Spontaneous Polarization of Quantum Dots

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We have performed spin density functional theory calculations to determine the electronic structure of lateral, $GaAs - AlGaAs$ quantum dots. We have developed a highly efficient, quasi-adiabatic subband method which allows us to solve the Schrödinger equation for dot electron number $N \lesssim 200$. We find that the direct Coulomb energy of putting two electrons into up and down spin of the same spatial state at the Fermi surface, combined with the polarization dependent, exchange-correlation induced splitting of the spin levels, typically exceeds the average single particle level spacing and leads to spontaneous spin polarization of the dot.

1. Introduction

The calculations described here are motivated by recent experimental studies on tunneling transport through lateral semiconductor quantum dots [1,2]. Two significant conclusions of the experiments are that (1) fluctuations in the "charging energy" E_C for adding an electron to the dot, as a function of co-varying gate voltage V_g and electron number N greatly exceed the fluctuations in the dot single particle level spacings despite the predictions of the zeroth order "constant interaction" (CI) model that they should be equal; and (2) within the CI model analysis, spin degeneracy appears to be absent from the effective single particle spectrum.

The principle simplification of the CI model is to treat the Coulomb energy classically by introducing a device capacitance C which is, by hypothesis, independent of N and V_g. Our calculations, within spin density functional theory (SDFT), of the electronic structure of lateral quantum dots allow the determination of the interacting dot-leads-gates system total free energy $F(N, V_g)$ as a function of N and V_g and, in principle, single particle level occupancies (more practically, the temperature), without employing phenomenological capacitances.

The calculations reveal that the dot spectrum is composed of "scarred" wavefunctions, which are quasi-one dimensional states that recapitulate unstable periodic orbits in the classical confining potential, and "chaotic" wavefunctions, which more smoothly occupy the entire dot area. Fluctuations in the charging energy and polarization proceed from fluctuations in the direct and exchange pairwise (screened) interaction between electrons in occupied levels. These interactions are dominated first by whether the two electrons are in spatially distinct states or are in the two spin states of a single spatial wavefunction. Second, the interactions depend strongly on whether the states are quasi-one dimensional scars or else dot-filling, chaotic states.

2. Coulomb oscillations and the constant interaction model

In recent experimental investigations [1,2], Coulomb oscillations in the low temperature, low source-drain voltage tunneling conductance from 2D source, through the dot to 2D drain, as a function of a chosen gate voltage V_g, are employed to study differences in the total interacting free energies $F(N, V_g)$ of the dot-leads-gates system at neighboring values of N. The CI model for relating the gate voltage spacing of Coulomb oscillations and the charging energy derive from the following parameterization of the total energy of the dot

$$E(N, V_g) = \frac{N^2 e^2}{2C} + e\alpha V_g N + \sum_{k=1}^{N} \epsilon_k^0 \qquad (1)$$

where C is the dot self-capacitance, α is the so-called "lever arm" which is the ratio of the dot-gate capacitance to the self-capacitance, and ϵ_k^0 is the dot single particle spectrum. The capacitances and spectrum are assumed to be independent of N and V_g. In eq. 1 we have used the symbol "E" for the energy within the CI model to distinguish it from the total free energy which we calculate microscopically within SDFT.

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Coulomb oscillations [3] occur at values of V_g for which a degeneracy occurs in the total energy evaluated at two neighboring charge states, i.e. $E(N+1,V_g) = E(N,V_g)$. The V_g spacing between two *successive* oscillations is simple to determine from eq. 1

$$e\alpha\Delta V_g = \frac{e^2}{C} + \Delta_N \tag{2}$$

where $\Delta_N \equiv \epsilon^0_{N+1} - \epsilon^0_N$, is the dot single particle level spacing at the Fermi surface. Consequently, assuming C is independent of N and V_g, the observed spacings of Coulomb oscillations in the transport through the dot should reflect directly the single particle level spacings of the dot. These experimental studies involve dots with electron number anywhere from 10^2 to 10^3. Thus precise predictions of the Δ_N values, which depend in addition on crystal imperfections, random impurities, lithographic variations etc., appears infeasible. *Statistical* properties of eigenvalues and eigenfunctions of Hamiltonian systems, on the other hand, have been exhaustively investigated in the context of nuclear excitation energies under the formalism of random matrix theory [4]. Treating the dot spectrum as such a random system, various predictions emerge for the statistical properties of an ensemble of $e\alpha\Delta V_g$ values, notably that the r.m.s. deviation of the distribution should go as $\sim 0.52\Delta$, where Δ (without subscripts) is the mean level spacing. Furthermore, according to eq. 2, if spin degeneracy persists in the dot, an even-odd fluctuation in $e\alpha\Delta V_g$ should exist, reflecting the fact that every other value of Δ_N should be zero.

