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Fluctuations in quantum dot charging energy

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Abstract

We demonstrate that large fluctuations in the charging energy, and in the spacing of Coulomb oscillations, of lateral, GaAs–$Al_xGa_{1-x}As$ semi-conductor quantum dots occur quasi-periodically as a function of externally applied gate voltage V_g. Using density functional theory, self-consistent electronic structure calculations, we show that large fluctuations correspond to gate voltages where strongly scarred states, which are the remnants of periodic orbits in the classical confining potential, become doubly occupied. We calculate the direct Coulomb matrix elements between self-consistent eigenstates and show that (1) diagonal elements are uniformly greater than off-diagonal elements and (2) diagonal elements for strongly scarred states, due to their one dimensional nature, are significantly larger than for more "chaotic" states, whose probability density is spread more uniformly across the dot. In consequence of the large energy cost of occupying both spin states of a strong scar, these states tend to remain, half filled, at the Fermi surface as V_g and electron number are swept.

Keywords: Quantum dots; Chaos; Scarred orbits

Inquiry into the interplay of disorder and interaction is at the forefront of condensed matter physics. Quasi-zero dimensional systems, i.e. quantum dots, insofar as several of the parameters of disorder and interaction are experimentally manipulable, form an important subfield of that inquiry. Of particular interest lately is the evolution of the ground state energy of a disordered or chaotic quantum dot as the electron number N is varied [1]. Density functional theory (DFT), which is a theory of the ground state of interacting electron systems, is ideally suited for application to this problem.

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The ground state energy is probed via spacings of the Coulomb oscillations of tunneling transport through the dot as an external gate voltage V_g is varied. The zeroth-order theory from which deviations are measured [2] includes interaction only through a smoothly varying capacitance and disorder through a single Wigner–Dyson non-interacting spectrum. By contrast, we have recently developed efficient algorithms which allow the computation of the self-consistent electronic structure of lateral, GaAs–$Al_xGa_{1-x}As$ quantum dots for $N \sim 150$ [3]. Further, we calculate the total free energy of the interacting dot-leads-gates system without recourse to capacitance parameters.

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Here we present results of our recent calculation of the electronic structure of the dots studied in Ref. [1] (see Ref. [3] for details). We substantiate the existence of a self-consistent level attraction at the Fermi surface [3,4], which is a manifestation of the fact that the direct Coulomb matrix element between electrons in different spatial states is in general weaker than between electrons in the same spatial state with different spins (we refer to this latter case as "self-interaction"). We show that quasi-periodic fluctuations in the charging energy E_C of the dot result from the filling of the second spin state of strongly scarred spatial states. These scarred states are the remnants of periodic orbits in the dot and, as such, are essentially one dimensional and occur periodically as additional nodes are added to the wave function (cf. Fig. 2). Due to their 1-d character their self-interaction is appreciably larger than for states which fill the entire dot (Fig. 3).

The device gate pattern is shown in the inset of Fig. 1. The excess metal in the rightmost gate significantly distorts the dot profile and the resulting eigenstates are chaotic. The gate voltages are set to deplete the 2DEG underneath and form tunnel barriers to the source and drain. The "plunger" gate V_g (inset Fig. 1) is varied in steps of 5 mV over a range such that N changes by about 70 electrons [5]. In the calculation, N is generally not an integer. Its equilibrium value at V_g (denoted simply as N) is determined by setting the dot Fermi energy E_F equal to that of the leads (which is our energy zero).

