Even-odd correlations in capacitance fluctuations of quantum dots

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We investigate effects of short-range interactions on the addition spectra of quantum dots using a disordered Hubbard model. A correlation function $\mathcal{S}(q)$ is defined on the inverse compressibility versus filling data, and computed numerically for small lattices. Two regimes of interaction strength are identified: the even/odd fluctuations regime typical of Fermi-liquid ground states, and a regime of structureless $\mathcal{S}(q)$ at strong interactions. We propose to understand the latter regime in terms of magnetically correlated localized spins. [S0163-1829(96)50544-8]

Coulomb interactions and disorder in electronic systems have posed a major challenge to condensed matter physics for quite some time. Quantum dots with discrete electronic spectra offer a new avenue to this problem. A direct probe to the ground-state energy is given by Coulomb blockade peaks in the conductance as the gate voltage is varied.[1–4] Theory of spectral fluctuations of noninteracting electrons has made much progress during the last decade due to the advent of semiclassical approximations, random matrix theory,[5] and the nonlinear σ model approach.[1,6] However since Coulomb interactions are essential for the ''Coulomb blockade'' effect, one may wonder as to the validity of noninteracting approximations to quantum dots in general. In particular: is the ground state qualitatively similar to or different than a Fock state of the lowest single electron orbitals?

To gain insight into this question, we consider a system of interacting electrons on a finite tight binding lattice with on-site disorder. The inverse compressibility at consecutive fillings is

$$\Delta(N)=E(N+1)-2E(N)+E(N-1), \tag{1}$$

where $E(N)$ is the ground-state energy of a dot with N electrons. (We assume weakly coupled leads such that N is well defined within the area of the dot.) By varying a gate potential φ, the dot's energy is modified to $E_N^{\varphi}=E(N)-e\varphi N$. Conductance peaks through the leads are observed at $E^{\varphi}(N)=E^{\varphi}(N+1)$, i.e., at potentials $e\varphi_N=E(N+1)-E(N)$. Thus differences between the peak potentials φ_N yield direct measurements of $\Delta(N)$ which can be defined as e^2 times the *discrete inverse capacitance* of the dot.

We shall model the single electron part of the dot's Hamiltonian by a site-disordered tight-binding model

$$\mathcal{H}_0=\sum_{is} w_i c_{is}^{\dagger}c_{is}-\sum_{\langle ij\rangle} t_{ij}(B)c_{is}^{\dagger}c_{js} \tag{2}$$

$$=\sum_{ns} \epsilon_n \alpha_{ns}^{\dagger}\alpha_{ns}, \tag{3}$$

where $c_{is}^{\dagger}$ creates an electron at site i with spin s, $\langle ij\rangle$ denote nearest neighbors on the lattice, and w_i are random site energies taken from a uniform distribution in the domain $[-W/\sqrt{3},W/\sqrt{3}]$. $\alpha_{ns}^{\dagger}$ creates an electron in eigenstate ϕ_n and spin s. An orbital coupling to a magnetic field is included by defining $t_{ij}(B)=te^{ie\mathbf{A}\cdot\mathbf{x}_{ij}}$, where $\nabla\times\mathbf{A}=\mathbf{B}$.

In the absence of electron interactions, the inverse compressibility is given by

$$\Delta_0(N)=\begin{cases} 0, & N=2n+1 \\ \epsilon_{n+1}-\epsilon_n, & N=2n, \end{cases} \tag{4}$$

where ϵ_n are defined in Eq. (3). We find it useful to define the ''Ising'' variables,

$$S(N)\equiv\frac{\Delta(N)-\Delta(N-1)}{|\Delta(N)-\Delta(N-1)|}, \tag{5}$$

and a corresponding correlation function on a series of L consecutive data points,

$$\mathcal{S}(q)\equiv\frac{1}{L^2}\sum_{i,j=1}^{L} S(N_i)S(N_j)\exp[-i(N_i-N_j)q]. \tag{6}$$

Obviously, the noninteracting spectrum has perfect ''long-range antiferromagnetic correlations'' i.e., $\mathcal{S}_0(\pi)=1$.

