Spin and interaction effects in quantum dots: a Hartree-Fock-Koopmans approach

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(submitted February 27, 2002)

We use a Hartree-Fock-Koopmans approach to study spin and interaction effects in a diffusive or chaotic quantum dot. In particular, we derive the statistics of the spacings between successive Coulomb-blockade peaks. We include fluctuations of the matrix elements of the two-body screened interaction, surface-charge potential, and confining potential to leading order in the inverse Thouless conductance. The calculated peak-spacing distribution is compared with experimental results.

The traditional description of Coulomb blockade in quantum dots has been the constant-interaction (CI) model, in which the electrons occupy single-particle levels in a confining potential, and the interaction is taken as a constant charging energy. In dots with chaotic dynamics, the fluctuations of the single-particle levels and wave functions can be described by random-matrix theory (RMT). The CI plus RMT model was successful in describing some of the observed statistical properties of such dots, e.g., the conductance peak-height distribution. However, several experiments have demonstrated that other statistics, such as the distribution of the spacings between Coulomb-blockade peaks, are affected by electron-electron interactions.

At low temperatures the peak spacing is given by the second-order difference of the ground-state energy versus the number of electrons. In the CI model and for spin-degenerate levels, the peak-spacing distribution is bimodal, i.e., a superposition of a $\delta$ function and a (shifted) Wigner-Dyson distribution. However, none of the measured distributions are bimodal and all deviate from Wigner-Dyson statistics.

In the spinless case, exact diagonalization for a small number of electrons [3], Hartree-Fock (HF) [4] and density functional [5] calculations, as well as a random interaction matrix model [6] explained the deviation from Wigner-Dyson statistics as an interaction effect. Spin degrees of freedom were included using exact diagonalization for dots with a small number of electrons and small values of the Thouless conductance $g$ [7,11,12]. Spin effects were studied in the limit $g \to \infty$ using the so-called universal Hamiltonian [14,15]. Interaction effects (in the presence of a magnetic field) were included using a Strutinsky approach for quantum dots [12]. The resulting distributions showed qualitative differences when compared with experiments. Temperature effects were shown to be important at temperatures as low as $T \sim 0.1 - 0.2 \Delta$ [13] ($\Delta$ is the mean spacing between spin-degenerate levels). The spacing statistics were also studied in a spin density functional theory for dots with $\sim 10$ electrons [10].

Here we develop a theory that includes spin and interaction effects and is based on a HF-Koopmans approach. The theory is generic and does not require the actual solution of the HF equations. Rather than using the non-interacting basis, we choose as a reference state the $n$-electron dot with spin $S = 0$ ($n$ even) and work in its HF basis. We then consider the addition energies and the energy differences between various spin configurations in Koopmans’ limit where the single-particle wave functions do not change [17]. Previously, Koopmans’ approach was discussed for dots with spinless electrons [7] and was used to study spectral scrambling [13]. Here we derive the generic statistics of the spin and peak spacings, assuming that the HF levels of the reference state satisfy statistics typical of a diffusive or chaotic dot. The theory is valid for large $g$, and for $g \to \infty$ it reduces to the universal Hamiltonian [14]. For finite $g$, we include fluctuations of the diagonal matrix elements to leading order in $1/g$. Compared with the approach of Ref. [12], our theory requires the statistics of only a few levels around the Fermi energy, and some of its results are qualitatively different.

The Hamiltonian of the quantum dot in the “disorder” basis $|i\sigma\rangle = a_{i\sigma}^\dagger|0\rangle$ ($i$ denotes a spatial orbit and $\sigma = \pm 1$ is the spin variable) is given by

$$H = \sum_{i\sigma} \epsilon_i^{(0)} a^\dagger_{i\sigma} a_{i\sigma} + \frac{1}{2} \sum_{ijkl\sigma\sigma'} v_{ijkl} a_{i\sigma}^\dagger a_{j\sigma'}^\dagger a_{l\sigma'} a_{k\sigma},$$