In contrast to these predictions, the experiments have shown that fluctuations in gate voltage spacings are as much as a factor of five greater than the prediction and no even-odd asymmetry, within experimental error, can be discerned.

3. Self-consistent calculation

We have described the calculation of the electronic structure of lateral, *GaAs – AlGaAs* quantum dots, within the effective mass approximation and within the local density approximation to density functional theory at length [5]. In a more recent publication [6] we have discussed the modifications to those calculations resulting from spin dependent exchange-correlation potentials. To briefly recapitulate, Poisson's equation is solved on a 3D mesh which incorporates the (conduction) band offset between the *GaAs* and the *AlGaAs* of the heterostructure (material varies only in the z-direction, i.e. the growth direction). Boundary conditions on the surface are fixed by the gate pattern and the various applied gate voltages. The density is determined by a solution of Schrödinger's equation in the dot and a Thomas Fermi approximation for the electrons in the leads. We employ an adiabatic treatment of the quantum description which takes advantage of the strong confinement of electrons to the heterostructure interface. That is, we solve Schrödinger's equation in the z-direction at each point in the x-y plane, resulting in subband energies $\epsilon_n(x,y)$ and wavefunctions $\xi^{xy}_n(z)$ which depend parametrically on x and y. Generally we employ only the lowest subband (the structures which we model are all in the electric quantum limit) whose energy becomes an effective potential in the x-y plane. In comparison to solving a full three dimensional Schrödinger equation, which would require diagonalization of a sparse matrix of order $\sim 10^6$, this method, combined with an eigenfunction expansion for the 2D states in the dot, is highly efficient. The solution of a dot state with $N \sim 200$ and one set of gate voltages typically consumes less than one hour on a DEC-ALPHA workstation.

Calculation of the total free energy from the results of the self-consistent electronic structure requires that we include the work provided by the power supplies connected to leads and gates [5], as well as double counting and exchange correlation terms in the energy. The free energy functional is thus

$$\begin{aligned} F(N,V_i) &= \sum_p f_p\epsilon_p - \frac{1}{2}\int d\mathbf{r}\phi(\mathbf{r})\rho(\mathbf{r}) \\ &+ \int d\mathbf{r}[n_\uparrow(E^\uparrow_{xc}(\mathbf{r}) - v^\uparrow_{xc}(\mathbf{r})) \\ &+ n_\downarrow(E^\downarrow_{xc}(\mathbf{r}) - v^\downarrow_{xc}(\mathbf{r}))] \\ &+ \frac{1}{2}\int d\mathbf{r}\phi(\mathbf{r})n_+(\mathbf{r}) - \frac{1}{2}\sum_i Q_iV_i \end{aligned} \tag{3}$$

where the (multiple) gate voltages are V_i, the Kohn-Sham level energies are ϵ_p, with occupancies f_p, the electrostatic potential is $\phi(\mathbf{r})$, total conduction band electron density is $\rho(\mathbf{r})$, $E^{\uparrow(\downarrow)}_{xc}(\mathbf{r})$ is the exchange-correlation energy per particle for up (down) spin, and $v^{\uparrow(\downarrow)}_{xc}(\mathbf{r})$ is the corresponding potential. Also, $n_+(\mathbf{r})$ is the density of positively charged donors, Q_i and V_i are the charge and voltage respectively of gate i.

We *define* the dot charging energy as

$$E_C \equiv [-2F(N,V_g)+F(N+\Delta N,V_g)+F(N-\Delta N,V_g)]/\Delta N^2 \tag{4}$$

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 and that here we are assuming that only a single gate voltage, V_g, will vary.

4. Results

In this report we present results for a device which was used in reference [1]. As in the experiment, we sweep a gate voltage and allow N to change so that the dot remains in equilibrium with the leads. Thus V_g and N change simultaneously. The gate pattern and a typical effective 2D potential is shown in the inset to Fig. 1. Results for $E_C(N)$ and the spectrum near the Fermi surface, in the case where complete spin degeneracy is assumed, are also 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-correlation. 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.

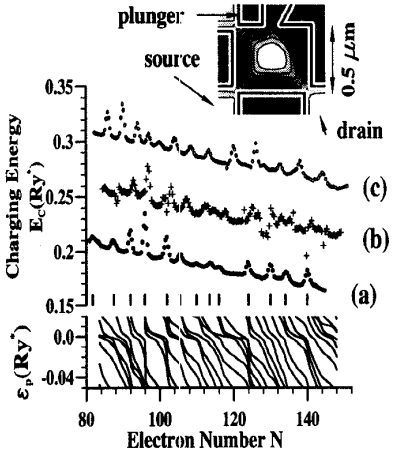

Figure 1: Upper panel: Charging energy E_C as a function of dot equilibrium electron number. Curves: (a) pure Hartree; (b) exchange-correlation included in local density approximation (LDA), same gate voltages as (a); (c) gate voltages other than plunger different to change shape. All results at $T = 0.1\ K$. Lower panel: Kohn-Sham energy levels vs. N near E_F. Total depth of Fermi sea $\sim 1.2\ Ry^*$. 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 fluctuations in E_C indicated in Fig. 1 cause the distribution of Coulomb oscillation gate voltage spacings to be significantly broader than the distribution of single particle level spacings, in agreement with experimental findings.

