Density-functional theory of spin-polarized disordered quantum dots

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Using density-functional theory, we investigate fluctuations of the ground-state energy of spin-polarized, disordered quantum dots in the metallic regime. To compare to experiment, we evaluate the distribution of addition energies and find a convolution of the Wigner-Dyson distribution, expected for noninteracting electrons, with a narrower Gaussian distribution. The width of the Gaussian is accurately given by the fluctuations in the *screened* Coulomb interaction between a pair of electrons at the Fermi energy.

The interplay of disorder and electron-electron interactions in quantum dots has recently attracted much attention. Experiments using quantum dots—small islands fabricated in a two-dimensional electron gas[1]—measure the spacings between conductance peaks in the Coulomb blockade region. Since the peak spacings reflect differences between ground-state energies *for different numbers of electrons* one should not cavalierly apply random matrix theory[2] to evaluate the spectrum of peak-spacing fluctuations. Indeed, experiments find a more symmetric distribution than the Wigner-Dyson form. Experiments disagree, however, on the magnitude of the fluctuations.[3–6] Sivan *et al.*[3] observed fluctuations several times as large as the inferred mean level spacing $\langle \Delta_0 \rangle$, and concluded that the fluctuations are a fixed percentage 10–15% of the total charging energy e^2/C, where C is the dot capacitance. In contrast, Patel *et al.*[5] found fluctuations in GaAs dots comparable to the mean level spacing. Simmel *et al.*[6] performed the experiment in small Si dots and found fluctuations roughly of order $\langle \Delta_0 \rangle$.

Theoretical treatments also disagree regarding the magnitude of the peak-spacing fluctuations. Sivan *et al.*[3] found large fluctuations scaling as $(0.10-0.17)e^2/C$ for a small lattice model. Similar results were found by Koulakov *et al.* for the classical, strong interaction regime $r_s \gg 1$,[7] where electrons form a Wigner lattice.[8] Blanter *et al.*[9] used the random-phase approximation (RPA)[10] for weakly interacting dots and concluded that, for dimensionless conductance $g \gg 1$,[11] the contribution to fluctuations from interactions should be parametrically *smaller* than the mean level spacing $\langle \Delta_0 \rangle$. While the above results can be reconciled as applying to different regimes of r_s and g, recent work employing the self-consistent Hartree-Fock equations[12–14] found peak-spacing fluctuations several times as large as $\langle \Delta_0 \rangle$ even for $r_s \sim 1$ and $g \gg 1$, where RPA should still provide a good approximation.[10]

The purpose of the present paper is to clarify the origin, magnitude, and distribution of peak-spacing fluctuations in spin-polarized disordered quantum dots in the regime $g \gtrsim 1$ and $r_s \sim 1$. Density-functional theory (DFT) provides us with accurate ground-state energies including electron-electron interaction, confinement, and disorder for realistic quantum dots. We find that the distribution of peak spacings is the convolution of a Wigner-Dyson distribution, expected for noninteracting electrons, with a *narrower* Gaussian distribution due to interactions. The width of the Gaussian is accurately given by the fluctuations in the screened Coulomb interaction between a pair of electrons at the Fermi energy. The total peak spacing *fluctuations* are hence smaller than the mean level spacing $\langle \Delta_0 \rangle$. (This central result is expected to also apply to unpolarized quantum dots, even though the noninteracting spectrum in that case is not Wigner-Dyson.) Use of an *unscreened* interaction between electrons, either direct or exchange, is found to greatly overestimate the magnitude of the fluctuations. Furthermore, since interactions add a symmetric contribution to the distribution of peak-spacing fluctuations, the third moment of the total distribution is independent of interactions. Hence, we predict that experimental application of a magnetic field will reduce the third moment by a universal factor of 0.405, corresponding to a change from the Gaussian orthogonal ensemble to the Gaussian unitary ensemble.

