Rotational levels in quantum dots(*)

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Abstract. – Low-energy spectra of isotropic quantum dots are calculated in the regime of low electron densities where Coulomb interaction causes strong correlations. The earlier developed pocket state method is generalized to allow for continuous rotations. Detailed predictions are made for dots of shallow confinements and small particle numbers, including the occurrence of spin blockades in transport.

Much of our present understanding of small quantum dots, with observable discrete level structure [1,2], concentrates on the regime of relatively high carrier densities where the interaction and charging energy is comparable to the kinetic (Fermi) energy in magnitude [3–6]. Similar to real atoms effective single-particle orbitals establish a reasonable approximation to the electronic states. The spins follow from Hund's rule [5,6] which is a perturbative result though it accords well with experimental findings in small quantum dots at high particle densities [1].

At lower densities Coulomb interaction is expected to destroy this single-particle picture, leaving strongly correlated or even crystallized electrons with collective low-energy excitations. While in the homogeneous two-dimensional case r_s should exceed $r_c = 37$ to reach this regime [7] ($r_s = (\pi n_s)^{-1/2}$ measures the ratio between Coulomb and kinetic energy and is regulated by the two-dimensional carrier density n_s), disorder is predicted to reduce this value considerably to $r_c = 7.5$ [8]. An even more pronounced reduction of r_c in comparison with the homogeneous value is found for the transition into the "Wigner regime" in quantum dots [9,10]. Careful quantum Monte Carlo (QMC) studies based on the spin sensitivity of the density-density correlation function yielded $r_c = 4$ for parabolic quantum dots [10]. Experimentally, this regime has been addressed using capacitance spectroscopy [11] which only probes ground state energies. Non-linear transport behaviour [1,2,12,13] has not yet been investigated to detect the interesting correlation effects for the low-energy excitations.

Numerical investigations of the low-density regime, emphasizing the spin states of rotating three electron Wigner molecules, have been carried out for shallow parabolic dots [14]. Investigations for larger particle numbers have focussed on dots of low symmetry where corners in the

(*) Dedicated to Alfred Hüller on the occasion of his 60th birthday.

confining potential or impurities suppress zero modes to delocalize the charges in the Wigner regime by so that "pocket states" can be introduced [15], which are well suited to describe localized charges. The "pocket states" served as basis to map the spin-sensitive low-energy physics to the one of lattice models of the Hubbard form [16] that account for quantum correlations by hopping between nearest places. The applicability of this archetype for correlation phenomena has been demonstrated, *e.g.*, in quantum dots of polygonal geometry [9].

This mapping to a lattice model cannot be carried out straightforwardly if zero modes cause charge delocalization which by symmetry actually happens in most experimental quantum dots. They are fairly well described by an isotropic and in fact parabolic model [3, 17]

$$H = \sum_{i=1}^{N} \frac{\boldsymbol{p}_i^2}{2m^*} + V\,, \tag{1}$$

where

$$V = \frac{m^*}{2}\omega_0^2 \sum_{i=1}^{N} \boldsymbol{x}_i^2 + \sum_{i<j} \frac{e^2}{\kappa|\boldsymbol{x}_i - \boldsymbol{x}_j|}\,. \tag{2}$$

Here, the effective mass m^* and the dielectric constant κ are material parameters, and $\boldsymbol{x}_j$ ($\boldsymbol{p}_j$) are electron positions (momenta) in two dimensions. This model does not explicitly involve spin (as opposed to real atoms spin-orbit coupling is negligible in quantum dots) so that all of its eigenstates are simultaneously eigenstates to the square of the total spin $\widehat{S}^2$ to eigenvalues $S(S+1)$. The present work extends the pocket state method (PSM) to allow for rotational symmetry and compares with results obtained by QMC studies [10]. Being based on a recently developed multilevel blocking algorithm [18] to circumvent the infamous fermion sign problem, this QMC allows for high accuracy to resolve reliably even the low-energy spin structure at particle numbers significantly larger than those treatable by diagonalizations.

At low densities the charge carriers form a finite piece of an electron crystal [14], a Wigner molecule (WM), that might, classically [19], be arbitrarily oriented. Superposition of all of the azimuthal degeneracies leads to an isotropic charge density distribution, as required by the symmetry of (2) [20]. For analytical progress it is tempting to separate out the normal coordinate related with the overall rotation and with total angular momentum quantum numbers ℓ (in strictly harmonic confinements ℓ refers to the relative part of the Hamiltonian since the center-of-mass motion just adds integer multiples of ω_0 to all of the eigenvalues and does not affect the spin of any of the states [21, 22]). However, the remaining normal coordinates then would in general no longer describe identical quantum particles obeying Pauli's principle and Fermi (or Bose) statistics but they would correspond to linear combinations of such particles. Within the PSM it is crucial to know the result of particle permutations in order to assign eventually the correct total spins S to the eigenstates and eigenenergies [23].

