Excitation spectra of two correlated electrons in a quantum dot

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Abstract. Measurements and a theoretical interpretation of the excitation spectrum of a two-electron quantum dot fabricated on a parabolic Ga[Al]As quantum well are reported. Experimentally, excited states are found beyond the well-known lowest singlet- and triplet states. These states can be reproduced in an exact diagonalization calculation of a parabolic dot with moderate in-plane anisotropy. The calculated spectra are in reasonable quantitative agreement with the measurement, and suggest that correlations between the electrons play a significant role in this system. Comparison of the exact results with the restricted Hartree–Fock and the generalized Heitler–London approach shows that the latter is more appropriate for this system because it can account for the spatial correlation of the electron states.

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Two-electron quantum dots (2eQDs) are the simplest man-made structures that allow to study the effects of electron–electron interaction including exchange and correlation. Pioneering experiments measuring the excitation spectrum of such systems with finite bias spectroscopy have been reported on vertical few-electron quantum dots [1, 2]. Measurements on lateral dots became possible [3, 4, 5] driven by the potential use of such systems for the implementation of qubits [6].

Here we present finite bias spectroscopy measurements performed on a system designed to have a *g*-factor close to zero and a small lateral anisotropy. We find that the magnetic field dependent excited state spectrum found experimentally can be closely reproduced by exact diagonalization calculations and is reasonably well described by a generalized Heitler–London (GHL) approach. The calculations suggest that electronic correlations are significant at low magnetic field, leading to spatially separated single-particle GHL orbitals, reminiscent of the formation of a Wigner molecule.

The dot is fabricated with electron-beam lithography defined top-gates on a 55 nm wide parabolic Ga[Al]As quantum well. The aluminium concentration varies in growth direction and was chosen such that the Zeemansplitting of electronic levels is negligible. The particular gate geometry of the dot (see Fig. 1, inset) leads to a moderate spatial anisotropy of the confinement potential. Further details about the sample can be found in Ref. [5].

Measurements were performed in a dilution refrigerator with an electronic temperature of 300 mK as determined from the width of conductance resonances in the Coulomb blockade regime. Finite bias I(V) traces were recorded using standard DC current measurement techniques. Later on the data was numerically differentiated to obtain the differential conductance dI/dV_{bias} .

The sample was tuned into the Coulomb-blockade regime in the region of the transitions between one and two, and two and three electrons on the dot. The electron number was confirmed by measurements of Coulomb-blockade diamonds [5] and by using the on-chip quantum point contact as a charge detector. In this region, the dot had a single-particle level spacing of 5 meV and a charging energy of 6.9 meV.

The magnetic field dependent excitation spectrum shown in 1 is measured employing tunneling spectroscopy with varying plunger gate voltage V_{pg} at a fixed bias voltage of 2.5 mV. We plot the derivative dI/dV_{pg} . Resonances in this quantity correspond to resonances in the differential conductance dI/dV_{bias} . Two families of resonances can be seen corresponding to transitions between electron numbers N = 1 and 2, and N = 2 and 3.

First we concentrate on the N = 1 and 2 region. At zero magnetic field we observe – in addition to the wellknown singlet ground state S_0 and triplet excited state T_+ that can be seen in Fig. 1 – an additional triplet state T_- split from the T_+ state by the confinement anisotropy (not seen in the figure, see [5]). At finite magnetic fields we find an additional excited singlet state S_2 (see Fig. 1) which shows an avoided crossing with S_0 at a magnetic field beyond the singlet–triplet transition (at 4 T) in the ground state of the system. Another excited state $T_{+,CM}$ even higher in energy than S_2 is found and attributed to

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FIGURE 1. Measured excited state spectra as a function of magnetic field and plunger gate voltage. Inset: sample geometry.

a triplet state combining an excitation in the relative and the center of mass motion (see below). In the transition region between N = 2 and 3 we observe resonances that correspond to transitions between the two-electron states S_0 and T_+ and the three-electron ground state.

The experimental findings can be quantitatively interpreted by comparing to the results of exact diagonalization (EXD) calculations for two electrons in an anisotropic harmonic confinement potential. Details of the model can be found in Ref. [5]. The calculated magnetic field dependent energy splitting $J(B) = E_{T+} - E_{S0}$ between the two lowest states S₀ and T₊ is found to be in remarkable agreement with the experiment. All the additional excited states observed in the experiment can be unambiguously identified with calculated excited states of the two-electron dot [5].

The EXD calculations give strong evidence for the importance of correlation effects. Closer inspection of the total electron densities and conditional probabilities (CPDs) reveals a strongly correlated ground state at zero magnetic field, in which the electrons do not occupy the same single-particle state. Already at magnetic fields below the singlet-triplet transition, the CPDs indicate localization of the two electrons and formation of a state resembling an H₂-like Wigner molecule.

We gain further insight into the importance of correlations in the system by comparing the results for J(B) calculated exactly and within different approximations. Figure 2 shows the predictions obtained from EXD calculations, restricted Hartree–Fock (RHF) calculations and



FIGURE 2. Comparison of J(B) calculated with different methods and the experimental results. Inset: single-particle orbitals (modulus square) of the GHL approach.

from a generalized Heitler–London (GHL) approach in comparison with the measured data. Details about the approximative calculations can be found in [7]. The RHF and GHL schemes have the advantage that they minimize the energy using single-particle orbitals. It is evident from Fig. 2 that the RHF method, which assumes that both electrons occupy the same single-particle orbital, is not able to reproduce the experimental findings. The GHL approach, which allows the two electrons to occupy two spatially separated states, appears to be a good approximation. Plotting the two single-particle orbitals resulting from this approach clearly demonstrates that the two electrons do not occupy the same spatial orbital, but rather fill single-particle states that are spatially separated significantly (inset).

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