The ground-state energies of spin-polarized, disordered quantum dots are obtained within density-functional theory with the exchange-correlation part of the electron-electron interactions treated in the local-density approximation. Specifically, we solve the following Kohn-Sham equations[15] numerically, and iterate until self-consistent solutions are obtained;[16]

$$\left[-\frac{\hbar^2}{2m^*}\nabla^2 + \frac{e^2}{\kappa}\int \frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}' + \frac{\delta E_{\mathrm{xc}}[\rho,\zeta]}{\delta\rho(\mathbf{r})} + V_{\mathrm{ext}}(\mathbf{r}) \right]\Psi_i(\mathbf{r})$$
$$= \epsilon_i \Psi_i(\mathbf{r}), \tag{1}$$

where the density is

$$\rho(\mathbf{r}) = \sum_i^N |\Psi_i(\mathbf{r})|^2. \tag{2}$$

Here $E_{\mathrm{xc}}[\rho,\zeta]$ is the exchange-correlation energy functional[17] with local spin polarization $\zeta(\mathbf{r})=1$. The summation in the density (2) is taken over the N lowest energy Kohn-Sham orbitals. In a previous work,[18] we have shown that the DFT method gives very accurate ground-state energies for *clean* parabolic GaAs quantum dots, in agreement

with exact calculations for up to five electrons.[19] Comparison with quantum Monte Carlo calculations[20] confirms that DFT is valid for interaction strengths up to $(e^2/\kappa l_0)/\hbar\omega_0=6$ ($r_s \simeq 8$) and up to $N=8$ electrons.

The external potential for our disordered dots is the sum of a confining parabola and multiple ''impurity'' potentials each with a Gaussian profile:

$$V_{\text{ext}}(\mathbf{r}) = \frac{1}{2}m^*\omega_0^2 r^2 + \frac{1}{2\pi\lambda^2}\sum_i^{N_{\text{imp}}} \gamma_i \exp\left(-\frac{|\mathbf{r}-\mathbf{r}_i|^2}{2\lambda^2}\right). \tag{3}$$

The impurity potentials are randomly distributed with density $n_{\text{imp}}=1.03\times10^{-3}$ nm^{-2} and strength γ_i uniformly distributed on $[-W/2,W/2]$ with $W=10\hbar^2/m^*$. The width of each impurity is taken as $\lambda=l_0/(2\sqrt{2})$, where $l_0 = \sqrt{\hbar/m^*\omega_0}\simeq19.5$ nm. Here we use the effective mass for GaAs, $m^*=0.067m$, and $\hbar\omega_0=3.0$ meV. The strength of the Coulomb interaction is controlled by changing the dielectric constant κ, where $\kappa=12.9$ for GaAs. The resulting dimensionless interaction strength is measured by $(e^2/\kappa l_0)/\hbar\omega_0$ or $r_s(=1/\sqrt{\pi\rho_0}a_B^*)$, where $a_B^*=\hbar^2\kappa/m^*e^2$ is the effective Bohr radius, and ρ_0 is the electron density at the center of the dot. From a scattering phase-shift analysis we find the mean-free-path of electrons $l=v_F\tau\simeq170$ nm to be slightly larger than the dot diameter $L=120-160$ nm, where the dot diameter increases with r_s. Therefore the dots are marginally in the ballistic regime and have a dimensionless conductance $g=2-4$.[11]

At low temperatures, electron hopping into a dot containing $N-1$ electrons is suppressed except when the ground-state free energy $E(N-1)-(N-1)\mu$ is equal to the ground-state free energy for N electrons $E(N)-N\mu$. This degeneracy condition determines the position of the Nth conductance peak as a function of the electron chemical potential $\mu_N=E(N)-E(N-1)$, or equivalently, as a function of an applied gate voltage.[21] The increase in μ needed to put an extra electron in the dot, which we will refer to as the addition energy Δ, is given by $\Delta=E(N+1)-2E(N)+E(N-1)$. From our solution of the Kohn-Sham equations, the ground-state energy of a dot with N electrons is obtained from

$$E(N) = \sum_i^N \epsilon_i - \frac{e^2}{2\kappa}\int\frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}\,d\mathbf{r}'$$

$$- \int \rho(\mathbf{r})\frac{\delta E_{\text{xc}}[\rho,\zeta]}{\delta\rho(\mathbf{r})}d\mathbf{r} + E_{\text{xc}}. \tag{4}$$