Therefore, we treat all of the possible particle exchanges on equal footing, including discrete overall rotations of the WM if they correspond to particle permutations. It depends on the geometry of the WM whether rotations by $2\pi/p$ with $p > 1$ leave the electron places invariant so that the Pauli principle relates ℓ with S. Such a relationship is well known, for instance from the example of solid hydrogen H_2, where the even ℓ are necessarily $S = 0$ singlet states while the odd ℓ are $S = 1$ triplets (in this example the spins refer to the protons), the reason being the equivalence of rotations by 180 degrees with the exchange of two identical spin-half fermions. Other examples are discussed in [24].

The validity of the PSM requires that the spin-sensitive excitation energies Δ, to be calculated by this method, should be smaller than charge (plasmon) excitations [23]. In the absence of continuous symmetries this condition is easily fulfilled at small densities due to the almost

exponential decay of $\Delta \sim \exp[-\sqrt{r_s}]$. Plasmon energies decrease only according to a power law $\sim r_s^{-3/2}$ for Coulomb repulsions. With their faster decay $1/2I = (2\pi m^* \int_0^\infty \mathrm{d}r\ r^3 n(r))^{-1} \sim r_s^{-2}$ (depending on the radial charge density distribution $n(r)$, I is the moment of inertia) the total angular momentum excitations, however, still decay faster than the plasmons so that eventually the low-energy levels will follow only from electron interchanges among the places defining the WM [25], including overall rotations by $2\pi/p$, *i.e.* by processes permuting identical quantum particles [26].

From classical [19] as well as from quantum [10] Monte Carlo studies it is known that up to $N \leq 8$ Wigner molecules in the parabolic quantum dots are very symmetric: the electrons form one spatial shell ($N \leq 5$) so that $p = N$, or one electron occupies the center (*i.e.* $p = N - 1$). Here we focus on $N \leq 6$. The method can be generalized straightforwardly to larger N and more complicated geometries of the WM.

The transition amplitudes for all possible particle permutations constitute the entries t of the pocket state matrix [15]. In the classically forbidden cases t can be estimated within the WKB approximation as discussed in [16, 23]. The complete potential (2), including the interaction, goes into this estimate. Often the most important entries involve only two or three adjacent particles, as in quantum dots of polygonal shapes [9], which then determine the hopping terms in the equivalent Hubbard model. This is different for the zero modes: there a much larger number of particles can be involved into a certain permutational transition, such as a rotation by $2\pi/p$ in isotropic quantum dots. Corresponding entries t_R to the pocket state matrix are not of tunneling type and, therefore, not exponentially small. In those cases $t_R = -p^2/8\pi^2 I$ is fixed by the energy constant $1/2I$ for rotational excitations (I follows from $n(r)$).

This way all of the relevant entries to the pocket state matrix can be estimated. Its diagonalization yields eventually the complete set of low-energy eigenvalues. Advantage can be taken from the fact that pocket states constitute a faithful representation of the symmetric group S_N, so that diagonalization can be carried out analytically for small systems, $N \leq 4$, otherwise numerical help is required. Only irreducible representations $[N/2+S,\ N/2-S]$ are compatible with Pauli's principle for spin-half fermions [23, 27]. This fixes the spin S for each eigenvalue.

The entries $|t| \sim \mathrm{e}^{-\sqrt{r_s}}$ and $|t_R| \sim r_s^{-2}$ vary differently with the strength of the Coulomb interaction, so that the ratio t/t_R is a measure for the interaction strength. We use

$$y := \frac{1}{1 + t/t_R} > 0$$

ranging from $1/(1+(\pi^2/4)p)$, since $|2t|$ cannot exceed the Fermi energy in the non-interacting limit, up to unity at strong interactions, $y \to 1$.

Figure 1 shows the low-energy spectrum *vs.* y for $N = 3$. Our description is designed for evaluating *excitation* energies, *i.e.* the *differences* between the energies of different spin states. As expected for weak interactions ($y < 0.5$), the ground state is unpolarized [22]. A transition into the spin-polarized ground state $S = 3/2$, not found in earlier diagonalization studies, is seen above a certain interaction strength which for Coulomb interactions and GaAs parameters can be estimated to happen when $\omega_0 < 0.5\,\mathrm{meV}$ [10]. This result complies with the QMC studies and can also be seen when carefully examining fig. 1 of the study [14] of a large quantum dot. We would like to emphasize that this spin polarization is an exact consequence of correlations and not the result of the mean-field approximation or a magnetic field. In transport experiments, when contacting quantum dots with electron reservoirs, it should show up as a "spin blockade" [28], since the ground states of $N = 2$ and $N = 3$ in