Coulomb interactions are treated by separating the interactions into the long- and short-range parts. A crude approximation to these two terms is given by an infinite range term and an on-site term

$$H_{\rm int}=e^2\frac{N(N-1)}{2C}+U\sum_i n_{i\uparrow}n_{i\downarrow}, \tag{7}$$

where $n_{is}=c_{is}^{\dagger}c_{is}, s=\uparrow,\downarrow$. It is clear that the first term simply adds a constant e^2/C to $\Delta(N)$, and therefore does not alter $\mathcal{S}(q)$. Thus in our model, deviations of $S(q)$ from $\mathcal{S}_0(q)$ must therefore be a consequence of the Hubbard interactions described by the second term in Eq. (7).

We restrict ourselves to a square lattice of $\mathcal{N}$ sites with periodic boundary conditions, and to disorder strength W appropriate for the ''diffusive'' regime, i.e., the mean free path

l for the noninteracting electrons is of the order of, or smaller than, the system's linear length L. Using $l=v_F\tau$, where the inverse lifetime is calculated in the Born approximation to be $\tau=8t\hbar/2\pi W^2$, we find $L/l=(\pi/16)\sqrt{\mathcal{N}}(W/t)^2$.

Perturbation theory: We diagonalize $\mathcal{H}_0$ on a square lattice of size $\mathcal{N}$, with periodic boundary conditions for a given realization $\{w_i\}$. The single electron eigenenergies $\{\epsilon_n\}$ and wave functions $\psi_n(i)$ are assumed to be known. The first order correction to Eq. (4) are given by second differences of the first-order energies,

$$E_1(N)=U\sum_{n_\uparrow\leqslant n_{F\uparrow}}\sum_{n_\downarrow\leqslant n_{F\downarrow}}|\psi_{n_\uparrow}|^2|\psi_{n_\downarrow}|^2. \tag{8}$$

Here we appeal to the random matrix properties of $\mathcal{H}_0$ in the diffusive regime, in order to estimate the magnitude and fluctuations of E_1 analytically. We assume a random vector model (RVM) where all eigenvectors ψ_n are random complex unit vectors of dimension N whose ensemble averaged correlations are[8]

$$\langle\psi_n(\mathbf{x}_i)\psi_m(\mathbf{x}_j)\rangle_{\mathrm{RVM}}\propto\delta_{ij}\delta_{nm}. \tag{9}$$

Using the orthonormalization constraints for ψ_n we obtain after some algebra[9] that the RVM estimate for the average first order correction to Δ is

$$\Delta_1^{\mathrm{RVM}}(N)=\begin{cases}\dfrac{3U}{\mathcal{N}+2}, & N \text{ odd}\\ -\dfrac{2U}{\mathcal{N}+2}, & N \text{ even.}\end{cases} \tag{10}$$

We have compared the RVM estimates to numerical results for disorder averaged $\Delta_1^{\mathrm{num}}(N)$ for odd and even N, respectively. Calculations have been done for lattice sizes $\mathcal{N}=56,110,210,420$, with the disorder varied in the range $l/L\in[0.1,2.5]$. We find that

$$\Delta_1^{\mathrm{num}}(N)\in\begin{cases}[2.3,3.5]\times\dfrac{U}{\mathcal{N}}, & N \text{ odd}\\ [-1.3,-2.7]\times\dfrac{U}{\mathcal{N}}, & N \text{ even.}\end{cases} \tag{11}$$

Comparison of Eq. (10) to Eq. (11) shows the RVM estimates to be in the right ballpark in relation to those obtained by numerically determining ψ_n of the disordered tight-binding model. The main lesson learned by this calculation is that first order Hubbard corrections *reduce* on average the fluctuations in $\Delta(N)$ since they are positive for odd N and negative (on average) for even N.

