Interactions in chaotic nanoparticles: Fluctuations in Coulomb blockade peak spacings

Denis Ullmo and Harold U. Baranger

1Laboratoire de Physique Théorique et Modèles Statistiques (LPTMS), 91405 Orsay Cedex, France
2Department of Physics, Duke University, Box 90305, Durham, North Carolina 27708-0305

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We use random matrix models and a Fermi-liquid approach to investigate the ground state energy of electrons confined to a nanoparticle. Our expression for the energy includes the charging effect, the single-particle energies, and the residual screened interactions treated in Hartree-Fock. This model is applicable to chaotic quantum dots or nanoparticles—in these systems the single-particle statistics follows random matrix theory at energy scales less than the Thouless energy. We find the distribution of Coulomb blockade peak spacings first for a large dot in which the residual interactions can be taken constant: the spacing fluctuations are of order the mean level separation $\Delta$. Corrections to this limit are studied using the small parameter $1/k_FL$: both the residual interactions and the effect of the changing confinement on the single-particle levels produce fluctuations of order $\Delta/\sqrt{k_FL}$. The distributions we find are qualitatively similar to the experimental results. Thus, models beyond Fermi-liquid theory are not needed to describe this quantity.

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I. INTRODUCTION

The ground state properties of electrons in nanoparticles or quantum dots—finite systems of fermions interacting via Coulomb forces—remain incompletely understood. One valuable way to probe these systems is via electrical transport (for reviews see Refs. 1–5). The dominant effect is the suppression of the conductance $G$ because of the charging energy needed to add an electron to the nanoparticle and so allow current to flow. This Coulomb blockade (CB) is an essentially classical effect, and the charging energy is simply $E_C=e^2/2C$ where $C$ is the capacitance of the nanoparticle. The blockade can be lifted by adjusting the potential on the particle—with a gate voltage $V_g$, for instance—so that the energy for $N$ electrons is the same as that for $N+1$. The conductance through the nanoparticle has peaks at these degeneracy points; in fact, $G(V_g)$ is periodic with a spacing of $e^2/C$ between the peaks.1,2

At low temperature the electrons remain coherent in the dot and so quantum interference effects must be included in the Coulomb blockade.6–8 Two main effects occur: (1) the peak heights fluctuate from peak to peak because of varying coupling between the wave function in the nanoparticle and the leads and (2) the spacings between the peaks fluctuate because of interference contributions to the ground state energy.

In this paper we focus on quantum effects in the peak spacings (for peak heights see Ref. 9, and references therein). One obvious interference contribution to the ground state energy is the single-particle energy levels caused by the confinement in the quantum dot. But there may also be contributions from the interactions in the dot. We will show that for typical parameters characterizing experimental quantum dots or nanoparticles the contributions coming from the interactions must be included.

The simplest treatment of Coulomb blockade peak spacing fluctuations results from writing the ground state energy as the sum of the classical electrostatic energy and the energies of the occupied single particle states.6 Such an expression for a many Fermion system has a long and venerable history in atomic and nuclear physics.10–12 Using $E$ for the energy in the presence of the gate voltage while reserving $E$ for $V_g=0$, we have for $N$ particles

$$E_N(V_g)=E_N-(C_g/C)eNV_g,$$

$$E_N=(e^2/2C)N^2+\sum_{\text{occupied } i\sigma}\epsilon_{i\sigma}.$$  (1)

where $C_g$ is the capacitance of the gate to the dot and $\{\epsilon_{i\sigma}\}$ are the single-particle energy levels. This treatment, known as the constant-interaction (CI) model, is still an important point of reference in the field. The gate voltage $V^*_g$ at which a peak in the conductance occurs is given by the condition $E_N(V^*_g)=E_{N+1}(V^*_g)$. The spacing between two peaks is, then, proportional to the (discrete) inverse compressibility $\Delta^2E$

$$(V^*_g)_{N\rightarrow N+1}-(V^*_g)_{N-1\rightarrow N}\approx E_{N+1}+E_{N-1}-2E_N=\Delta^2E_N.$$  (2)

In the case of the CI model, one gets the simple prediction13

$$\Delta^2E_N=e^2/C \quad \text{for odd } N,$$

$$\Delta^2E_N=e^2/C+(\epsilon_{N/2+1}-\epsilon_{N/2}) \quad \text{for even } N.$$  (3)

Note the drastic odd/even effect in the quantum correction to the classical result $e^2/C$ because of the spin of the electron.

A number of experiments have studied the distribution of CB peak spacings in semiconductor quantum dots,13–18 a particularly flexible and convenient type of nanoparticle. In these high mobility quantum dots, the transport mean free path is significantly larger than the system size (at least twice as large), so that a ballistic description is required. (Experiments could also be performed on diffusive dots but, as far as we know, have not been.) The overall result has been striking disagreement with the CI model. The main discrepancy is that there is no sign of the strong odd/even effect predicted by Eq. (3), though there is evidence of a weak odd/even
effect.\textsuperscript{17,18} In addition, in some of the experiments the widths of the observed distributions are significantly larger than predicted by the CI model assuming the single-particle energy levels \( \varepsilon_i \) are distributed according to the appropriate Gaussian random matrix ensemble. These discrepancies indicate unambiguously that interaction effects play a significant role in the fluctuating part of the Coulomb Blockade peak position.

Two theoretical approaches have been pursued to account for this agreement. First, several groups proceeded via direct calculation. Exact multiparticle diagonalization\textsuperscript{13,19} can, however, only treat problems with a few electrons (\( \lesssim 15 \)) while typically the dots contain a large number of electrons (\( \sim 100–1000 \)). To reach these \( N \), self-consistent techniques have been used. The lack of screening of the exchange term in Hartree-Fock calculations\textsuperscript{20–23} makes those results suspect, however. Density functional theory in the local density or local spin density approximation\textsuperscript{24–26} handles screening properly but approximates the exchange term. Since the subtle interplay between exchange, residual direct interactions, and single-particle level fluctuations is critical in describing quantum dot physics, results from these methods are difficult to generalize.

A second theoretical approach is to formally carry out a screening calculation and then use a short-range screened interaction in the subsequent evaluation of various effects.\textsuperscript{27–29,8} The nature of the screened interaction used is, then, important. Since experimentally the system is much larger than the screening length, the natural choice is the bulk RPA screened interaction. For momentum smaller than \( 2k_F \) this is correctly approximated by the Thomas-Fermi expression. Throughout this paper we consider two-dimensional systems, in which case

\[
V_{TF}(\mathbf{r}) = \int \frac{d\mathbf{q}}{(2\pi)^2} \hat{V}_{TF}(\mathbf{q}) \exp[\mathbf{i} \cdot \mathbf{q} \cdot \mathbf{r}],
\]

\[
\hat{V}_{TF}(\mathbf{q}) = \frac{\nu^{-1}}{1 + r_s^{-1}(|\mathbf{q}|/k_F)/\sqrt{2}}, \tag{4}
\]

where \( \nu \) is the mean density of states (including spin degeneracy), and \( r_s = r_0/a_0 \) with \( \pi r_0^2 \) the average area per electron and \( a_0 \) the Bohr radius in the semiconductor. Although \( \hat{V}_{TF} \) is proportional to \( r_s \) for small \( r_s \), it becomes largely independent of \( r_s \) when \( r_s \gtrsim 1 \), which is the case experimentally. In fact, within any RPA-like approach the interaction cannot become stronger than

\[
V_{\text{zero-range}}(\mathbf{r}) = \nu^{-1} \delta(\mathbf{r}) \tag{5}
\]

which we refer to as the zero-range limit of the potential. Thus, once screening is included, the interaction is small, and one has the usual picture of weakly interacting Landau quasiparticles, a Fermi liquid, appropriate for good conductors.

