, Phys. Rev. B 54, 16850 (1996).