(1)

where $\epsilon_i^{(0)}$ are the single-particle energies and $v_{ijkl}$ are the (spin-independent) matrix elements of the Coulomb interaction. The Hamiltonian [10] can be solved in the HF approximation. For each value of the spin projection $S_z$, we use Slater determinants with $n_+$ ($n_-$) spin up (down) orbitals. Usually, the HF single-particle energies $\epsilon_{i\sigma}$ and orbitals $\phi_{i\sigma}$ depend on the spin $\sigma$. However for even $n$, the HF equations have a solution where $\epsilon_{i\sigma}$ and $\phi_{i\sigma}$ are independent of $\sigma$, and the lowest $n/2$ levels are doubly occupied. Such a Slater determinant has good $S = 0$. We choose this solution as our reference state, and work in its HF basis $|\alpha\rangle$. This $S = 0$ state is an eigenstate of the following diagonal many-particle Hamiltonian

$$H_d = \sum_{\alpha\sigma} \epsilon_{\alpha\sigma}^0 \hat{n}_{\alpha\sigma} + \frac{1}{2} \sum_{\alpha\beta} v^A_{\alpha\beta} \hat{n}_{\alpha\sigma} \hat{n}_{\beta\sigma} + \sum_{\alpha\beta} v_{\alpha\beta} \hat{n}_{\alpha\sigma} \hat{n}_{\beta\sigma},$$

(2)
where $\hat{n}_{\alpha \sigma}$ is the number operator of the state $\alpha \sigma$, and $v_{\alpha \beta} \equiv v_{\alpha \beta \alpha \beta}$, $v_{\alpha \beta}^{\text{ex}} \equiv v_{\alpha \beta \beta \alpha}$, and $v_{\alpha \beta}^{\text{ex}} \equiv v_{\alpha \beta} - v_{\alpha \beta}^{\text{ex}}$ are diagonal, exchange and antisymmetric matrix elements, respectively.

The Hamiltonian (3) also has eigenstates with $S \neq 0$. We label by $\alpha = 0$ the last occupied level of the $S = 0$ state. The lowest energy state for each spin $S$ is obtained by promoting $S$ spin down electrons from $\alpha = 0, \ldots, -(S-1)$ to spin up electrons in $\alpha = 1, \ldots, S$. The resulting Slater determinant describes a maximal spin projection state $S = S$ and has good spin $S$. Using (2), the energy difference $\delta E_n(S) \equiv E_n(S) - E_n(S = 0)$ between the lowest states with spin $S$ and spin $S = 0$ can be written in terms of $\epsilon_0^{(n)}$ and a few matrix elements. For example

$$\delta E_n(S = 1) = (\epsilon_1^{(n)} - \epsilon_0^{(n)}) - v_{10} .$$

(3)

The spin $S_n$ of the ground state of the $n$-electron dot is determined by minimizing $\delta E_n(S)$. Similarly, the ground-state spin $S_{n+1}$ of the dot with $n + 1$ electrons can be determined from the energy differences $\delta E_{n+1}(S) \equiv E_{n+1}(S) - E_{n+1}(S = 1/2)$ for half-integer $S$. Assuming Koopmans’ limit, we can express $\delta E_{n+1}(S)$ in terms of the HF levels and matrix elements of the reference state $(n, S = 0)$. For example

$$\delta E_{n+1}(S = 3/2) = (\epsilon_n^{(2)} - \epsilon_0^{(n)}) - v_{10} - v_{20} + v_{21}^A .$$

(4)

The addition energy $\mu_{n+1} \equiv E_{\text{gs}}(n + 1) - E_{\text{gs}}(n)$ is

$$\mu_{n+1} = \mu_{n+1}(0 \rightarrow 1/2) + \delta E_{n+1}(S_{n+1}) - \delta E_n(S_n) ,$$

(5)

where in Koopmans’ limit $\mu_{n+1}(0 \rightarrow 1/2) = \epsilon_1^{(n)}$.

The spacing $\Delta_2$ between successive peaks is given by the difference in addition energies. We have to distinguish between even-odd-even (“odd”) and odd-even-odd (“even”) transitions in particle number. We consider here the odd transition $n \rightarrow n + 1 \rightarrow n + 2$ (n even), for which $\Delta_2 = \mu_{n+2} - \mu_{n+1}$ (similar results can be derived for the even transition [19]). $\mu_{n+1}$ is given by (3) and $\mu_{n+2}$ is calculated from $\mu_{n+2} = \mu_{n+2} (1/2 \rightarrow 0) + \delta E_{n+2}(S_{n+2}) - \delta E_{n+1}(S_{n+1})$ with $\mu_{n+2} (1/2 \rightarrow 0) = \epsilon_1^{(n)} - v_{11}$. $\delta E_{n+1}(S)$ is given by, e.g., Eq. (3), while $\delta E_{n+2}(S)$ is calculated from, e.g.,

$$\delta E_{n+2}(S = 1) = (\epsilon_2^{(n)} - \epsilon_1^{(n)}) - v_{11} + v_{21}^A .$$

(6)