To obtain results very different from those of the CI model, one possibility is to abandon the Fermi-liquid-like approach and to assume, for instance, that screening is not as efficient in a nanoparticle as in the bulk, and so conclude that large zero-range interactions, not amenable to a perturbative treatment, should be used.\textsuperscript{13,19,27,28} Giving up a Fermi-liquid description of the interacting electrons is, however, a drastic step. It implies, in short, that the physics of quantum dots has little connection to that for unconfined electrons in this density range. This is an exciting possibility, but also one which requires skeptical consideration. It therefore seems reasonable to see how far one can go within the more standard Fermi-liquid description. Indeed, even in this framework, we know that the CI model is inadequate.\textsuperscript{27–34}

An accurate treatment of the spacing or inverse compressibility certainly requires that the average effect of the residual interactions be added to the CI model. By “residual interactions” we mean interactions beyond the simple classical effect taken into account in the constant interaction model and, in particular, interactions between the quasiparticles after the bare electrons are dressed through screening. While they make a smaller contribution to the total energy than those considered in Eq. (1), residual interactions make a contribution of order the mean level separation to the second difference \( \Delta^2 E \), comparable to the single-particle contribution in Eq. (3).\textsuperscript{27,29–33} In the limit of a large nanoparticle—whose typical dimension \( L \) is many times the electron wavelength \( k_F L \gtrsim 1 \)—only the average effect of residual interactions needs to be added to the CI model.\textsuperscript{29–32} We shall refer to this approximation as the constant exchange and interaction (CEI) model. The intuitive argument for this is that the effect of residual interactions involves integrating wave functions over the entire volume of the nanoparticle and so self-averages. Even if one neglects the fluctuations of the residual interaction terms, because of fluctuations of the single-particle levels, the “constant exchange” contribution can modify the total spin of the nanoparticle by favoring nontrivial occupation of the single-particle orbitals and, as a consequence, will significantly modify the fluctuations of the peak spacings.\textsuperscript{27,29} The ground state spin of a large nanoparticle in this regime has been examined in detail,\textsuperscript{30,32–34} but curiously the corresponding distribution of Coulomb blockade peak spacings has not appeared explicitly (see, however, Ref. 29). Therefore, before turning to effects caused by fluctuations in the interactions—our main interest in this paper—we present below the distribution for the CEI model (\( k_F L \gtrsim 1 \)) to use as a reference point.

In the experiments, however, \( k_F L \) is not very large because of the need to have the mean level separation larger than the temperature. This suggests that the variations in interactions caused by properties of individual wave functions are important. The main goal of this paper is, therefore, to study the effect of fluctuations in the electron-electron interactions on the distribution of CB peak spacings.

We limit our study here to the case of chaotic dynamics within the dot, for which a random matrix description of the noninteracting limit can be used.\textsuperscript{35} Much as in the case of diffusive transport, completely chaotic dynamics introduces an important additional energy scale (and only one): the Thouless energy, \( E_{Th} = \hbar/\tau \), is the inverse of the transit time across the dot. It is generally larger than the mean level separation \( \Delta \) but smaller than the Fermi energy; we will assume \( \Delta \ll E_{Th} \ll E_F \). On energy scales less than \( E_{Th} \), it is
now well established that the statistical fluctuations of eigenenergies and eigenvectors of chaotic systems are universal and described by the "classical" random matrix ensembles [e.g., the Gaussian Unitary Ensemble (GUE) for systems which are not time-reversal invariant]. 36 For larger energies (shorter times) the individual dynamics of the system comes into play. Because the primary energy scale of importance here is the mean level spacing, we neglect all effects caused by individual dynamics (they could be added as corrections at a later stage). Even so, we will see that taking account of the additional energy scale $E_{\text{FB}}$ is critical in describing CB peak spacings in nanoparticles.

We focus on two effects in particular: (1) the fluctuation in the residual Hartree and Fock contributions to the ground state energy and (2) the change in the single-particle energies because of changes in the mean field potential as electrons are added to the dot, 29 an effect we refer to as "scrambling." For two-dimensional quantum dots, the changes in energy are evaluated to leading order in the smallness of the dot, the CEI model being used as reference point. We find that both effects contribute terms which are of order $\Delta \sqrt{k_F L}$, up to logarithmic corrections.

From the resulting distributions of CB peak spacings, the debate concerning the necessity to go beyond a Fermi-liquid description to explain the experimental data can be settled: a more conventional approach does explain the main features of the experiment. Thus, a drastic step away from Fermi-liquid theory is unjustified.

The paper is organized as follows. Our starting point is a semiclassical expansion of the ground state energy described in Sec. II—an expansion in $1/k_F L$. The important large dot limit is presented in Sec. III. The next two sections discuss the two main issues—fluctuations of the residual interaction contributions (Sec. IV) and scrambling (Sec. V). The numerical evaluation of the magnitude of these two fluctuation effects is presented in Sec. VI. Section VII takes a first step beyond the Gaussian model introduced in Sec. IV. Finally, our conclusions, including a discussion of the experimental results, appear in Sec. VIII.

II. APPROACH: SEMICLASSICAL CORRECTIONS

In the Coulomb blockade through quantum dots we are faced with a classical theory which works remarkably well to which we want to add quantum effects to leading order. The small parameter is the standard semiclassical one, $\hbar$. In our context, this is equivalent to an expansion in $1/k_F L$ where $k_F$ is the Fermi wave vector and $L$ is the typical size of the nanoparticle. Another useful way to view the corrections is in terms of the dimensionless conductance $g$, given as the ratio of the transit rate $h v_F / L$ to the mean level separation $\Delta$: in this case the expansion is in terms of $1/g$, as in the diffusive mesoscopic regime. 37 For two-dimensional systems, such as the ones we consider here, these two parameters are proportional ($g = k_F L / 2 \pi$).

We proceed by using the method of Strutinsky 38,39 in which the dependence of many-body ground state quantities on particle number can be decomposed into an average and a fluctuating part. While the average part varies smoothly with particle number, the fluctuating part reflects the shell structure of the system. The smooth part is the bulk energy per unit volume integrated over the finite-size system, and the oscillating contributions come from quantum interference effects explicitly caused by the confinement. By supposing that the smooth part is known while the unknown oscillatory contribution is a correction, Strutinsky introduced a physically motivated systematic approach to obtain the shell corrections. 38,39 This shell correction method is essentially a semiclassical approximation. It rests on the fact that the number of particles in the system considered is large, rather than on the interaction between the particles being weak. (One must, of course, work in a regime where the smooth starting point is basically valid.) Since the quantum dots in which we are interested contain on the order of 100 electrons, they are a perfect place to apply the Strutinsky method. We use the formulation recently developed specifically for quantum dots in Ref. 40.

Density functional theory guarantees that the ground state energy of a nanoparticle can be written as a functional of its density $E_{\text{DFT}} = \mathcal{F}_{\text{DFT}}[n_{\text{DFT}}]$. Neglecting quantum interference effects in this functional corresponds to a generalized Thomas-Fermi approximation (generalized because bulk local exchange and correlation can be included) which can also be written as a functional of a (smooth) density $E_{\text{GTF}} = \mathcal{F}_{\text{GTF}}[n_{\text{GTF}}]$. The quantum interference part of the energy $E_{\text{DFT}} - E_{\text{GTF}}$ can be obtained approximately by expanding $n_{\text{DFT}}$ about $n_{\text{GTF}}$ and solving the resulting density functional equations order by order in the oscillating part of the density $n_{\text{osc}}$. To carry out this procedure an explicit form for the exchange-correlation functional is required. Based on the local density approximation, the result derived in Ref. 40 is

$$E_{\text{DFT}} = E_{\text{GTF}} + \Delta E^{(1)} + \Delta E^{(2)},$$

where the first- and second-order correction terms are

$$\Delta E^{(1)} = \epsilon_{\text{osc}}^{\text{GTF}}[V_{\text{eff}}(n_{\text{GTF}})] - \sum_{i=1}^{N} \delta \epsilon_i,$$

$$\Delta E^{(2)} = \frac{1}{2} \int dr dr' n_{\text{osc}}^{\text{GTF}}(r) V_{\text{sc}}(r, r') n_{\text{osc}}^{\text{GTF}}(r').$$