Calculation of the free energy $F(N, V_g)$ of the interacting dot-leads-gates system is also discussed in Ref. [3]. We *define* the dot charging energy as

$$E_C \equiv [-2F(N,V_g) + F(N+\Delta N, V_g) + F(N-\Delta N, V_g)]/\Delta N^2 \quad (1)$$

that is, the discrete second derivative of F with respect to N, at fixed V_g. $F(N \pm \Delta N, V_g)$ are computed by varying the dot Fermi energy, typically by $\pm 0.2Ry^*$, which results in a ΔN close to unity. Note that E_C is defined at a fixed gate voltage. Given $F(N, V_g)$ it is possible to numerically extract the actual gate voltage spacing ΔV_g between "degeneracy points" where $F(N+1, V_g) = F(N, V_g)$. These are the locations of the Coulomb oscillations. Whereas fluctuations in E_C (Fig. 1) are typically positive relative to a smoothly varying base, fluctuations in ΔV_g are symmetric and resemble the experimental Gaussian shape. Also, relative to fluctuations in E_C, those in ΔV_g are much greater in that the self-consistent energy levels as well as other terms in $F(N, V_g)$ depend, even at fixed N, on V_g. Thus, in Ref. [1], a purely linear term in V_g can be extracted from the total energy and a closed expression for ΔV_g derived only insofar as the capacitance C and the single particle levels (η_k in Ref. [1]) are not themselves functions of V_g. This is, of course, an assumption of the constant interaction (CI) model with which those authors begin.

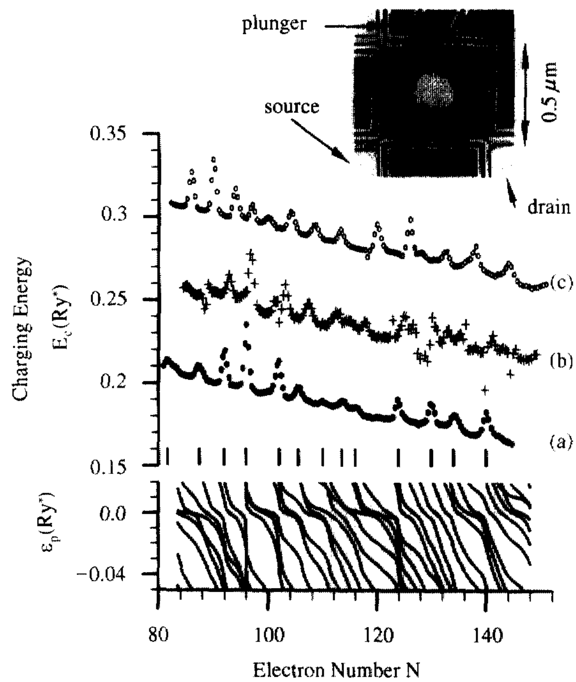

Fig. 1. Wave function moduli squared for $V_g = 1.26\,V$, $N \approx 124$. Other gates set as in curves (a) and (b), Fig. 3. Strong scar at $p = 62$ is at Fermi surface.

Results for $E_C(N)$ are shown in Fig. 1. The most striking feature of the results is the appearance of seemingly quasi-periodic peaks. The precise location of these peaks depends on the gate voltages (other than the plunger) as shown in the difference between curves (a) and (c). Curves (a) and (c) are pure Hartree, whereas curve (b) has identical gate voltages to (a) but also includes exchange