To describe the statistics of $\Delta_2$ it is necessary to model the fluctuations of the HF levels and wave functions of the $(n, S = 0)$ dot. The spectrum is assumed to follow RMT within $g$ levels around the Fermi energy, and the matrix elements are uncorrelated from the single-particle spectrum. An exception is the gap $\epsilon_1^{(n)} - \epsilon_0^{(n)}$. We find its statistics by comparing the single-particle spectrum of the $(n + 2)$-electron dot with the spectrum of the $n$-electron dot (both in their $S = 0$ configuration). We have the relation $\epsilon_1^{(n)} - \epsilon_0^{(n)} = \epsilon_1^{(n+2)} - \epsilon_0^{(n+2)} + v_{01}^A + v_{01} - v_{11}$, in which the levels $\epsilon_0^{(n+2)}$ and $\epsilon_1^{(n+2)}$ are both doubly occupied, and thus their spacing should follow Wigner-Dyson statistics. The gap distribution is then a convolution of a Wigner-Dyson distribution with a Gaussian describing the distribution of $v_{01}^A + v_{01} - v_{11}$ (see below).

We apply our HF-Koopmans approach in a restricted single-particle space of $\sim g$ levels around the Fermi energy. The long-range bare Coulomb interaction should then be replaced by an effective interaction. In the limit $r_s << 1$ we employ an effective RPA interaction calculated by excluding particle-hole transitions within the above strip of $\sim g$ levels [20]. This effective interaction is $v(r_1, r_2) = \epsilon^2/C + \nu_{nk}(r_1, r_2) + V(r_1) + V(r_2)$, where $\nu_{nk}(r_1, r_2)$ is a two-body screened interaction in the dot, and $V(r)$ is a one-body potential generated by the accumulation of surface charge in the finite dot [20].

We decompose the interaction in (2) into its average and fluctuating parts, and denote by $U_d = \bar{v}_{\alpha \beta}$ and $J_s = \bar{v}_{\alpha \beta}^{\text{ex}} (\alpha \neq \beta)$ the average values of the direct and exchange interaction, respectively. The average interaction is invariant under a change of the single-particle basis when $\bar{v}_{\alpha \alpha} = U_d + J_s$, and can be written in terms of the number operator $\hat{n}$ and total spin $S$ [21].

$$\bar{V} = \frac{1}{2} \left( U_d - \frac{J_s}{2} \right) \hat{n}^2 - \left( \frac{U_d}{2} - J_s \right) \hat{n} - J_s S^2 .$$

(7)

The average interaction is determined by the spatial correlations of the single-particle wave functions [20,22]. To leading order in $1/g$, $J_s = (\Delta/2)(1 + 2 - b_1/g)$ [23], where $b_1$ is a geometry-dependent coefficient determined from $b_1/g = \int dr \Pi(r, r)/A$. Here $\Pi(r_1, r_2)$ is the diffusion propagator and $A$ is the area of the dot.

FIG. 1. Statistics in the absence of fluctuations of the interaction matrix elements. The probabilities of various spin values $S$ are shown versus $J_s$. The solid (dashed) lines describe the orthogonal (unitary) symmetry. Inset: the standard deviation of the peak spacing $\sigma(\Delta_2)$ versus $J_s$ for the even, odd and combined (“total”) transitions. $J_s$ and $\sigma(\Delta_2)$ are measured in units of $\Delta$. Koopmans’ limit is exact when the fluctuations of the
matrix elements are ignored, and our calculations of $\Delta_2$ reduce to those in Ref. [11], i.e., they can be derived directly from an Hamiltonian that consists of a random one-body part plus an average interaction [11]. In the limit $g \to \infty$ this Hamiltonian is just the universal Hamiltonian with $J_s = \Delta/2$ [11]. A better estimate of $J_s$ can be obtained in RPA [11]; it increases monotonically with $r_s$ but remains below $\Delta/2$.

In the simple limit [11], the spin and spacing distributions are determined by a single parameter $J_s/\Delta$ [11]. This limit is demonstrated in Fig. 1 where we show the probabilities of various spin values versus $J_s$. The inset shows the standard deviation of $\Delta_2$ versus $J_s$ for the even and odd transitions as well as the combined one (“total”).