The first-order correction is the oscillatory part of the single-particle energy $\epsilon_{\text{osc}}$, calculated in the smooth Thomas-Fermi potential $V_{\text{eff}}[n_{\text{GTF}}]$; that is, it is the sum of the deviations $\delta \epsilon_i$ of the single-particle levels $\epsilon_i$ from their mean values. In the second-order correction, $n_{\text{osc}}$ is the deviation of the quantum mechanical density calculated in the Thomas-Fermi potential from the smooth starting point $n_{\text{osc}} = n[V_{\text{eff}}(n_{\text{GTF}})](r) - n_{\text{GTF}}$, and $V_{\text{sc}}(r, r')$ is the screened interaction in the nanoparticle. To the order that we are working, using $n_{\text{osc}}$ here is equivalent to using $n_{\text{DFT}} - n_{\text{GTF}}$. 40

In this approach, the ground state energy is, then, the sum of a classical contribution—the generalized-Thomas-Fermi result $E_{\text{GTF}}$—and two quantum contributions—$\Delta E^{(1)}$ and $\Delta E^{(2)}$. With the inclusion of only the first-order contribution, Eq. (6) is simply the CI model for the ground state energy. Because we have the corrections to this model, we can see
exactly when it is applicable. The second-order correction, Eq. (8), has a natural interpretation: the ripples in the density caused by interference (the Friedel oscillations in this setting) interact with each other via an interaction screened by the smooth Thomas-Fermi fluid already present. In fact, the bulk Thomas-Fermi interaction Eq. (4) can often be used for $V_{sc}$. Taking the $r_s \rightarrow \infty$ limit formally yields the zero-range interaction limit (5). This is equivalent to using a Hubbard Hamiltonian on a square lattice with hopping term $t$ and interaction $U$ with $U/t = \pi$.

The result Eq. (6) is, however, inadequate for our purposes here because of the local density approximation used. In particular, the spin degree of freedom is handled poorly in this approximation: while Eq. (8) gives the “direct” interaction between the density ripples, an analogous “exchange” term is missed. A better treatment of the interactions in density functional theory could presumably yield this additional term, by including off-diagonal terms in the density matrix, for instance. In fact, an RPA treatment of the interaction in the diffusive case yields an energy with exactly this structure.29,30 Motivated by these physical considerations, we simply add the exchange term by hand to the expression for the energy $E_{\text{DFT}}$.

For the purposes of deriving CB peak spacing distributions, we thus consider three contributions to the ground state energy: (1) the Thomas-Fermi energy, (2) the deviation of the single-particle energy from its mean, and (3) a Hartree-Fock like (first-order perturbation) treatment of the residual screened interaction. With regard to the last point, higher order corrections can be evaluated and seen to be numerically small for the quantum dots under consideration.34,41 Without the residual interactions, a $N$-particle state is, of course, a Slater determinant in the basis of single-particle eigenstates $\psi_i(r)$; it is characterized by the occupation numbers $(n_{0\sigma}, n_{1\sigma}, \ldots)$, where $n_{i\sigma} = 0$ or $1$, $\sigma = +1$ or $-1$ labels the spin degree of freedom, and $\sum n_{i\sigma} = N$. In this occupation number representation, the expression for the energy is then,

$$E_{\text{tot}}(\{n_{i\sigma}\}) = E_{\text{TF}} + \sum_{i\sigma} n_{i\sigma} \delta \epsilon_i + \frac{1}{2} \sum_{i\sigma,j\sigma'} n_{i\sigma} M_{ij} n_{j\sigma'},$$

where

$$M_{ij} = \int d\mathbf{r} d\mathbf{r}' \left| \psi_i(\mathbf{r}) \right|^2 V_{\text{TF}}(\mathbf{r} - \mathbf{r}') \left| \psi_j(\mathbf{r}') \right|^2,$$

and

$$N_{ij} = \int d\mathbf{r} d\mathbf{r}' \psi_i(\mathbf{r}) \psi_j^\ast(\mathbf{r}) V_{\text{TF}}(\mathbf{r} - \mathbf{r}') \psi_j(\mathbf{r}') \psi_i(\mathbf{r}')$$

are the direct and exchange contributions, respectively. This is the starting point for our study of CB peak spacings.

In order to derive the statistical properties of the CB peak spacings from Eq. (9), the statistical properties of the single-particle eigenvalues and eigenfunctions must be known. For this purpose we assume that the single-particle classical dynamics in the nanoparticle is completely chaotic. In this case it is well-known that the single-particle quantum mechanics is accurately described by random matrix theory (RMT) on an energy scale smaller than the Thouless energy $E_{\text{Th}}$ (which, again, is the inverse time of flight across the system).35,36

We consider only the case of no symmetry here, so that the energy levels $\{\epsilon_i\}$ obey Gaussian unitary ensemble (GUE) statistics. Because of the spatial integrals in the expressions for $M_{ij}$ and $N_{ij}$, the correlations of the wave functions are also needed. For the classical Gaussian ensembles, there is no spatial correlation for a given wave function, except for that arising from the normalization. This is a direct consequence of the fact that no basis should play a particular role for these matrix ensembles. Here, however, because of the particular role played by the kinetic energy, the plane wave basis is special. More precisely, we should implement that each eigenfunction is localized in this basis on the energy scale $E_{\text{Th}}$. We shall therefore build our random matrix ensemble with the following requirements (a practical implementation is given in Sec. VII).

(i) On energy scales smaller than the Thouless energy $E_{\text{Th}}$, one should recover standard GUE properties.

(ii) On scales larger than $E_{\text{Th}}$, an eigenstate of energy $\epsilon$ should appear localized on the energy surface $|\mathbf{k}| = \sqrt{2m\epsilon/h}$. More precisely, we require that in a plane wave basis an eigenstate has an envelope of width

$$\delta k = 1/L,$$

where $L$ is the typical size of the system.

(iii) Finally, we have to implement that the fluctuations of energy levels, and also importantly of local density, saturate at the Thouless energy. That is, if one considers a quantity $A$ expressed as a sum over the energy levels in an energy window $\delta \epsilon$ about the Fermi energy, as the window widens the fluctuations of $A$ increase until $\delta \epsilon \approx E_{\text{Th}}$ beyond which point the fluctuations of $A$ do not further increase.

Except for the fact that the Fermi surface is given an explicit width $\approx E_{\text{Th}}$ due to the finite size $L$ of the system, items (i) and (ii) are very similar to Berry’s modeling of chaotic wave functions42 which assumes that a random superposition of plane waves describes the correlation on scales smaller than the size of the system $L$. Item (iii) is motivated by the known saturation of, for instance, fluctuations in the density of states of chaotic systems at an energy scale of order $\hbar$ divided by the period of the shortest periodic orbit.35 Note that these requirements will yield a random matrix approach noticeably different from the ones discussed in Refs. 27, 30, and 33.

In this model, the wave function statistics that we find are as follows: for a two-dimensional dot of area $A$ (the case that we consider throughout this paper)

$$A(\psi_i(\mathbf{r}) \psi_j^\ast(\mathbf{r}')) = \delta_{ij} \delta(k_F | \mathbf{r} - \mathbf{r}'|)$$

for $|\mathbf{r} - \mathbf{r}'| \ll L$. (In a maximum entropy approach semiclassically restricted by the classical dynamics, this corresponds to keeping only the direct trajectory between $\mathbf{r}$ and $\mathbf{r}'$; the approach can be generalized to keep more classical paths yielding terms similar to those appearing in the diffusive.
regime.) In addition, it can be shown that within our random matrix modeling, one has, up to \(1/(k_F L)\) corrections
\[
\langle \psi_i(r_1)\psi^\dagger_j(r_2)\psi_j(r_3)\psi^\dagger_i(r_4) \rangle = \langle \psi_i(r_1)\psi^\dagger_j(r_2)\psi_j(r_3)\psi^\dagger_i(r_4) \rangle 
+ \langle \psi_i(r_1)\psi^\dagger_j(r_2)\psi_j(r_3)\psi^\dagger_i(r_4) \rangle.
\] (14)

Alternatively, these statistics can be obtained directly by assuming that the distribution of wave functions is Gaussian,\(^43\) though in this case special care must be taken with regard to the normalization constraint.

