Mesoscopic Fluctuations in the Ground State Energy of Disordered Quantum Dots

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The ground state energy of disordered quantum dots is studied experimentally as a function of dot population. The fluctuations are found to be considerably larger than those predicted by random matrix theory and to display different statistics. Exact diagonalization of the Hamiltonian pertaining to small clusters shows, indeed, that the random matrix statistics holds only for weak Coulomb interactions. As the interaction is made realistic, a crossover to a different statistics, similar to the experimental one, is observed. The statistics crossover is accompanied by appearance of short range spatial correlations in the electron density. [S0031-9007(96)00839-3]

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The spectrum of many complex systems display a remarkable statistical universality which is well described by the random matrix theory (RMT) [1]. Some examples are hydrogen atoms at strong magnetic fields, complex molecules, large nuclei, and the eigenmodes of chaotic microwave cavities. In recent years it became increasingly clear that the excitation spectra of disordered quantum dots display the same statistics [2]. The pioneering works by Efetov [3] and Altshuler and Shklovskii [4] have been followed by a large body of theoretical studies, and recently some predictions of RMT have been confirmed experimentally [5].

The RMT and its ramifications can in principle be employed to estimate the ground state level fluctuations in mesoscopic systems. We were therefore surprised to find in the experiments reported below that the ground state energy of disordered GaAs dots, as well as that of In_2O_{3-x} wires [6], follows a statistics different in the following ways: (a) The fluctuations are up to 5 times larger than those predicted by RMT. (b) The probability distribution for the fluctuations is different. (c) The fluctuations in the ground state energy do not scale with the level spacing which is the only energy scale in RMT.

Motivated by these experimental results we have numerically studied the addition spectra of small clusters and found the RMT prediction holds, indeed, only for weak Coulomb interaction. As the interparticle interaction is made larger, the fluctuations grow and acquire a statistics similar to the experimental one. In contrast to the RMT regime, where fluctuations are on the order of the average level spacing, here they scale with the charging energy.

The ground state energy was measured using the Coulomb blockade phenomenon. A small quantum dot is weakly coupled to two current leads while a third electrode, called gate, is used to vary its electrostatic potential. At temperatures lower than the dot's charging energy, the charge is typically quantized and constant, and the structure is hence insulating (Coulomb blockade). However, by varying the gate voltage, the dot's potential can be tuned to a point where the ground states with N and $N + 1$ electrons are degenerate, thus allowing for current flow through the dot. The resulting conductance vs gate voltage curve comprises a series of sharp peaks marking the degeneracy points between consecutive states differing by one electron. Such traces have been observed in semiconductors [7], metals [8], and metal oxide [9] structures. A careful analysis of the peak spacings yields the ground state energy as a function of dot population [10].

The degeneracy condition between the N and $N + 1$ states implies $E_N^{N+1} + \mu = E_{N+1}^{N+1}$, where μ is the leads chemical potential and E_i^j is the ground state energy of the dot with i electrons and a gate voltage V_G^j corresponding to the jth conductance peak. Since the average potential induced by the gate is linear in V_G, $E_i^j = \varepsilon_i^j - ei\alpha V_G^j$, where α is the average ratio between the dot's capacitance with respect to the gate to its total capacitance, and ε_i^j is the ground state energy, *including* possible mesoscopic fluctuations in the interaction between dot electrons and the gate. Substitution of E_i^j into the degeneracy conditions yields $\varepsilon_{N+1}^{N+1} - \varepsilon_N^{N+1} = \mu - e\alpha V_G^{N+1}$ or

$$\begin{aligned}\Delta_2^{N+1} &\equiv \varepsilon_{N+1}^{N+1} - \varepsilon_N^{N+1} - \varepsilon_N^N + \varepsilon_{N-1}^N \\ &= e\alpha(V_G^{N+1} - V_G^N)\,. \qquad (1)\end{aligned}$$

Since the dot's charge distribution may depend on V_G, ε_N^{N+1} is generally different from ε_N^N. The right hand side is the spacing between subsequent conductance peaks translated to actual potential in the dot. The proportionality coefficient α is accurately extracted from the experiment by fitting the conductance peaks to a derivative of a Fermi function at different temperatures [11]. Equation (1) thus constitutes a unique and accurate method for studying the ground state energy as electrons are added one at a time to the dot.