Next we discuss the fluctuations of the interaction matrix elements [20,21], which are approximately Gaussian variables. We discuss separately the bulk screened interaction and surface-charge potential. Using the diagrammatic approach for the two-body screened interaction, we have, for $r_s \ll 1$ and to leading order in $1/g$

\[
\text{GOE: } \sigma(v_{\alpha\beta}) = 2 \sigma_2; \quad \sigma(v_{\alpha\alpha}) = \sqrt{2} \sigma_2; \quad \sigma(v_{\alpha\beta}^e) = 2 \sqrt{2} \sigma_2
\]

\[
\text{GUE: } \sigma(v_{\alpha\beta}) = \sigma_2; \quad \sigma(v_{\alpha\beta}^e) = \sigma_2; \quad \sigma(v_{\alpha\alpha}) = \sqrt{2} \sigma_2 .
\]

Different matrix elements (including the direct $v_{\alpha\beta}$ and exchange $v_{\alpha\beta}^e$) are uncorrelated. We note that the coefficients of $\sigma_1$ and $\sigma_2$ in Eqs. (8) are different from those obtained for a zero-range interaction [22]. The parameter $\sigma_2$ is given by

\[
\sigma_2 = \left[ A^{-2} \int dr_1 \int dr_2 \Pi^2(r_1, r_2) \right]^{1/2} = c_2 \Delta / g ,
\]

where $c_2$ is a geometry-dependent coefficient. For a disk of radius $R$ and boundary conditions of vanishing normal derivative, we find [11]

\[
c_2 = 2 \left[ \sum_{l,m} x_{l,m}^{-4} \right]^{1/2} \approx 0.67
\]

where $x_{l,m}$ are the zeros of $J_l(x)$ ($J_l$ is the Bessel function of order $l$) and $g \Delta = 2 \pi \hbar D / R^2$ ($D$ being the diffusion constant). Since only a few matrix elements contribute to the peak spacing, the contribution of the two-body screened interaction is parametrically of the order $\Delta/g$, unlike the $\Delta/\sqrt{g}$ dependence found in Ref. [12].

The surface-charge contribution to an interaction matrix element is $v_{\alpha\beta} = V_\alpha + V_\beta$, where $V_\alpha \equiv \int |\psi_\alpha(r)|^2 V(r)$ is a diagonal matrix element of the surface-charge potential. We have

\[
\sigma(V_\alpha) = (2/\beta)^{1/2} \sigma_1 ; \quad V_\alpha V_\beta - V_\alpha V_\beta \approx 0 ,
\]

where

\[
\sigma_1 = \left[ A^{-2} \int dr_1 \int dr_2 V(r_1) \Pi(r_1, r_2) V(r_2) \right]^{1/2} = c_1 \Delta / \sqrt{g} .
\]

For an isolated two-dimensional (2D) circular disk of radius $R$, the surface-charge potential can be approximated by $V(r) = -(e^2 / 2 \kappa \varepsilon R)(R^2 - r^2)^{-1/2}$, where $\kappa = 2 \pi \varepsilon^2 \nu / \epsilon$ is the inverse screening length in 2D and $\epsilon$ is the dielectric constant [21]. We then find $c_1 = 2^{-1/2} \sum_{m \neq 0} \sin^2 (x_{0,m} / x_{0,0} J_1(x_{0,m}))^{1/2} \approx 0.087$ [12].

The above results can be generalized to a ballistic dot, using the ballistic supersymmetric $\sigma$ model obtained when a weak disorder with finite correlation length is added [23]. In particular, if the Lyapunov length of this smooth disorder is smaller than the dot’s size, relations similar to Eqs. (8) and (11) can be derived but with the ballistic propagator $\Pi_B$ replacing the diffusive propagator $\Pi$ in Eqs. (11) and (11). For a circular dot we define the ballistic Thouless conductance from the inverse time it takes to cross the diameter $2R$ of the dot, leading to $g = \pi \kappa F / (R^2) = \pi \nu (n/2)^{1/2}$ ($n$ is the number of electrons in the dot). In the ballistic case

\[
\sigma_2 = c_2 \Delta \left[ \ln(c'_2 g) \right]^{1/2} / g ,
\]

where $c_2 = \sqrt{3}/2$ is a geometry-independent constant. The direct propagation between $r_1$ and $r_2$ contributes to $\Pi_B(r_1, r_2)$ a term $1/(\pi \kappa F |r_1 - r_2|)$ which at shorter distances should be replaced by its quantum counterpart $J^2(k_F |r_1 - r_2|)$ [27]. The corresponding contribution needs to be renormalized such that its integral over $r_1$ (or $r_2$) vanishes [28]. The remaining part of $\Pi_B$ involves single or multiple scattering from the boundaries and calculating it requires knowledge of the semiclassical dynamics. It can be calculated analytically for a circular billiard with diffusive boundary scattering [23]. Using this model, we estimate $c'_2 = 0.81$ [13]. A similar estimate of (11) gives $c_1 = 0.123$.

We have studied the effects of fluctuations of the interaction matrix elements on the peak-spacing distribution. In Fig. 2 we show the standard deviation $\sigma(\Delta_2)$ versus $\sigma_1$ ($J_s = 0.3 \Delta$ and $\sigma_2 = 0.05 \Delta$) for the orthogonal (solid lines) and unitary (dashed lines) symmetries. $\sigma(\Delta_2)$ increases with $\sigma_1$ and it does so faster in the odd case.