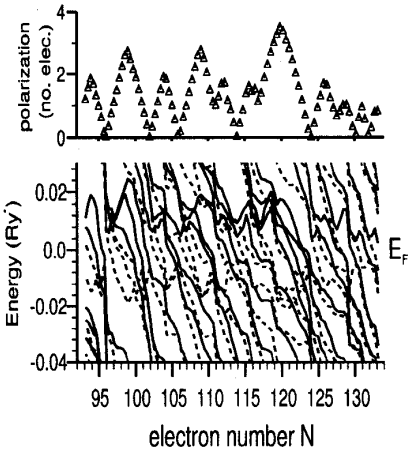

Figure 2: Spectrum and polarization of quantum dot within SDF theory. Note that while spontaneous polarization due to the unfavorable energy condition of double occupancy is to be expected, the tendency for the polarization to periodically return to zero is unexpected. In the lower panel E_F is the zero of energy.

When spin is incorporated into the calculation, the clustering of levels at E_F is replaced by spin splitting. In Fig. 2 we show the spectrum and the total dot polarization P as a function of N (again V_g co-varies). The polarization is adjusted so as to always be positive. In reality the direction of the total spin will probably be determined by spin orbit coupling. Fig. 2 shows that P tends to increase from zero as an odd electron is added to a spin-degenerate state. For example, near $N = 96$ if we take the dashed lines as the "up" spins, filling of $49_\uparrow$ results in non-zero polarization which creates a difference in the exchange energy experienced by up and down electrons and thereby causes all states to spin split. In addition, the charge distribution of $49_\uparrow$ is imperfectly screened at the Fermi surface. Therefore there is an added

direct Coulomb energy cost of filling $49_\downarrow$, in comparison to $50_\uparrow$. Consequently the next state to go below E_F is again a spin up state, which further increases the splitting. Finally $p = 51_\uparrow$, which is a strongly scarred state, fills. At this point the total dot polarization is maximum at 3 electrons.

Generally, strongly scarred states tend to produce a gap in the spectrum since they have a highly inhomogeneous spatial distribution. Empirically we find that the polarization tends to peak at the filling of the first spin state of a scar and drops to zero when it is the last polarized state to double fill. This occurs in Fig. 2 at, for example, $N = 102$. The $p = 51_\uparrow$ state undergoes anticrossings at $N \approx 100.5$ and $N \approx 101.4$, and rejoins its spin partner at $N = 102$, where the spin polarization goes to zero and all states become spin degenerate. The pattern of polarization and depolarization, modulated by strongly scarred states, repeats throughout the range of N considered in the calculation.

Clearly, the extent to which quantum dots will spin polarize depends upon the excess of diagonal Coulomb matrix elements over off diagonal elements (i.e. the cost to double occupy a state) as compared to the average bare level spacing Δ. Since Δ scales as L^{-2} and the Coulomb matrix elements scale as L^{-1}, one expects that for small dots spin polarization will be suppressed. This, however, does not take into account the regularity of the spectrum of small dots (due to, for example, approximately bi-parabolic confinement) which can produce level degeneracies. Also, while the typical Coulomb interaction goes as $1/L$, it is not clear that the excess of diagonal (same spatial state) over off-diagonal interaction also scales in the same way, particularly insofar as screening is likely to play an important role.

Note that it is by no means *a priori* obvious why the polarization should drop periodically to zero. One might expect, given a typical average difference between diagonal and off-diagonal Coulomb matrix elements and a typical level spacing, that some roughly constant, non-zero polarization would be favored (say one or two electrons) and the plot of P vs. N would show random fluctuations up and down from that average value. That polarization periodically collapses, a fact which above we have related to the unusually large diagonal matrix element of scarred states, still requires, we believe, a more elaborate explanation.

It is interesting to speculate on the possibilities for directly observing the fluctuating spin polarization and also for devices which employ the spin polarization. It is well known that a spin-valve effect exists for transport across an interface between magnetic materials which differ in their anisotropic pinnings [7]. However the direction of the magnetizations in this case are fixed by crystallographic properties, whereas in the quantum dot case the strength of pinning of P to a given direction, such as the perpendicular to the plane of the dot, is unknown. Nonetheless, if a polarized source of electrons can be generated, it is plausible that a giant magnetoresistance effect will occur in response to the reversal of a weak magnetic field, which will flip the dot total spin. We are currently investigating further these questions of pinning.

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