In the constant interaction (CI) model, which was extensively employed to interpret Coulomb blockade data, $\varepsilon_i^j = e^2 i^2/2C + \sum_{k=l}^{i} \eta_k$, where C is the dot's smoothly varying total capacitance and η_k is the kth single particle energy. Substituting this expression into Eq. (1) one finds $\Delta_2^{N+1} = e^2/C + \eta_{N+1} - \eta_N$. Within the CI model and RMT, the distribution function of Δ_2 should satisfy the Wigner surmise,

$$P\left[\frac{\Delta_2 - e^2/C}{\Delta}\right] = \frac{\pi}{2}\left[\frac{\Delta_2 - e^2/C}{\Delta}\right] \times \exp\left[-\frac{\pi}{4}\left(\frac{\Delta_2 - e^2/C}{\Delta}\right)^2\right],$$

where Δ is the average single particle level spacing. Consequently, the average fluctuations in Δ_2 should be given by $\delta\Delta_2 \equiv \sqrt{\langle\Delta_2^2\rangle - \langle\Delta_2\rangle^2} = \sqrt{4/\pi - 1}\,\Delta \cong 0.52\Delta$. We show below that the CI model fails to describe the experimental data. In fact we argue that the main source of fluctuations in the ground state energy is mesoscopic fluctuations in the Coulomb interaction (capacitance) rather than single particle level fluctuations.

The GaAs dots studied in the experiment are electrostatically defined in a modulation doped two dimensional electron gas having sheet mobility and carrier concentration of 5×10^5 V cm^2 s^{-1} and 3.1×10^{11} cm^{-2}, respectively. The lithographic area is 0.6×0.5 μm^2. The electrical area at low gate voltages, extracted from Aharonov-Bohm oscillations in the quantum Hall effect regime, is 0.15 μm^2, indicating a plausible 80 nm depletion region around the gates. The measured Δ_2 for two dots is depicted by squares and circles in Fig. 1 vs dot population, N (the second curve is shifted down by 200 μeV for clarity). As the dot is made larger, the average Δ_2 grows smaller, reflecting the increase in the average dot capacitance. Superimposed on the monotonous dependence, large fluctuations are observed. The characterization of these fluctuations is the subject of the present manuscript.

The prediction of the CI model combined with RMT (dashed lines in Fig. 1) fails to account for the data in the following ways: (a) Fluctuations larger than expected—for the dots of Fig. 1, $\Delta \cong 30$–60 μeV, implying according to the CI model $\delta\Delta_2 \leq 30$ μeV. This value is up to a factor of 5 smaller than the experimentally measured fluctuations. (b) Independence upon average level spacing—as the dot population is varied from $N \cong 60$ at the left hand side to $N \cong 130$ at the right hand side, Δ shrinks by a factor of 2 with no observable effect on $\delta\Delta_2$. (c) "Wrong statistics"—the histogram of the experimental spacing distribution of the top trace in Fig. 1, together with a similar distribution extracted from In_2O_{3-x} data [6], are depicted in Fig. 2. Evidently the experimental distributions are wider and more symmetric than the RMT one (solid line). The similarity between the GaAs and the InO data is striking in light of the 3D nature of the latter sample and its density of states being two orders of magnitude larger than that of GaAs dots (two linear dimensions are correspondingly smaller).

Considering the mentioned discrepancies we are led to conclude that the CI model fails to approximate the data. Since the Coulomb blockade is governed by the charging energy, we postulate that the fluctuations in Δ_2 are determined by the Coulomb interaction rather than Δ. In fact we find for all measured dots, as well as for In_2O_{3-x} data, $\delta\Delta_2 \cong 0.1 - 0.15e^2/C$, regardless of N and Δ.

The large capacitance fluctuations are characteristic of zero or small magnetic fields. At fields larger than 0.6–0.8 T, the fluctuations are replaced by considerably

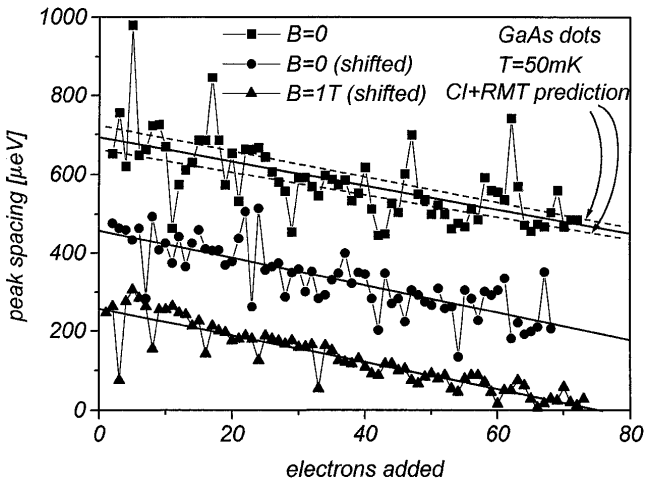

FIG. 1. Peak spacing vs number of electrons added to GaAs dots. Second and bottom traces have been shifted down by 200 and 400 μeV, respectively, for clarity. Solid line: linear fit.