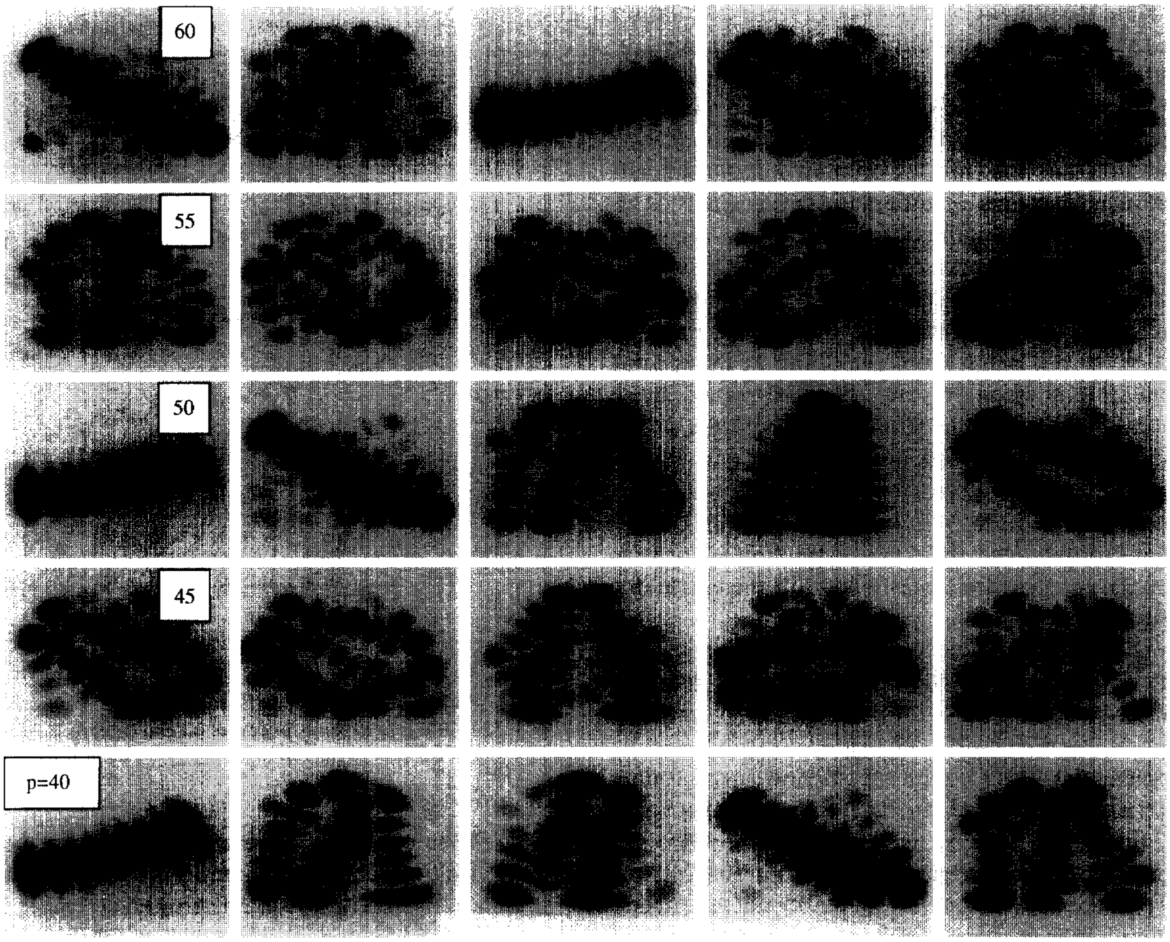

Fig. 2. Direct Coulomb matrix elements (range of scale from ~ 0.18 to ~ 0.3, in Ry*), same parameters as Fig. 1, showing strong interaction between spin pairs ("self-interaction") particularly for strong scars. Scars corresponding to same classical orbit also interact strongly, but interact weakly with other, more chaotic states.

correlation within the local density approximation [6]. The lower panel of Fig. 1 shows the self-consistent level structure $\varepsilon_p(N)$ near E_F, as a function of N, corresponding to curve (a). The levels tend to cluster near E_F and fluctuations in E_C occur when a gap opens up at E_F. Level clustering at E_F occurs when it is energetically favorable to occupy two or more different spatial states as opposed to occupying both spins of a single spatial state. This in turn results from the tendency, alluded to above, for the Coulomb self-interactions to be greater than the interaction between different spatial states.

Fig. 2 shows a sequence, in order of increasing ε_p, of self-consistent states (moduli squared) in the vicinity of the large fluctuation of E_C near $N = 124$, curve (a), Fig. 1. The uppermost filled state, $p = 62$, is a strong remnant of a simple periodic orbit across the dot (compare also $p = 50$). This state reaches the Fermi surface (as N increases) near $N = 116$ and remains there as other states pass through. Over the range from $N = 98$ to $N = 126$ the indicial location of this state changes from $p = 55$ to $p = 62$. This appears to be a general feature of strong scars that their energy tends to rise relative to other states as the number of electrons increases.