We consider fluctuations of the addition energy for $N=10$ electrons. Thus for each realization of disorder we calculate $\Delta\equiv E(11)-2E(10)+E(9)$. The disorder average is taken over more than 1,000 different impurity configurations. As a check of accuracy, we have confirmed that the ground-state energies obtained from DFT for disordered quantum dots with $N=2$ and 3 are in good agreement with exact diagonalization results for $0\leq r_s\leq5$.
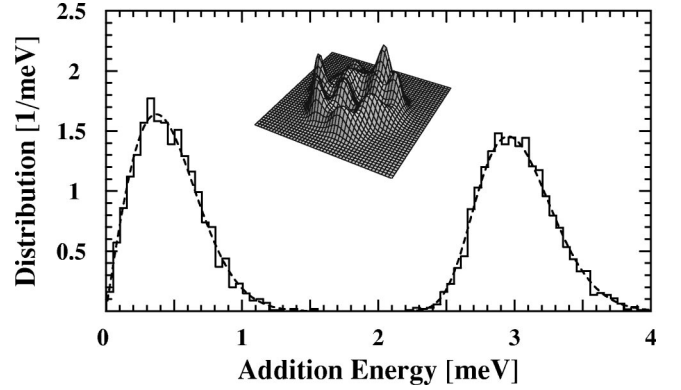

FIG. 1. Distribution of addition energies Δ to add the 11th electron for interacting dots with $(e^2/\kappa l_0)/\hbar\omega_0=2.39$ (right) and Δ_0 for noninteracting dots of the same size (left). The energy binwidth is 0.05 [meV]. The dashed lines show the distribution function obtained from Eq. (10). Inset: The charge density profile $\rho(\mathbf{r})$ for $N=10$ electrons with one configuration of impurities.

Figure 1 shows the distribution of addition energies Δ for interacting dots with $(e^2/\kappa l_0)/\hbar\omega_0=2.39$ (right), and the distribution of Δ_0 for noninteracting dots of the same size (left). In the inset, we show the charge density $\rho(\mathbf{r})$ for one realization of disorder. While interactions considerably enhance the average addition energy $\langle\Delta\rangle\simeq6.50\langle\Delta_0\rangle$, the fluctuation in the interacting case $\delta\Delta\equiv\sqrt{\langle\Delta^2\rangle-\langle\Delta\rangle^2}$ is only $\sim13\%$ larger than the noninteracting fluctuation $\delta\Delta_0 \equiv\sqrt{\langle\Delta_0^2\rangle-\langle\Delta_0\rangle^2}$. The distribution of level spacings Δ_0 in the noninteracting dots has the Wigner-Dyson form, while that in the interacting dots is somewhat more symmetrical. The symmetry continues to increase with increasing interaction strength.

Figure 2 shows the average addition energy $\langle\Delta\rangle$, its rms fluctuations $\delta\Delta$, and its third moment for disordered dots as a function of the Coulomb interaction strength $(e^2/\kappa l_0)/\hbar\omega_0$. For comparison, we have also plotted results for disordered, noninteracting dots of the same size, which we obtain as follows. First we find the effective potential for the clean dot, without impurities. Then we solve for the single-particle level energies ϵ_i^0 for this effective potential plus the random impurity potentials in Eq. (3). The addition energy is simply given by $\Delta_0=\epsilon_{N+1}^0-\epsilon_N^0$ with $N=10$. All dot sizes satisfy the relation $\delta\Delta_0=\sqrt{4/\pi-1}\langle\Delta_0\rangle\simeq0.52\langle\Delta_0\rangle$ predicted by random matrix theory for the Gaussian orthogonal ensemble.[2] In Fig. 2(a), the average addition energy in the noninteracting case is seen to decrease with increasing Coulomb interaction. This is because the increasing Coulomb repulsion among electrons causes the dot to grow and hence the level spacing to shrink. The average addition energy in the interacting case increases considerably with Coulomb interaction strength, as expected from the classical electrostatics relation $\langle\Delta\rangle\simeq e^2/C$, where C is the capacitance of the dot. However, Fig. 2(b) shows that the interactions only slightly increase the addition-energy *fluctuations*. For GaAs, $r_s\simeq2$, the enhancement is only about 10%, in rough agreement with the experiment of Patel *et al.*[5]