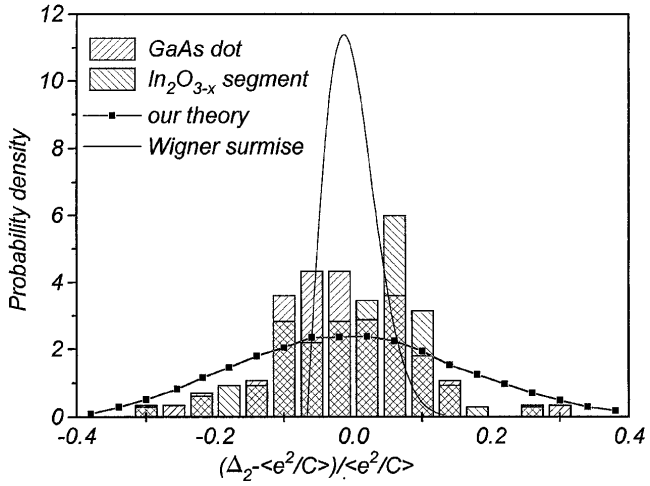

FIG. 2. Histogram of the normalized fluctuations of Δ_2 in GaAs dots and In_2O_{3-x} segments (bar graphs), the numerically obtained limiting distribution for a 4×4 cluster with eight electrons (solid squares), and the Wigner surmise (solid line).

smaller oscillations whose period can be directly related to the Landau level population. The experimentally measured Δ_2 at $B = 1$ T, is presented in Fig. 1 by triangles. There are five Landau levels at small population (corresponding to the five dips) and six Landau levels at larger populations (period of oscillation). Note the Landau levels related structure appears when the cyclotron energy becomes about twice the charging energy (1.2 compared with 0.6 meV). Our magnetic field data indicate that large spacings are accompanied by smaller conductances. In the zero field case, we never find such correlations.

Large capacitance fluctuations exist only at low temperatures. As the temperature is raised above 400–500 mK, they are gradually suppressed and at $T = 900$ mK, the peak spacing is practically uniform. The temperature scale for the disappearance of the fluctuations is consistent with a single particle level spacing Δ and about a factor of 5 smaller than the charging energy. However, the Coulomb blockade itself disappears around 1.5 K, namely, for $k_B T \cong 0.25 e^2/C$. Finally, when the dot is thermally cycled to room temperature and back, the exact peak positions change, though the average spacing remains the same. This sensitivity to impurity configuration is characteristic of mesoscopic systems.

In an attempt to account for the experimental results, we have studied numerically the ground state level statistics of interacting electrons on a small 2D cylinder. The Hamiltonian was given by

$$H = \sum_{k,j} \varepsilon_{k,j} a^{\dagger}_{k,j} a_{k,j} - V \sum_{k,j} (a^{\dagger}_{k,j+1} a_{k,j} + a^{\dagger}_{k+1,j} a_{k,j} + \text{H.c.}) + U \sum_{k,j>l,p} \frac{a^{\dagger}_{k,j} a_{k,j} a^{\dagger}_{l,p} a_{l,p}}{|r_{k,j} - r_{l,p}|}, \quad (2)$$

where $a^{\dagger}_{k,j}$ was the electron creation operator at a site $\{k, j\}$, $\varepsilon_{k,j}$ was the energy at that site chosen randomly in the range $[-W/2, W/2]$, V was the hopping matrix element, and U was the Coulomb interaction energy over one lattice constant. Distances were measured in lattice constant units. The inverse compressibility Δ_2 was computed by exact diagonalization of the above Hamiltonian with three consecutive numbers of electrons at a given impurity configuration. Statistics was obtained by averaging over 200–500 realizations of the disorder. The resulting $\delta\Delta_2/\langle\Delta_2\rangle$ is depicted in Fig. 3 vs e^2/CV for various sample sizes, disorder, and electron number. The charging energy was obtained by fitting the average ground state energy by $e^2N^2/2C$. For $U = 0$ the RMT prediction, $\delta\Delta_2/\langle\Delta_2\rangle = \sqrt{4/\pi - 1}$, is reproduced. As the interaction is turned on, $\langle\Delta_2\rangle$ grows from Δ to $e^2/C \gg \Delta$, and the relative fluctuations decrease and converge to $\delta\Delta_2/\langle\Delta_2\rangle \cong 0.10$–$0.17$ almost *independently* of the interaction strength, sample size, and electron population.

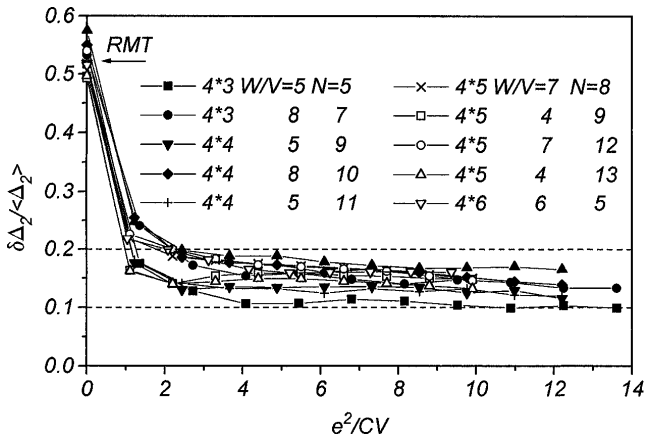

FIG. 3. Normalized fluctuations in Δ_2 vs cluster capacitance for various clusters and electron population (numerical calculations).