There are alternate routes to our very natural starting point Eq. (9). One that has been developed recently is the “universal Hamiltonian” approach\(^34,35,32,8\) which uses RPA to treat the interactions, RMT properties of the single-particle wave functions, and the small parameter \(1/g\) to arrive at an effective Hamiltonian.Treating this effective Hamiltonian in the Hartree-Fock approximation leads to the same expression for the ground state energy that we give to treat the interactions, RMT properties of the single-electron singlet state and the two neighboring states for \(N=1\) and \(N+1\) electrons are doublets. The peak spacing is then
\[
\Delta^2 E_N = \Delta^2 E_{GTF} + 2\delta \epsilon_{n+1} - \delta \epsilon_n + \sum_{i=1}^n \left\{ 2 (M_{n+1,i} - M_{n,i}) \right\}.
\]
(15)

On the other hand if \(N\) is odd, still assuming the simplest up-down filling, the peak spacing is
\[
\Delta^2 E_{N+1} = (2g)^2 E_{GTF} + \Delta M_{n+1,n+1}.
\]
(16)

The second difference of \(E_{GTF}\) is almost equal to the charging energy \(e^2/C\) but there is a small correction from the residual interactions \(M_{ij}\) as discussed below.

**III. LARGE DOT LIMIT**

The main simplifying feature in the large dot limit, \(k_F L \gg 1\), is that the fluctuations of the interactions can be neglected, much as for diffusive nanoparticles\(^44-47\) in the large \(g\) limit. The distribution of the spacings is determined, then, by the fluctuations of the single-particle levels and their interplay with the mean residual interactions. Of course, as has been studied previously,\(^27,30-32\) the ground state of the nanoparticle does not necessarily follow the simple filling used in Eqs. (15), (16), and this effect must be included in finding the distribution.

Let us consider the mean and variance of the \(M_{ij}\) and \(N_{ij}\) for levels near the Fermi level. We have, for instance, using Eq. (14)
\[
\langle M_{ij} \rangle = \int d^2r d^2r' \langle \psi_j(r) \psi^\dagger_j(r') \rangle V_{TF}(r-r') \langle \psi_i(r') \psi^\dagger_i(r) \rangle.
\]
(17)

We will make the simplifying assumption that the nanoparticle is a billiard so that the average density is constant \(\langle \psi(r) \rangle = 1/A\) where \(A\) is the area of the particle and the correlation function is given by Eq. (13). In this case the mean values are
\[
\langle M_{ij} \rangle = \frac{\Delta}{2} + \delta_{ij} V_{TF} \langle N_{ij} \rangle/2.
\]
(18)

where
\[
\langle V_{TF} \rangle = \int_0^{2\pi} \frac{d\theta}{2\pi} V_{TF} \sqrt{2(1 + \cos \theta)}.
\]
(19)

is the average on the Fermi surface of \(V_{TF}(k-k')\). In the zero-range interaction limit, these expressions simplify to
\[
\langle M_{ij} \rangle = (1 + \delta_{ij}) \Delta/2, \quad \langle N_{ij} \rangle = \Delta/2.
\]
(20)

The magnitude of the residual interaction effects is thus of order the mean level spacing \(\Delta\). (Note that the \(\Delta\) that we use here and throughout this paper is the spacing of the orbital levels alone and so does not take into account the spin degeneracy factor.) Since energies of this order are critical in determining the spacing of CB peaks—note, for instance, the case of standard up-down filling Eq. (15)—the residual interaction terms must be included in any theory. In particular, the reason for the failure of the simple constant interaction model, Eqs. (1) and (3), is now clear: it does not consistently keep all terms of order \(\Delta\).

The variance or covariance of any of the \(M_{ij}\) and \(N_{ij}\) will involve the correlation function of a wave function. For example, consider the variance of \(M_{ij}\):\[
\text{var}(M_{ij}) = \int d^2r_1 d^2r_2 d^2r_3 d^2r_4 \langle \psi_i(r_1) \psi^\dagger_i(r_2) \psi_j(r_3) \psi^\dagger_j(r_4) \rangle \frac{\langle \psi_i(r_1) \psi^\dagger_i(r_2) \psi_j(r_3) \psi^\dagger_j(r_4) \rangle}{\langle \psi_i(r_1) \psi^\dagger_i(r_2) \psi_j(r_3) \psi^\dagger_j(r_4) \rangle^2}.
\]
(21)

The first equality is due to the normalization of the wave function, and the second derives from Eq. (14). Because of the decay of the autocorrelation of \(\psi_i\), Eq. (13), the spatial integrals in Eq. (21) are restricted to a region where \(r_1\) is close to \(r_3\) and likewise \(r_2\) to \(r_4\). These restrictions produce...
factors of the small parameter $1/k_{F}L$ compared to the mean values. We will evaluate these factors below. However, for the large dot limit here, we see that to zeroth order in $1/k_{F}L$ all the fluctuations in the residual interactions may be neglected.\textsuperscript{30–32} As in the diffusive case,\textsuperscript{44–47} Qualitatively, this is natural because of the averaging implicit in the integral in $M_{ij}$ and $N_{ij}$; when the size of the system is much larger than the scale of oscillation of $\psi_{i}(r)$, particular features of $\psi_{i}$ and $\psi_{j}$ become less important compared to the mean behavior.

For the CB peak spacing distribution, we thus consider the expression for the ground state energy (9) with constant $\langle M_{ij}\rangle$ and $\langle N_{ij}\rangle$. The problem is still complicated because of the interplay of the fluctuations in the single-particle levels with these constants, which may, for instance, lead to $S=1$ or higher ground state spins.\textsuperscript{30–32} We thus evaluate the distribution of Coulomb blockade peak spacings numerically: Hamiltonians are taken at random in the GUE ensemble, and the lowest energy states for $N-1$, $N$, and $N+1$ particles are obtained by determining in each case the occupation numbers $\{n_{i}\}$ which minimize the expression Eq. (9).

The resulting distribution of CB peak spacings is shown in Fig. 1. We show it for two different strengths of interactions, the case of $r_{s}=1$ and the zero-range interaction limit. The first thing to note is that for both of these values the distribution differs substantially from that in the CI model, Eq. (3), with much less odd/even alternation present. Still, there is a $\delta$ function in the distribution, as is clear in the cumulative distribution functions in panel (c), because the constant residual interaction term gives a rigid shift to the spacing for odd $N$ and standard filling, Eq. (16). The origin of the spacing in these plots is $\Delta^{2}E_{GUE}$+$\langle M_{i+}\rangle$. This is the mean spacing for adding a particle into the next available state neglecting all spin/filling effects; it thus corresponds to the classical charging energy $e^{2}/C$.

The fluctuations of the peak spacings are smaller in this constant exchange model than in the CI model. This is natural since the even and odd parts of the distribution are brought closer together. The values are $\text{rms}(\Delta^{2}E_{N})/\Delta =0.58$ in the CI model and 0.28 or 0.24 in the present large dot case for $r_{s}=1$ or zero-range interaction, respectively.

**IV. FLUCTUATIONS OF RESIDUAL INTERACTIONS**

In order to evaluate the statistical effect of fluctuating residual interactions, we need to find the distribution of the $M_{ij}$ and $N_{ij}$. We will show that in the limit of large but not infinite $k_{F}L$, the $M$’s and $N$’s are Gaussian distributed and uncorrelated.