Variational theory for sign flips. The weak coupling regime is defined where the noninteracting ground state $|\Psi_0\rangle$ is variationally stable against particle hole excitations. When interaction strength exceeds a certain threshold, it is variationally advantageous to create spin-polarized electron-hole pairs, e.g., $c^\dagger_{n_{F\uparrow}+1}c_{n_{F\downarrow}}|\Psi_0\rangle$, which reduce the Hubbard interaction energy at the expense of enhanced single particle (kinetic) energy $\epsilon_{n_F+1}-\epsilon_{n_F}$. In this variational theory, for an even number of electrons the threshold for forming a triplet is given by the inequality

$$0>\Delta\epsilon-[E_1^t(2n)-E_1^s(2n)]=\Delta\epsilon+U\mathcal{F},$$

$$\mathcal{F}\equiv\sum_i\left((|\psi_{n+1}(\mathbf{x}_i)|^2-|\psi_n(\mathbf{x}_i)|^2)\left(\sum_{n'}^{n-1}|\psi_{n'}(\mathbf{x}_i)|^2\right)+|\psi_n(\mathbf{x}_i)|^4\right), \tag{12}$$

where $\Delta\epsilon=\epsilon_{n+1}-\epsilon_n$ and E_1^t,E_1^s are the interaction corrections to the singlet and triplet energies, respectively. Equation (12) can alternatively be written as an inequality for $\Delta(N)$ which includes up to first-order corrections in U:

$$0>\Delta(2n)-\Delta(2n-1)-U\sum_i(|\psi_{n+1}(\mathbf{x}_i)|^2-|\psi_n(\mathbf{x}_i)|^2)|\psi_n(\mathbf{x}_i)|^2. \tag{13}$$

For extended random wave functions, the last term in Eq. (13) is readily seen to be of order $\mathcal{N}^{-3/2}$ and thus negligible in comparison to $\Delta(2n)-\Delta(2n-1)$. Equations (12,13) establish the connection between formation of triplets and sign flips of $S(N)$ [defined in Eq. (5)]. The sign flips degrade the ''antiferromagnetic'' correlations of $\mathcal{S}(q)$ as U is increased. We shall proceed to estimate the leading dependence of $\mathcal{S}(\pi;U)$ by a statistical calculation based on the properties of the noninteracting spectrum of $\mathcal{H}_0$ with and without an orbital magnetic field.

We assume that the level spacing statistics of H_0 in the diffusive regime of $L/l\simeq1$ is that of a random matrix in the Gaussian orthogonal ensemble (GOE). In the presence of a magnetic field B whose flux through the dot is of order of one flux quantum divided by $\mathcal{N}$[1/4,7] the level spacing statistics of $\mathcal{H}_0$ turns into the Gaussian unitary ensemble (GUE). These assumptions were checked numerically and verified quite well for the Hamiltonian (3) with and without external flux, on lattices up to $\mathcal{N}=420$ sites.

The Wigner distributions of $\Delta\epsilon$ are[8]

$$P_2^{\mathrm{GOE}}(\Delta\epsilon)=\frac{\Delta\epsilon\pi}{2\overline{\Delta^2}}e^{-[\pi(\Delta\epsilon)^2/4\overline{\Delta^2}]},$$

$$P_2^{\mathrm{GUE}}(\Delta\epsilon)=\frac{32(\Delta\epsilon)^2}{\pi^2\overline{\Delta^3}}e^{-[4(\Delta\epsilon)^2/\pi\overline{\Delta^2}]}, \tag{14}$$

where $\overline{\Delta}$ is the mean level spacing.