![Fig. 2. The standard deviation $\sigma(\Delta_2)$ versus the standard deviation $\sigma_1$ of the surface-charge potential for $J_s = 0.3 \Delta$ and $\sigma_2 = 0.05 \Delta$. The solid and dashed lines describe the orthogonal (left) and unitary (right) symmetries, respectively.](image-url)
for $\sigma_2 = 0.025\Delta$ and $\sigma_1 = 0$, 0.03$\Delta$ and 0.06$\Delta$. Signatures of the bimodality can still be observed for $\sigma_1 = 0$ but they disappear at $\sigma_1 = 0.03\Delta$. Nevertheless, the distributions remain asymmetric (more so in the unitary case).

An additional contribution to the fluctuations of $\Delta_2$ arises from the variation of the gate voltage between peaks. In general, the change of the gate voltage between two peaks leads to a spatially non-uniform change $U(r) = -V(r) + \bar{V}(r)$ in the confining potential, where $\bar{V}$ originates in the mutual dot-gate capacitance [15]. This leads to scrambling of the HF levels between peaks. For example, let us consider the odd transition. As the gate voltage changes between $V_g^{n+1}$ and $V_g^{n+2}$, the reference HF levels $\epsilon^{(n)}_\alpha$ change by $\delta\epsilon^{(n)}_\alpha = U_\alpha = \int d\mathbf{r} |\psi_\alpha(r)|^2 U(r)$. We therefore substitute $\epsilon^{(n)}_\alpha \rightarrow \epsilon^{(n)}_\alpha + U_\alpha$ in the calculation of $\mu_{n+2}$ and in Eq. (3) ($\mu_{n+1}$ is unchanged and calculated from Eq. (2)). This level scrambling can also lead to spin rearrangement in the dot. The ground-state spin of the $n + 1$-electron dot at gate voltage $V_g^{n+2}$ (just below the transition) is found from (4) (and its generalization to higher values of $S$) after the substitution $\epsilon^{(n)}_\alpha \rightarrow \epsilon^{(n)}_\alpha + U_\alpha$.

The fluctuation properties of $U_\alpha$ are similar to those of $V_g$ in Eqs. (11), except that $V(r)$ is replaced by $U(r)$ in Eq. (11).

We now compare our theory with the experimental results of Ref. [3] at the lowest measured temperature of $T = 0.22\Delta$. At this temperature it is necessary to include the effect of excited states and in particular the contribution from both lowest $S = 0$ and $S = 1$ states [4]. We model the gate-voltage scrambling by a potential $\bar{V}$ whose matrix elements are uncorrelated from the matrix elements of $V$ and have the same variance. It is difficult to estimate $\sigma_1$ and $\sigma_2$ for the ballistic dot used in the experiment. The simple estimates based on a billiard with diffusive surface scattering (see the second paragraph after Eq. (3)) for $n \approx 340$ electrons (i.e., $g \approx 41$) give $\sigma_1 = 0.02\Delta$ and $\sigma_2 = 0.04\Delta$. Fig. 4 compares the experimental distribution of Ref. [3] in the presence of a magnetic field (solid histograms) with the corresponding theoretical distribution (dashed histograms) that includes an experimental noise of 0.1$\Delta$. The theoretical distribution describes rather well the asymmetry of the experimental distribution and its width 0.27$\Delta$ is slightly below the experimental width of (0.29 ± 0.03)$\Delta$. In chaotic billiards our estimates of $\sigma_1$ and $\sigma_2$ can be enhanced by up to a factor of 2 and lead to better agreement with the data.

![FIG. 4. Calculated peak-spacing distribution at $T = 0.22\Delta$ for the unitary symmetry (dashed histograms) and $J_g = 0.28\Delta$, $\sigma_1 = 0.02\Delta$ and $\sigma_2 = 0.04\Delta$ is compared with the experimental distribution of Ref. [3] (solid histograms)](image)

In conclusion, we have developed a HF-Koopmans approach to study spin and interaction effects in diffusive or chaotic quantum dots. In particular we have studied the dependence of the peak-spacing distribution on the fluctuations of the interaction matrix elements to leading order in the inverse Thouless conductance. We find good agreement with the lowest temperature data of Ref. [3].

This work was supported in part by the U.S. DOE grant No. DE-FG-0291-ER-40608. We acknowledge useful discussions with H.U. Baranger, Y. Gefen, A. Polkovnikov and G. Usaj, and in particular with A.D. Mirlin.
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