The first step is to evaluate the variance of the $M$’s and $N$’s. Using the random matrix ensemble introduced in Sec. II, we find to a good approximation

$$\text{var}(M_{i+})=\frac{8}{\pi A^{2}}\int_{\pi L}^{2k_{F}-\pi LL} dq \frac{V_{TF}(q)^{2}}{q(2kL)^{2}-(qL)^{2}}$$

$$\approx \frac{1}{\pi A^{2}} \ln(kL) \left[ 2\hat{V}_{TF}^{2}(0)+\hat{V}_{TF}^{2}(2k) \right], \quad (23)$$

\begin{align}
\text{var}(N_{i+}) & \approx \frac{2}{\pi A^{2}} \int_{\pi L}^{2k_{F}-\pi LL} dq \frac{[\hat{V}_{TF}(q)+\hat{V}_{TF}(2k)]^{2}}{q(2kL)^{2}-(qL)^{2}}, \\
\langle M_{i+}N_{i+}\rangle & - \langle M_{i+}\rangle \langle N_{i+}\rangle \\
& \approx \frac{4}{\pi A^{2}} \int_{\pi L}^{2k_{F}-\pi LL} dq \frac{[\hat{V}_{TF}(q)+\hat{V}_{TF}(2k)]}{q(2kL)^{2}-(qL)^{2}}; \quad (24)
\end{align}

FIG. 1. The probability density of CB peak spacings in the large dot limit ($k_{F}L \to \infty$). The total distribution (solid) as well as that for $N$ even (dashed) and odd (dotted) is given for two strengths of the interactions: (a) $r_{s}=1$ and (b) the zero-range interaction limit. The presence of a $\delta$ function in the distribution is particularly clear in the cumulative distribution functions (the integral of the probability density) shown in (c). Insets show the probability of occurrence of ground state spins in the two cases. The spacing is in units of the mean level separation $\Delta$, and the origin corresponds to the classical spacing $e^{2}/C$.

Details for the case of $\text{var}(M_{i+})$ are given in the Appendix. The diagonal part of the direct residual interaction has an
extra contribution because of the additional way in which the wave functions can be paired:

$$\text{var}(M_{ij}) = 2\text{var}(M_{i+j}) + \frac{8}{\pi a^2} \int_1^\infty \frac{f_{2x-\pi/4} dq}{q}$$

$$\times \left( \hat{V}\hat{V} q \left[ \hat{V}\hat{V}(0) + \hat{V}\hat{V} \left( \sqrt{(2k^2-q^2)} \right) \right] \right) \left( (2k^2)-(qL)^2 \right).$$

(26)

In the zero-range interaction limit, the expressions for the variance of the \( M \)'s and \( N \)'s simplify considerably, as for the mean values given in Eq. (20). In this case, we find

$$\text{var}(M_{ij}) = \text{var}(N_{i+j}) = \frac{3\Delta^2}{4\pi} \frac{\ln(kL)}{(kL)^2} (1 + 3\delta_{ij}).$$

(27)

Note that the decay of the wave function correlations appearing in the variance produces a factor of \( 1/kF L \) in the rms compared to the mean. The \( \ln(kL) \) factor is special to two dimensions; it comes from the \( 1/kL \) decay of the wave function correlator in this case. Thus in the large dot limit \( kF L \gg 1 \), the distributions of the \( M \)'s and \( N \)'s are narrowly peaked about their mean values. This is the justification for ignoring these fluctuations to a first approximation in the last section.

Higher moments of the distribution involve coupling between more pairs of wave functions. The non-Gaussian part of the distribution is described by the cumulants, in which all of the wave functions are coupled in ways not present in the lower moments. In this case, the spatial integrals will have more restrictions coming from the wave function correlation (13); these restrictions will in turn produce additional factors of the small parameter \( 1/kF L \). Thus in the large dot limit all higher moments can be neglected. In the case of third and fourth moments this can be easily verified by explicit calculation. Likewise it is straightforward to check that cross correlations among the \( M \)'s and \( N \)'s do not need to be considered, except for the obvious correlation between \( M_{ij} \) and \( N_{ij} \) which are equal in the zero-range interaction limit. We shall return to this point in Sec. VII. In this section, however, we shall either take \( M_{ij} = N_{ij} \) in the zero-range interaction limit, or neglect this correlation for \( r_i = 1 \), and therefore take the \( M_{ij} \) and \( N_{ij} \) to be uncorrelated random Gaussian variables with mean and variance given above.

We now use the statistics of the \( M_{ij} \), \( N_{ij} \), and single-particle levels (assumed to follow the random matrix GUE ensemble as in Sec. III) to deduce the statistics of the CB peak spacings. It is clear from the expressions for the spacing in the simple up-down filling case, Eq. (15), that sums such as \( \Sigma_j M_{ij} \) will be important. Because of the saturation of the fluctuations, item (iii) in the random matrix model of Sec. II, the variance of such a sum involves only levels within \( E_{TF} \) of each other. In calculating \( \text{var}(\Sigma_j M_{ij}) \), then, the sum should be taken over of order \( kF L \) independent \( M \)'s:

$$\text{var} \left( \Sigma_j M_{ij} \right) \propto \frac{\ln(kF L)}{kF L} \Delta^2.$$

(28)

The variance of other sums over the residual interaction terms will have the same dependence on \( kF L \).

Up to a \( \sqrt{\ln} \) factor, we thus expect the contribution of the fluctuations of the residual interactions to the CB peak spacings to be of order \( \Delta / \sqrt{kF L} \) in rms.

V. SCRAMBLING: ADDED ELECTRON CHANGES

CONFINEMENT

As electrons are added to the nanoparticle, the mean field potential which confines the electrons changes. This causes a change in the single-particle levels, whose energies appear in the expression for the ground state energy Eq. (9), which then in turn change the CB peak spacing. Because we consider a chaotic quantum dot, the change in the energy levels will be unpredictable; hence, we call this effect “scrambling.” Note that it is an intrinsic effect connected to the charge of the electron not the geometry or environment of the dot. This effect has been studied previously for diffusive quantum dots in Ref. 29; here we give a derivation directly in the context of ballistic chaotic dots.

We know from the Strutinsky approach (Sec. II) that the change in the single-particle levels should be found from the change in the smooth Thomas-Fermi potential, Eq. (7). So we first evaluate this change in potential, and then use perturbation theory to find the change in the energy levels. Perturbation theory is justified since the resulting shift is smaller than the mean level separation \( \Delta \) by a factor depending on \( kF L \).

A. Evaluation of \( \delta V_{\text{eff}} \)

The generalized Thomas-Fermi problem is specified in terms of the density functional \( F_{\text{GTF}}(n) = E_{\text{TF}}(n) + \nu_{\text{ext}}(n) \) where the first term is the kinetic energy and the second is the potential energy, including the interaction with the external potential, the Coulomb term, and exchange correlation. The effective potential in which the electrons move is defined by

$$V_{\text{eff}}(r) = \delta(V_{\text{ext}} + V_{\text{Coul}}) \delta E_{\text{TF}}(n_{\text{GTF}}).$$

(29)

The Thomas-Fermi density is found by solving

$$\frac{\delta F_{\text{GTF}}}{\delta n}(n_{\text{GTF}}) = \mu_{\text{GTF}}.$$

(30)

where \( n(r) \) is the electronic density and \( \mu_{\text{GTF}} \) is found such that \( \int n(r) \text{d}r = N \).

Let \( n_{\text{GTF}} = n_{\text{GTF}}(N+1) - n_{\text{GTF}}(N) \) be the change in the Thomas-Fermi density when one electron is added to the system. Bearing in mind that Eq. (30) is a classical-like equation for which \( \delta n_{\text{GTF}} \) is small, one can write

$$\int d\mathbf{r}' \frac{\delta^2 E_{\text{TF}}}{\delta n^2}(n_{\text{GTF}}(\mathbf{r}, \mathbf{r}')) \delta n_{\text{GTF}}(\mathbf{r}')$$

$$+ \int d\mathbf{r}' \frac{\delta V_{\text{eff}}}{\delta n}(\mathbf{r}, \mathbf{r}') \delta n_{\text{GTF}}(\mathbf{r}') = \delta \mu_{\text{GTF}}.$$

(31)
with \( \delta \mu_{\text{GTF}} \) such that \( \int d\mathbf{r} \delta n_{\text{GTF}}(\mathbf{r}) = 1 \). In two dimensions, the Coulomb energy of the added charge will dominate any variations in the kinetic energy. One can therefore write the variation of the density as \( \delta n_{\text{GTF}} = \delta n_{\text{GTF}}^0 + \delta n_{\text{GTF}}^1 + \cdots \) with \( \delta n_{\text{GTF}}^0 \) the solution of the electrostatic problem

\[
\delta \mu^0 = e^2 \int d\mathbf{r}' \frac{\delta n_{\text{GTF}}^0(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|},
\]

where we have assumed that the quantum dot is isolated.