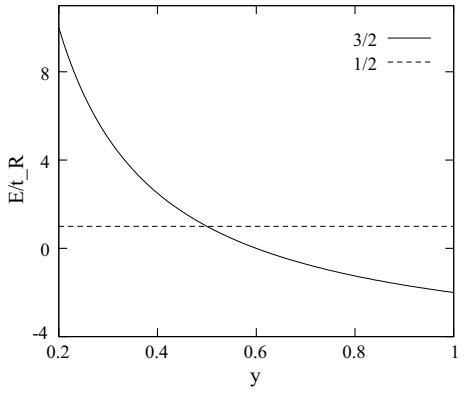

Fig. 1

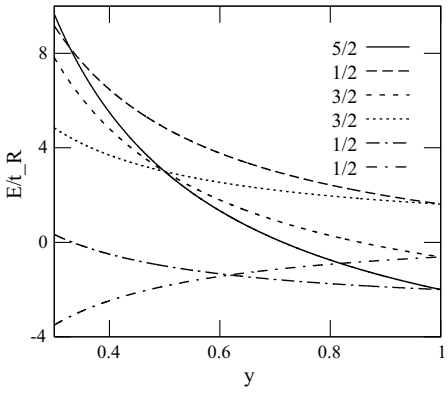

Fig. 2

Fig. 1 – Low energy levels using pocket states *vs.* y for $N = 3$ in units of t_{R}.

Fig. 2 – Low energy levels using pocket states *vs.* y for $N = 5$ in units of t_{R}.

sufficiently *large* quantum dots differ then in spin by more than $\Delta S = 1/2$ (by which entering or escaping single electrons can change spin) since the $N = 2$ ground state (with time reversal symmetry) is always a singlet [29].

For $N = 4$ (not shown here) we confirm the Hund's rule result of a $S = 1$ ground state, as obtained already in density functional calculations [5,6,20]. New is its persistence up to strong interactions. The lowest singlet level $S = 0$ approaches this ground level $\sim \exp[-\omega_0^{-1/3}]$ as ω_0 decreases. The rotationally first excited state $\ell = 1$ consists only of triplet $S = 1$ levels while the spin polarized level $S = 2$ belongs to the doubly excited rotational state, $\ell = 2$, together with another singlet $S = 0$ level.

For $N = 5$ (fig. 2), on the other hand, the polarized state $S = 5/2$ joins the unpolarized ground state $S = 1/2$ in the lowest rotational level at strong interactions. This low-energy high spin state makes negative differential conductances in the non-linear transport likely, due to the spin blockade [28]. Rotationally excited levels consist of $S = 1/2$ as well as of $S = 3/2$ spin states.

The sixth electron is predicted [10], also classically [19], to occupy the center of a 5-fold ring. This complicates the pocket state analysis since new types of pair exchanges appear (exchange with the central electron) and also the triple exchange t_3 (cyclic permutations of three adjacent electrons, including the central one) turns out as important, in accordance with WKB estimates [30]. Indeed, the PSM spectra do not compare with the low energy levels obtained from QMC unless t_3 is included with a similar magnitude as the pair exchanges.

This demonstrates how our approach complements most favorably the QMC simulations for quantum dots which yields abolute values for the many particle energies to high accuracy, contrary to the method based on pocket states. Very reliable estimates for the t-parameters can be achieved which otherwise would have to be guessed by less trusty approximative means. On the other hand, QMC is incomplete for the low energy levels since only the lowest eigenenergies to given z-component can be simulated.

For $N = 6$ and confining energies $\omega_0 \approx 0.13\,\mathrm{meV}$ (GaAs) we find, with increasing energy, the spin sequence 1-0-3-2-1-0-2-1-2-1-1-0. The spin $S = 1$ indicates another interaction-induced change in the ground state spin since from the non-interacting levels the point of view $N = 6$ corresponds to a "noble gas" configuration implying an unpolarized ground state

spin $S = 0$ [6]. This result also has to be contrasted with the conjecture $S = 2$ following from a static antiferromagnetic WM [16] of pentagonal symmetry. The rotational ground state $\ell = 0$ includes all possible spin states $S = 1, 0, 3, 2$, with the fully polarized state, $S = 3$, being lower in energy than the lowest $S = 2$ state, in accordance with QMC. This again suggests the possible occurrence of negative differential conductances for the transition to $N = 5$.

In conclusion, generalizing the pocket state method, we have developed a description for the low-density regime in isotropic such as parabolic quantum dots. Low energy levels, including spin quantum numbers were determined for $N \leq 6$. Detailed predictions are made for spin blockades as they should be detectable in linear and non-linear transport through shallow quantum dots of confinement energies below $\omega_0 \lesssim 0.4\,\mathrm{meV}$ (GaAs).

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I am particularly indebted to my teacher A. Hüller for long-standing support and encouragement. Numerous very fruitful discussions with Ch. Creffield, R. Egger, H. Grabert and J. Jefferson are acknowledged. This work has been carried out during stays at the University of Freiburg, the University of Jyväskylä, and the King's College London. Support has been received from the DFG (through SFB 276) and the EPSRC (UK).

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