To relate the experiment to theory we use r_s, the ratio between the Coulomb interaction among two neighboring electrons and the Fermi energy. For half filled band, the tight binding model gives $r_s = (U/4V)\sqrt{\pi/2}$. The crossover to the almost constant capacitance fluctuations regime hence occurs at $r_s \geq 0.75$. In all our experiments, $r_s > 1$, implying they were carried out in the latter regime. The agreement of theory with the experiment might hence indicate universal mesoscopic fluctuations in the capacitance of real dots.

The crossover from an RMT statistics to a new one is also apparent from the distribution function $P(\Delta_2/\langle\Delta_2\rangle)$ depicted in Fig. 4. For noninteracting electrons, the Wigner surmise is reproduced while for stronger interaction, the distribution becomes almost symmetric and independent of interaction strength. Note in this figure the Wigner surmise appears wider than the finite interaction

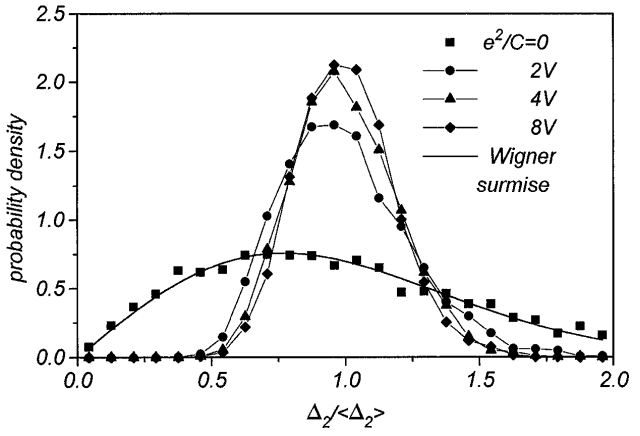

FIG. 4. Distribution function of the normalized fluctuations in Δ_2 for different interaction strengths for a 4×4 cluster with eight electrons. Note convergence to a limiting distribution.

distribution. This results from the normalization being Δ for $U = 0$ and $e^2/C \gg \Delta$ for finite interactions. The absolute fluctuations grow with U. The limiting distribution is compared to the experimental one in Fig. 2. It clearly agrees with the experiment better than RMT. Numerical Hartree-Fock calculations on considerably large samples also reveal increased absolute fluctuations as the interaction is made stronger [12]. It is emphasized that all figures present fluctuations in the ground state energy. The excitation spectra satisfy the RMT statistics in the full range of interaction strength studied here [13].

A failure of RMT as the interaction is made stronger is in retrospect expected. In the extreme case of Wigner crystallization, a preferred basis of the Hamiltonian appears thus violating the main assumption of RMT, namely, the equivalence of all bases. To characterize spatial ordering we have numerically evaluated the two point correlation function $C(r_{i,j}) = [a_i^\dagger a_i - \langle a_i^\dagger a_i \rangle][a_j^\dagger a_j - \langle a_j^\dagger a_j \rangle]/\langle a_i^\dagger a_i \rangle \langle a_j^\dagger a_j \rangle$ and found the statistics crossover is accompanied by the appearance of short range spatial correlations (Wigner crystallization occurs at considerably stronger interaction). A theory that deals with spatial ordering and its incommensurability with disorder or dot boundaries is not available. This is, however, a plausible mechanism for producing large mesoscopic fluctuations in the ground state energy. We mention in this context that the ground state energy of classical electrons on a sphere [14] is characterized by $\delta\Delta_2/\langle\Delta_2\rangle \cong 0.1$ due to the mismatch of the electronic ordering with the sphere geometry.

Finally, recent studies [15] of the Coulomb blockade peak height statistics approximately agree (up to the magnetic flux scale) with RMT, though the Δ_2 fluctuations there are also considerably larger than Δ [16]. The reason for this agreement, in spite of the non-RMT Δ_2 fluctuations, is not clear.

In summary, the fluctuations in the ground state energy of quantum dots are found to be considerably larger than those predicted by the CI model and RMT and to display a different statistics. Numerical calculations on small clusters show a crossover from an RMT statistics to a different one as the interaction is made realistic in terms of r_s. The latter statistics agrees better with the experimental data. We propose the interplay between spatial ordering due to Coulomb interaction and impurity or boundary constraints as the mechanism underlying these mesoscopic thermodynamic fluctuations.

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