The probability distribution of the interaction term in Eq. (12) is denoted by

$$P_1(U\mathcal{F})=\frac{1}{U}P_1'(\mathcal{F}), \tag{15}$$

where P_1' is a dimensionless function of its dimensionless argument. The probability of satisfying Eq. (12) is given by the double integral

$$P^{\mathrm{flip}}(U)=\int_0^\infty d\mathcal{F}P_1'(\mathcal{F})\int_0^{U\mathcal{F}}d(\Delta\epsilon)P_2(\Delta\epsilon), \tag{16}$$

which at weak coupling $U\ll\overline{\Delta}$ we can evaluate using the low-energy expansion of P_2 and obtain

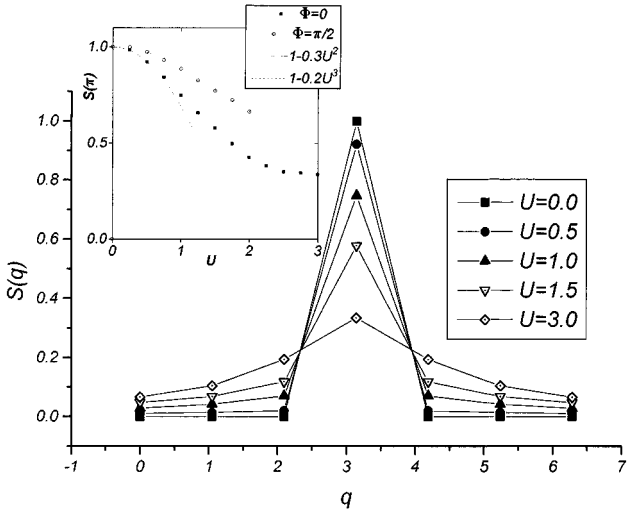

FIG. 1. Averaged $\mathcal{S}(q)$, Eq. (6), for the disordered Hubbard model on a 3×3 torus. Inset: $\mathcal{S}(\pi)$ for the $\phi=0$ and $\phi=\pi/2$ cases, fitted to estimates of RVM variational theory.

$$P_{\mathrm{GOE}}^{\mathrm{flip}} \approx U^2 \frac{\pi}{2\overline{\Delta^2}} \int_0^\infty dx P_1'(x) \int_0^x d\epsilon\epsilon = U^2 \frac{\pi}{4\overline{\Delta^2}} \langle \mathcal{F}^2 \rangle$$

$$P_{\mathrm{GUE}}^{\mathrm{flip}} \approx U^3 \frac{32}{\pi^2 \overline{\Delta^3}} \int_0^\infty dx P_1'(x) \int_0^x d\epsilon\epsilon = U^3 \frac{32}{3\pi^2 \overline{\Delta^3}} \langle \mathcal{F}^3 \rangle. \tag{17}$$

At weak coupling the reduction in the perfect even-odd correlations are proportional to P^{flip}, and therefore we find that

$$1-\mathcal{S}(\pi) \propto P^{\mathrm{flip}}(U) \propto \begin{cases} U^2, & \mathrm{GOE} \\ U^3, & \mathrm{GUE}. \end{cases} \tag{18}$$

Numerical diagonalizations. The variational theory and the random matrix estimates were checked by numerically diagonalizing the full interacting Hamiltonian on a lattice size 3×3 with periodic boundary conditions with a magnetic flux ϕ threading the lattice at its center. We varied the number of electrons for each specific realization from $N=5$ to $N=13$, each time diagonalizing the corresponding $\binom{N}{2,N}$ matrix (i.e., maximum matrices of size 48620×48620 for $N=9$) for various values of U. Then $S(\pi)$ was calculated and averaged over 500 different realizations.

Two different values of ϕ were considered: $\phi=0$ and $\phi=\pi/2$, which should correspond to the GOE and GUE statistics. Because of the small size of the samples the maximum flux of $\phi=\pi/2$ is not strong enough to completely remove time reversal symmetry, and the level spacing is a combination of the GOE and GUE level spacing. The results of $S(\pi)$ for both cases are presented in Fig. 1. It can be seen that the general behavior predicted in Eqs. (18) is observed.