At this level of approximation, the variation of the potential inside the dot is just the constant \( \delta \mu^1 \), which will just shift the one particle energies \( \epsilon_i \) but not give rise to any fluctuation. One should keep in mind that Eq. (30) applies only at places where \( n_{\text{GTF}}(\mathbf{r}) \neq 0 \). Therefore the boundaries of the effective potential can be affected, in a way that depends, of course, largely on the external confining potential, and in particular on the second derivative of this latter at the boundary. If the curvature is weak, fairly large displacements of the boundary can occur, which could play a significant role.

If, on the other hand, one assumes a billiardlike confining potential, the only source of modification of the effective potential comes from \( \delta n_{\text{GTF}}^1 \), which is obtained through the equation

\[
\int d\mathbf{r} \frac{\delta^2 \mathbf{V}_{\text{TF}}}{\delta n^2}(\mathbf{r}, \mathbf{r}') \delta n_{\text{GTF}}^0(\mathbf{r}') + \int d\mathbf{r}' \frac{\delta \mathbf{V}_{\text{eff}}}{\delta n}(\mathbf{r}, \mathbf{r}') \delta n_{\text{GTF}}^1(\mathbf{r}') = \delta \mu^1.
\]

Noting that \( \int (\delta \mathbf{V}_{\text{eff}}/\delta n) \delta n_{\text{GTF}}^1 \), is, up to an inessential additive constant, the variation \( \delta \mathbf{V}_{\text{eff}} \) of the effective potential we are interested in, and that

\[
\frac{\delta^2 \mathbf{V}_{\text{TF}}}{\delta n^2}(\mathbf{r}, \mathbf{r}') = \nu^{-1} \delta \mathbf{V}(\mathbf{r} - \mathbf{r}'),
\]

we get

\[
\delta \mathbf{V}_{\text{eff}}(\mathbf{r}) = -\frac{\Delta A}{2} \left[ \delta n_{\text{GTF}}^0(\mathbf{r}) - \delta n_{\text{GTF}}^0(\mathbf{r}') \right] = -\Delta \delta \mathbf{V}(\mathbf{r}),
\]

where \( \delta \mathbf{V} \) is a smooth function of order 1 (\( A \) is the area of the dot and the factor of 1/2 accounts for the spin degeneracy). Note that we choose \( \delta \mathbf{V}_{\text{eff}} \) with mean value zero. This ensures that there is no change in the mean energy levels; any such change in the mean should be incorporated in the charging energy.

### B. Consequences for the peak spacing distribution

The variation of the effective potential induces a change of the one particle energies. We let \( \epsilon_N \) denote the single-particle energy of the \( N^{th} \) electron, whatever orbital it is in (for \( N \) even and simple up/down filling, for instance, \( \epsilon_N = \epsilon_{N/2} \)). In perturbation theory, \( \delta \epsilon_j = \delta \epsilon_j^{(1)} + \delta \epsilon_j^{(2)} + \cdots \) with

\[
\delta \epsilon_j^{(1)} = \langle \psi_j | \delta \mathbf{V}_{\text{eff}} | \psi_j \rangle
\]

\[
\delta \epsilon_j^{(2)} = \sum_{k \neq j} \left| \langle \psi_j | \delta \mathbf{V}_{\text{eff}} | \psi_k \rangle \right|^2 \epsilon_j - \epsilon_k.
\]

When going back to the inverse compressibility, one should bear in mind, however, that the variation of \( \mathbf{V}_{\text{eff}} \) when going from \( N-1 \) to \( N \) electron is the same as when going from \( N \) to \( N+1 \). As a consequence, it can be checked easily that the linear variation \( \delta \epsilon_j^{(1)} \) cancels for all levels except the two top ones. At this level of approximation one therefore gets

\[
\Delta^2 \epsilon_N = \Delta^2 \epsilon_N^{\text{CI}} + \Delta^2 \epsilon_N^{\text{scramb}},
\]

where \( \Delta^2 \epsilon_N^{\text{CI}} \) is the constant interaction model result given in Eq. (3), and

\[
\Delta^2 \epsilon_N^{\text{scramb}} = \delta \epsilon_{N+1} + \delta \epsilon_N + \sum_{j=1}^{N-1} \delta^2 \epsilon_j
\]

is the correction due to scrambling. The levels used for \( \Delta^2 \epsilon_N^{\text{CI}} \) are those for the \( N \)-electron \( \mathbf{V}_{\text{eff}} \). Throughout this scrambling argument we assume that the filling of the levels does not change.

#### First order terms

The mean of the first order terms is zero by construction. The variance of \( \delta \epsilon_j^{(1)} \) is

\[
\text{var}(\delta \epsilon_j^{(1)}) = \Delta^2 \int d\mathbf{r} d\mathbf{r}' |\langle \psi_j(\mathbf{r}) \psi_j^*(\mathbf{r}') \rangle|^2 \delta \mathbf{V}(\mathbf{r}) \delta \mathbf{V}(\mathbf{r}').
\]

Using the wave function correlations (13) and approximating

\[
J_0^2(k_F |\mathbf{r} - \mathbf{r}'|) \approx 1/(\pi k_F |\mathbf{r} - \mathbf{r}'|)
\]

plus oscillating terms \( (k_F |\mathbf{r} - \mathbf{r}'| > 1) \), we find

\[
\text{var}(\delta \epsilon_j) = \frac{\Delta^2}{k_F L}.
\]

The prefactor depends on the geometry of the dot under consideration:

\[
\alpha = \frac{1}{\pi} \int d^2 u d^2 u' \frac{1}{|\mathbf{u} - \mathbf{u}'|} \delta \mathbf{V}(\mathbf{u}) \delta \mathbf{V}(\mathbf{u}')
\]

is a dimensionless quantity of order 1 (note \( L = r/L \) where \( L = \sqrt{A} \). For a circular dot, for instance, with diffuse boundary scattering to make it chaotic, \( \Delta \mathbf{V} = [(1 - (r/R)^2)^{-1/2} - 2]/4 \) where \( R \) is the radius; in this case, \( \alpha = 0.017 \).

Bearing in mind that \( k_F L = \sqrt{2} \pi N \) (taking into account the spin degeneracy), we see that the typical amplitude of the variation of the \( \delta \epsilon_j \) scales as \( N^{-1/4} \) times the mean level spacing. This may, in fact, be numerically significant for experimental dots—for a circular dot with 100 electrons, for instance, the magnitude would be \( \approx 0.05 \Delta \).

#### Second order terms

The variance of the sum \( \sum_{j=1}^{N-1} \delta^2 \epsilon_j \) can be evaluated along the same lines. Since it turns out to be parametrically smaller, we will only sketch the argument.

Approximating the wave function correlations as for Eq. (41), we find that the magnitude of the off-diagonal matrix element is
The fluctuation of the energy levels is given by the difference between the level spacing and the classical level spacing. The difference is removed by the variation of the top levels, and this adds drastically modified the resulting distribution of CB peak spacings. Figure 2 shows the resulting distribution of CB peak spacings. The fluctuations act to smooth out all the sharp features of the kF L→∞ case: the δ function becomes a finite peak, and the discontinuity at the origin is removed. The difference between the N even and odd cases is quite clear in the r s = 1 case—even for the relatively small N = 80 (for which 1/√kF L = 0.21). For N even, the large spacing tail coming from the level-spacing distribution (Wigner surmise) survives even for less than 100 electrons. When the interactions are stronger, as in the zero-range case shown, even and odd distributions are much more similar. There is still a discernible difference which is, however, compatible with experimental data.\cite{17,18}

The fluctuation of the residual interaction terms and the scrambling of the top levels lead, of course, to an increase in the fluctuation of the spacing. We now find rms(Δ 2E N)/Δ = 0.33–0.32 for r s = 1 and N = 80–200 compared with 0.28 in the absence of such fluctuations.