We have calculated the matrix of direct Coulomb interactions between self-consistent states. The computation proceeds as follows. Upon obtaining

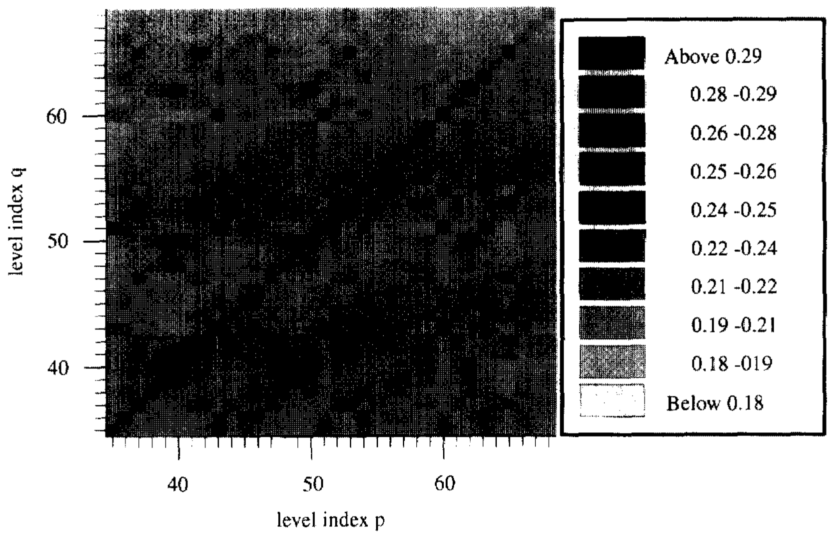

Fig. 3. Upper panel: Charging energy E_C as a function of dot equilibrium electron number. Curves: (a) pure Hartree; (b) exchange-correlation included in LDA, same gate voltages as (a); (c) gate voltages other than plunger different to change shape. All results at $T = 0.1$ K. Lower panel: Kohn-Sham energy levels vs. N near E_F. Total depth of Fermi sea ~ 1.2 Ry*. Fluctuations in E_C correspond to filling of second spin state of strong scars. Inset: gate pattern and typical self-consistent effective 2D potential at 2DEG level.

the self-consistent solution at a given (N, V_g), each eigenstate density, $|\psi_p(\boldsymbol{r})|^2$ is treated as a source term in the solution of Poisson's equation, with the same boundary conditions (e.g. gate voltages) as used in the self-consistent solution. The electrostatic potential for that state alone (note, screening by the *gates* is included automatically) is then computed by solving Poisson's equation and the interactions of that state with all states (including itself) is determined by a quadrature. Note that what we are calculating here is, in many-body language, the *unscreened* (by dot electrons) direct Coulomb matrix element between self-consistent Hartree states with exchange-correlation included via the local density approximation to DFT. This is in contrast to the screened matrix element between bare states, which is often calculated in RPA to determine, among other things, the plasmon modes of the system. Within the context of a mean field theory it is these matrix elements that we are computing which contribute to the total free energy and thereby to the charging energy.

The "Coulomb matrix" for the parameters at which the states in Fig. 2 are computed is shown in Fig. 3. It is immediately evident that, as claimed, the diagonal terms are much greater than the off diagonal terms. Further, the order of magnitude of the terms is roughly given by E_C. Also, the diagonal terms related to scars are significantly greater than those of more "chaotic" states. Finally, families of scars are evident by their off-diagonal couplings since they occupy identical orbits with only a change in the number of nodes.