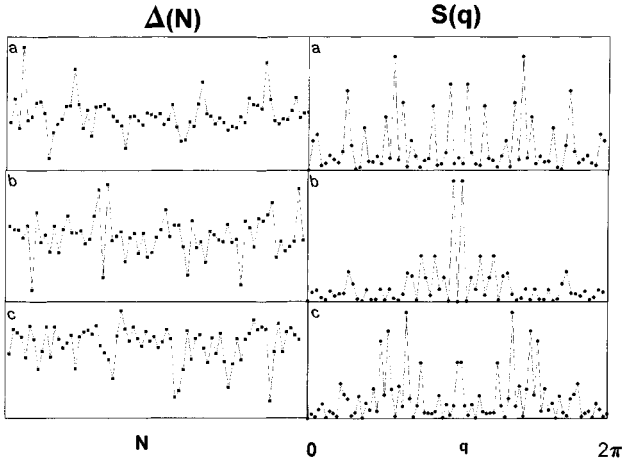

FIG. 2. Experimental second energy differences from quantum dots of GaAs, Ref. 10 (top and middle), and InO, Ref. 11 (bottom), with corresponding correlations $\mathcal{S}(q)$. Correlations indicate that the top and bottom data are in the strongly interacting regime.

A reasonable fit for the numerical results in the regime $U\leqslant1$ is obtained by $S(\pi)=1-0.3U^2$ for $\phi=0$ and $S(\pi)=1-0.2U^3$. This supports the validity of the GOE (GUE) statistics for the case $\phi=0$ ($\phi=\pi/2$).

Experimental $\mathcal{S}(q)$. In Fig. 2 we have plotted three sets of data for $\Delta(N)$ and $\mathcal{S}(q)$. The top two sets were measured on two quantum dots formed in a high mobility $GaAs/Al_xGa_{1-x}As$ two-dimensional electron gas by metal gating. The lithographic areas of the dots were 0.5×0.5 pm, and they both contain about 50–100 electrons.

The bottom data, measured on InO, was taken from Ref. 11. It is interesting to note that $\mathcal{S}(q)$ shows strong even-odd correlations only for the middle data set, while it has no remnance of these correlations for the top and bottom sets.

We do not presently understand the experimental conditions under which even-odd periodicity appear. Based on our modeling and variational theory, we dare to speculate that flatness of $\mathcal{S}(q)$ correlations is associated with creation of magnetically correlated ground states (probably of short range) which differ from Fermi-liquid-type Fock states. We note that two recent papers[12,13] show spin-related structure in dots with low electron fillings. However, the implications of these correlations on the thermodynamic limit are beyond the scope of this work. Complementing this work, Ref. 14 treats the effects of direct *long-range* Coulomb interaction and disorder on the fluctuations in $\Delta(N)$.

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[1] For extensive reviews see Rev. Mod. Phys. **64**, 849 (1992); U. Meirav and E.B. Foxman, Semicond. Sci. Technol. **10**, 255 (1995), and references therein.

[2] T. Heinzel *et al.*, Europhys. Lett. **26**, 689 (1994).

[3] T. Sakamoto, S.W. Hwang, Y. Nakamura, and K. Nakamura, Appl. Phys. Lett. **65**, 875 (1994).

[4] Y. Isawa, J. Phys. Soc. Jpn. **64**, 14 (1995).

[5] Random matrix theory also applies to excitations in interacting systems. Here, however, we are interested in ground-state energies.

[6] For a review see A.D. Stone and H. Bruus, Physica B **189**, 43 (1993).
[7] B.D. Simons and B.L. Altshuler, Phys. Rev. Lett. **70**, 4063 (1993).
[8] M.L. Mehta, *Random Matrices* (Academic Press, New York, 1964).
[9] O. Prus, M.S. thesis, Technion, 1996.
[10] The measurements were carried out by U. Sivan and Y. Aloni. Experimental details are reported in Y. Aloni, M.S. thesis, Technion, 1993.
[11] V. Chandrasekhar and R.A. Webb, J. Low Temp. Phys. **97**, 9 (1994).
[12] S. Tarucha *et al.* (unpublished).
[13] R.C. Ashoori *et al.*, Phys. Rev. Lett. **71**, 613 (1993).
[14] U. Sivan, R. Berkovits, Y. Aloni, O. Prus, A. Auerbach, and G. Ben-Yoseph, Phys. Rev. Lett. **77**, 1123 (1996).