VII. BEYOND THE GAUSSIAN MODEL

Our treatment of the fluctuations of the residual interactions in the previous sections relies on a certain number of assumptions. We have for instance neglected correlations among the M ij and N‘ i,j ‘ except for the obvious M ij = N ij for a zero-range interaction) when we know, for instance, that the correlation coefficient of M ij with N ij is of order 1 even in the infinite size limit. Similarly, we have assumed the absence of correlation between these residual interaction terms and the one particle energies ε i. Furthermore, we have used expressions such as Eqs. (23)–(26) which are derived in the large kF L limit. To check that these assumptions do not drastically modify the resulting distribution of CB peak spacings including the scrambling effect on the top two levels as well as the fluctuations in the residual interactions. The total distribution (solid) as well as that for N even (dashed) and odd (dotted) is given for two sizes of quantum dot (N = 200, left column or 80, right column) and two strengths of interaction (r s = 1, top row, or the case of a zero-range interaction, bottom row). All of the features are smoothed compared with Fig. 1. Note that the clear odd/even effect for r s = 1 and the relatively small N = 80 remains discernible even for the stronger zero-range interaction. The spacing is in units of the mean level separation ∆, and the origin corresponds to the classical spacing e 2/C.

FIG. 2. The probability density of CB peak spacings including the scrambling effect on the top two levels as well as the fluctuations in the residual interactions. The total distribution (solid) as well as that for N even (dashed) and odd (dotted) is given for two sizes of quantum dot (N = 200, left column or 80, right column) and two strengths of interaction (r s = 1, top row, or the case of a zero-range interaction, bottom row). All of the features are smoothed compared with Fig. 1. Note that the clear odd/even effect for r s = 1 and the relatively small N = 80 remains discernible even for the stronger zero-range interaction. The spacing is in units of the mean level separation ∆, and the origin corresponds to the classical spacing e 2/C.

\begin{equation}
|\langle i | \delta V_{\text{eff}} | j \rangle | ^2 = \begin{cases} 0 & \text{if } \delta k L > 1, \\
\alpha \Delta^2 / \pi k F L & \text{if } \delta k L < 1,
\end{cases}
\end{equation}

where \( \delta k = k_i - k_j \). This result is now inserted in the expression (37) for the energy shift \( \delta e_i^{(2)} \) and then for the inverse compressibility Eq. (39). Assuming the matrix elements \( \langle i | \delta V_{\text{eff}} | j \rangle \) are Gaussian distributed independent variables, we express the variance of the second-order scrambling corrections \( \sigma_2^2 \) in terms of the variance of a single matrix element. The number of independent matrix elements involved is not the total number of electrons \( N \) appearing in the sum in Eq. (39) but rather is reduced to the number \( N_c = k_F L \) because of the saturation of the fluctuations of the levels, point (iii) in the random matrix criteria of Sec. II. The resulting estimate is

\begin{equation}
\sigma_2^2 \propto k_F L \left( \frac{\Delta}{k_F L} \right)^2
\end{equation}

which is parametrically smaller than the first-order result (41).

We therefore see that the effect of scrambling is dominated by the variation of the top levels, and that this adds contributions to the peak spacing fluctuations which while parametrically small on the scale of \( \Delta \) could be numerically important for dots which are not too large.

VI. NUMERICS

In order to evaluate the magnitude of the effects discussed in the last two sections, we turn to numerical evaluation. We consider a collection of \( E_{\text{th}} / \Delta \) levels with GUE energies and Gaussian residual interaction terms. It is a simple matter to incorporate the scrambling of the top two levels: in random matrix theory, the change in an energy level upon varying a parameter is uncorrelated with its value.\cite{48,49} so we simply choose random \( \delta e_N \) and \( \delta e_{N+1} \) from a Gaussian distribution with variance Eq. (41). Then we fill the levels with \( N-1, N \), and \( N+1 \) particles successively, and find the occupation numbers \( \{ n_{i\alpha} \} \) minimizing the energy Eq. (9) in each case.

VII. BEYOND THE GAUSSIAN MODEL

Our treatment of the fluctuations of the residual interactions in the previous sections relies on a certain number of assumptions. We have for instance neglected correlations among the \( M_{ij} \) and \( N'_{i,j'} \) (except for the obvious \( M_{ij} = N_{ij} \) for a zero-range interaction) when we know, for instance, that the correlation coefficient of \( M_{ij} \) with \( N_{ij} \) is of order 1 even in the infinite size limit. Similarly, we have assumed the absence of correlation between these residual interaction terms and the one particle energies \( \varepsilon_i \). Furthermore, we have used expressions such as Eqs. (23)–(26) which are derived in the large \( k_F L \) limit. To check that these assumptions do not drastically modify the resulting distribution of CB peak spacings.

FIG. 2. The probability density of CB peak spacings including the scrambling effect on the top two levels as well as the fluctuations in the residual interactions. The total distribution (solid) as well as that for N even (dashed) and odd (dotted) is given for two sizes of quantum dot (N = 200, left column or 80, right column) and two strengths of interaction (r s = 1, top row, or the case of a zero-range interaction, bottom row). All of the features are smoothed compared with Fig. 1. Note that the clear odd/even effect for r s = 1 and the relatively small N = 80 remains discernible even for the stronger zero-range interaction. The spacing is in units of the mean level separation ∆, and the origin corresponds to the classical spacing e 2/C.
ings, we shall in this section implement the random matrix ensemble described in Sec. II [paragraph containing Eq. (12)] by directly performing numerical Monte Carlo calculations.

To implement the constraints of our random matrix model, we start by considering the system to be a square of size $L$, for which we can use a plane wave basis

$$|n_x,n_y\rangle = L^{-2} \exp[i 2\pi n_x x/L] \exp[i 2\pi n_y y/L]$$

ordered by increasing momentum. In this basis, we write the one particle Hamiltonian as $H = D + V$ where $D$ is diagonal with $D_{ii} = i$. $V$ is taken to be a banded random Hamiltonian with independent complex matrix elements which are Gaussian distributed with variance $v_{ij}^2 = a(i) \exp[ -b(i)|i-j|]$. For $a(i)$ in the correct range $^{50}$ such an ensemble yields (1) eigenvalues with a mean spacing close to one, (2) local fluctuations which are typical of the GUE, and (3) eigenfunctions which are localized in energy. The localization in energy can be characterized by the inverse participation ratio in the plane wave basis $I_j = \sum_{n_x,n_y} |\langle n_x,n_y|\epsilon_j\rangle|^4$. Our requirement Eq. (12) implies the constraint

$$I_j^{-1} = (3/4)(k_f L).$$

In practice, we use a simple algorithm to tune the value of $a(i)$ and $b(i)$ in such a way that this constraint holds. Diagonalizing the random matrix specified leads to eigenvalues and eigenvectors which are then used to evaluate ground state energies for different number of particles via Eq. (9), and hence arrive at the CB peak spacings in this model.

In this approach, deviations from the wave function correlations (13) required by normalization are naturally present. Furthermore, correlation between eigenvalues and eigenfunctions is present as well. Of course, in a real physical system, these effects would be automatically included because of the scattering at the boundary of the system—the transit time $\tau$, for instance, gives both the scale over which fluctuations saturate $(h/\tau)$ and the length scale at which the Bessel function correlations become suspect $(v_s \tau)$. We believe that our random matrix model includes the main features of these effects in a generic way.