Heretofore our discussion has been in terms of simple DFT wherein spin degeneracy is assumed. Thus a scarred state which remains near E_F due to a strong Coulomb interaction between up and down spins is, in a more accurate description, a pair of strongly spin-split states which straddle E_F. The lowering of total Coulomb energy by partial occupation of spin degenerate states is, more properly speaking, an indication of Coulomb regulated spin polarization. We have thus found it essential to complement our DFT calculations with full spin density functional theory (SDFT) calculations [6] for selected parameter regions. A portion of the self-consistent spectrum in the SDFT case is shown in Fig. 4. Throughout most of the spectrum the dot has a polarization of ~ 1–3 spins. Once again, when the second spin state of a strong scar passes

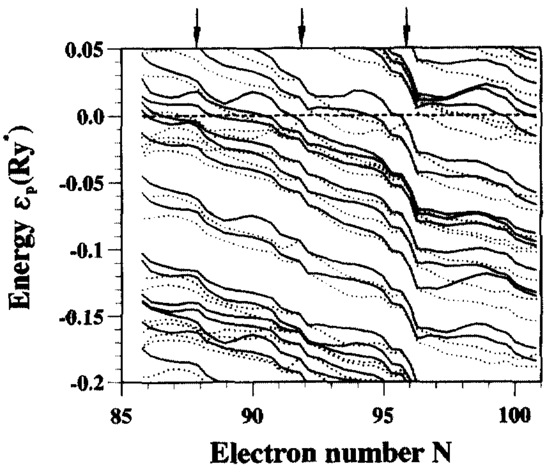

Fig. 4. Results of Kohn–Sham energies from SDFT calculation; solid line (arbitrarily) spin up, dotted line spin down. Arrows indicate locations of fluctuations in E_C, which are predominantly unchanged from simple DFT results. Note at $N = 96$ as strong scar spin up state passes below E_F spin polarization disappears and *all* states become spin degenerate. The same occurs at $N = 92$ however dot remains spin polarized at $N = 87$.

though E_F a large fluctuation in E_C occurs. However now this is observed to cause a complete *collapse* of spin polarization of the dot. The physics is as follows. At E_F the cause of spin splitting is the energy cost to occupy the same spatial state twice, particularly for a strong scar. When finally the changing gate voltage drives the second spin state below E_F it is because it is no longer energetically possible to pull any higher energy state down. The scar thus tends to be the *last* state in a sequence to be joined below E_F by its spin partner. For states not at E_F the origin of splitting is the exchange energy related to the polarization itself [6]. Thus when a strong scar is doubly occupied below E_F all states become exactly spin degenerate, as is seen in Fig. 4 near $N = 96$, which is the location of a major peak in E_C.

Our main conclusions are as follows. Large fluctuations in E_C occur quasi-periodically as V_g and N are varied. The fluctuations in E_C calculated at fixed V_g occur due to the formation of gaps at the Fermi surface and are therefore fluctuations upward from a smoothly varying background. Consideration of the fluctuations at neighboring oscillation gate voltages is necessary to recover the symmetric pattern of fluctuations observed in experiment. The physical origin of the fluctuations is the presence of strongly scarred states in the spectrum. Direct calculation of Coulomb matrix elements shows that all states have larger self-interaction (i.e. between up and down spins of the same spatial state) than their interaction with other states and that strong scars in particular have especially large self-interaction, owing to their essentially one-dimensional nature. This large self-interaction causes the scarred states to dwell at the Fermi surface, with only a single spin state filled, as V_g is varied. When the second spin state is finally filled, a fluctuation in E_C is seen as a shift in the self-consistent energies of all the levels. Calculations employing full spin density functional theory show that peaks in E_C, where the fillings of the second spin state of strong scars occur, are often accompanied by a collapse of spin polarization in the dot.

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[5] Specifically: $V_g \in [-1.0, -1.8]$V, correspondingly $N \tilde{\in} [150, 80]$. Other gate voltages, clockwise from plunger, -1.0, -0.9 and -1.15 V for Fig. 1 curves (a) and (b), -1.0, -1.0, -1.05 V, curve (c). All results here computed at $T = 0.1$ K.
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