Figure 3 shows numerical results for this model. We consider only the smaller value of $N, N=80$, because the effects are expected to be only significant for smaller quantum dots. On the other hand, we present results both with and without the scrambling effect of Sec. V, and for different interaction strengths $-r_s$ corresponds to a typical value for a GaAs quantum dot while the zero-range interaction case corresponds to the maximum effect possible within a perturbative approach to the interactions. When these results are compared to those of Fig. 2, we see that there is a small but noticeable effect. This is highlighted in the quantile-quantile plots in the insets to Fig. 3 where an ordered set of RMT spacings is plotted against a similar set for the Gaussian approximation. Insets: Quantile-quantile plots of the random matrix model versus the Gaussian model; note the differences at large spacings. Throughout, the spacing is in units of the mean level separation $\Delta$, and the origin corresponds to the classical spacing $e^2/C$.

**VIII. CONCLUSIONS**

In this paper we have studied the Coulomb blockade peak spacings for large but not infinite quantum dots in the case that the interactions can be treated perturbatively and the single-particle properties described by random matrix theory. Motivated by the Strutinsky approach to finite Fermi systems, $^{38-40}$ we use an expression for the ground state energy [Eq. (9)] which includes the charging energy, the single-particle quantization effects, and the residual (screened) interactions treated in first order (Hartree-Fock).

We presented results in four cases.

1. In the limit $k_f L \rightarrow \infty$, fluctuations of the residual interactions are negligible. The CB peak spacing distribution has a discontinuity at $e^2/C$ and a $\delta$ function because of the odd
The distribution is very different from that predicted by the simple CI model which neglects some terms of order $\Delta$ while keeping others.

(2) Fluctuations in the residual interactions contribute a term typically of order $\Delta/\sqrt{k_F L}$ to the CB peak spacings.

(3) The addition of an electron in the CB process changes the confinement potential in the quantum dot and so changes the single-particle properties. This scrambling effect also contributes a term typically of order $\Delta/\sqrt{k_F L}$ to the CB peak spacings. Combined with point (2), we find that the sharp features of the infinite dot limit are rounded out (Fig. 2).

(4) We took a first step towards investigating corrections to the Gaussian uncorrelated model for the fluctuations of the residual interactions. We found that the large spacing part of the distribution is somewhat underestimated in the Gaussian model.

Before ending this paper, we compare our theoretical distributions to those obtained experimentally.\textsuperscript{13–18} The observed distribution has little structure, being Gaussian near its peak but with longer tails for both larger and smaller spacings. The width of the distribution is contentious: early work claimed that the width scaled with the charging energy,\textsuperscript{13,14} but this has not been verified by more recent experiments.\textsuperscript{15–18} In the more recent experiments, the width is clearly related to the mean level separation $\Delta$; it is $\sim 0.3–0.5 \Delta$ in Refs. 16-18 and $\sim 1.75 \Delta$ in the Si dots of Ref. 15.

The most important conclusion of our work is that it is not sufficient to resort to a non-Fermi-liquid description of the interacting electrons to obtain peak spacing distributions similar to the experiments. Indeed, looking at the lower panels of Fig. 2, one observes that the zero-range potential Eq. (5) produces distributions whose shape resembles the experimental ones. Thus, this zero-range interaction is stronger than the RPA screened interaction, it remains "small" in the sense that treating it in perturbation theory is adequate. Therefore, even if, as we discuss below, not all aspects of these CB experiments are elucidated, it is within the relatively conservative framework of Fermi-liquid theory that they should be addressed, and that they will presumably be solved in the near future.

On the other hand, the specific model we consider—electrons confined within a chaotic dot with RPA screened interactions—does not provide an adequate quantitative description, even though the resulting distributions look significantly more similar to the experiments than the benchmark CI model. Indeed, looking at the top left panel of Fig. 2, for instance, $(r_x=1, N=200, \text{ which is typical of the actual dots})$, one observes features which are clearly not seen in the experiments. To start with, the magnitude of the fluctuations is actually less than in the CI model (0.32 versus 0.58 for $N=200$) and so differs from experiment. Moreover, one sees a substantial odd/even effect, in particular because of the long Wigner surmise tail present for even spacings.

It should be stressed that large changes in the distribution can be produced by a modest change in the mean values $\langle N_{ij} \rangle - \Delta/2$ and $\langle N_{ij} \rangle$, Eqs. (18)-(20). For instance, the change from 0.3$\Delta$ for $r_x=1$ to 0.5$\Delta$ in the zero-range limit causes the large difference seen in Figs. 2 and 3. The sensitivity to the numerical value of these quantities is sensible since we know that the Stoner instability, in which a spontaneous polarization of the spin of many electrons takes place, occurs for $\langle N_{ij} \rangle = \Delta$. These observations suggest an experiment: placing a good conductor close to the 2DEG in such a way that it can screen the interaction between electrons within the quantum dot should significantly alter the peak spacing distribution. If the auxiliary screening is strong enough, a strong odd/even effect should be visible. A double quantum well structure seems like to a possible setting for such an experiment.

We can divide the potential explanations for the quantitative discrepancy between theory and experiment into basically two categories. The first one, which we cannot absolutely exclude, is that noise in the experiments is corrupting the results.\textsuperscript{16,51} The second is that effects not included here, but interpretable within Fermi liquid theory, dominate. Some candidates are (a) the effect of the changing gate potential on the single-particle levels,\textsuperscript{52,53} this would, however, somewhat contradict the remarkable stability observed in the wave function—(b) boundary effects in the scrambling which need to be considered using a realistic description of the dot, and (c) consequences of non-chaotic dynamics within the dots.\textsuperscript{55,53} Further experimental and theoretical work is needed to discern between these options.

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APPENDIX: VARIANCE OF THE $M_{ij}$

In this appendix we sketch the argument which leads to Eqs. (23)-(26). As in Sec. VII, we assume that the configuration space is a square of size $L$, for which the plane wave basis $|k\rangle = |n_x, n_y\rangle$ introduced in Eq. (45) can be used.

Expanding the eigenfunctions in this basis

$$|\varphi_i\rangle = \sum_k u_{ik}|k\rangle,$$

we can write the $M_{ij}$’s as

$$M_{ij} = \sum_{k_1, k_2, k_3, k_4} v_{ik_1}^* v_{ik_2}^* v_{jk_3} v_{jk_4} \delta_{k_1-k_2-k_3+k_4} \hat{V}_{TF}(k_1-k_2),$$

$$= \sum_q \hat{V}_{TF}(q) W_{iq} W_{jq}^*,$$

where we define

$$W_{iq} = \sum_{k_1-k_2=-q} v_{ik_1}^* v_{ik_2}^*.$$

Our random matrix model (Sec. II) implies
\left\langle v_{i k}, v_{j k}^{*} \right\rangle = \left( k_{F} L \right)^{-1} \delta_{ij} \delta_{k, k_{i}} \quad \text{if} \quad \delta k < \pi / L,

\quad = 0 \quad \text{if} \quad \delta k > \pi / L \quad (A4)

with \( \delta k = |k_{i} - k_{j}| \) and \( k_{i} = \sqrt{2mE/\hbar} \). From this, we deduce

\[
\text{var}(M_{i_{0}j}) = \frac{1}{A} \sum_{q \neq 0} V_{q}^2 \left| \langle W_{qj} \rangle \left/ \langle |W_{qj}|^2 \rangle \right. \right| . \quad (A5)
\]

\( |W_{qj}|^2 \) can be interpreted as \( (2\pi k_{i})^{-2} \) times the area of the intersection of two rings of diameter \( k_{i} \), and width \( 2\pi / L \), centered at a distance \( |q| = q \). Simple geometry therefore gives, for \( 2\pi / L \leq |q| \leq 2k_{i} - 2\pi / L \)

\[
\langle |W_{qj}|^2 \rangle = \frac{4}{(qL)\sqrt{(2k_{i}L)^2 - (qL)^2}}. \quad (A6)
\]

We obtain for \( i = j \)

\[
\text{var}(M_{i_{0}i}) = \frac{8}{\pi A^2} \int_{\pi L}^{2\pi / L} dq \frac{V_{q}^2}{q(2k_{i}L)^2 - (qL)^2}. \quad (A7)
\]

The variance of \( M_{ij} \) and \( N_{ij} \) and the covariance between \( M_{ij} \) and \( N_{ij} \) can be computed along the same lines.

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