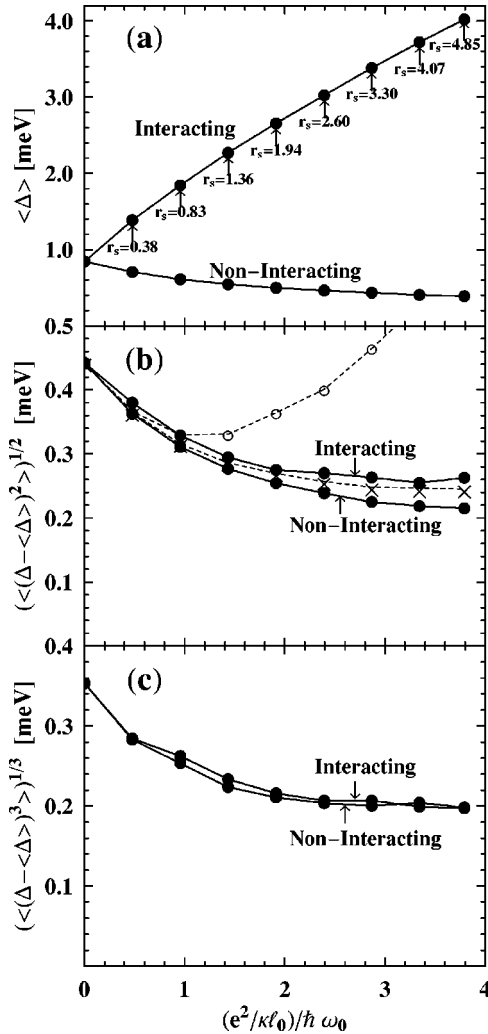To understand the magnitude of addition-energy fluctua-

FIG. 2. (a) Average addition energy $\langle \Delta \rangle$, (b) fluctuation $\delta \Delta$, and (c) cube-rooted third moment $\sqrt[3]{\langle (\Delta - \langle \Delta \rangle)^3 \rangle}$, as a function of electron-electron interaction strength $(e^2/\kappa l_0)/\hbar \omega_0$. The measure of interaction strength $r_s(=1/\sqrt{\pi \rho_0}\, a_B^*)$ indicated by arrows in (a) can be applied to the data in all panels. For each data point, the disorder average is taken over more than 1000 different impurity configurations. At each r_s, the noninteracting data are taken for dots of the same size as the interacting dots, and the relation $\delta \Delta_0 \simeq 0.52 \langle \Delta_0 \rangle$ expected for noninteracting level-spacing statistics is always satisfied. Also plotted in (b) are the fluctuations due to noninteracting level spacings plus the *screened* Coulomb interaction between two electrons at the Fermi surface (crosses), or the *unscreened* exchange interaction between the two electrons (open circles).

tions, we use the phenomenological framework presented by Blanter *et al.*[9] for the regime $r_s \ll 1$, where RPA is valid, and show that it applies to the DFT results at least up to $r_s \simeq 5$. Consider first a dot containing $N-1$ electrons in the ground state. Addition of the Nth electron to form the N electron ground state requires an electron chemical potential μ_N. To form instead the first excited state of N electrons requires the higher chemical potential $\mu_N + \Delta \epsilon$. For an ensemble of disordered metallic dots, $\Delta \epsilon$ will have Wigner-Dyson statistics,

with $\langle \Delta \epsilon \rangle$ equal to the mean noninteracting level spacing $\langle \Delta_0 \rangle$, since the lowest excitation of a Fermi liquid is a single electron promoted across the Fermi surface.

The addition energy Δ is the *increase* in chemical potential from μ_N required to add one more electron to the dot and thus form the $N+1$ electron ground state. This $(N+1)$st electron must have an extra energy $\Delta \epsilon$ to occupy the lowest empty level *plus* an extra energy $U_{N,N+1}$ due to its Coulomb interaction with the Nth electron. The total addition energy Δ will be approximately given by the sum of these two contributions,

$$\Delta \simeq \Delta \epsilon + U_{N,N+1}. \tag{5}$$

The distribution of $\Delta \epsilon$ is given by the Wigner-Dyson distribution of level spacings for a noninteracting dot of the same size. The average interaction energy $\langle U_{N,N+1} \rangle$ is the capacitive charging energy e^2/C. We estimate the fluctuations in $U_{N,N+1}$ by calculating the screened Coulomb interaction between two electrons at the Fermi surface.[9] Specifically, we treat the screening effect in the Thomas-Fermi approximation as

$$U_{N,N+1}^{TF} = e \int \varphi_N(\mathbf{r}) \rho_{N+1}^0(\mathbf{r}) d\mathbf{r}. \tag{6}$$

The screened potential due to the Nth electron in Fourier representation is[22]

$$\varphi_N(\mathbf{q}) = \frac{2 \pi e}{\kappa} \frac{\rho_N^0(\mathbf{q})}{|\mathbf{q}| + q_0}, \tag{7}$$

where $\rho_N^0(\mathbf{r}) = |\phi_N^0(\mathbf{r})|^2$ is the density of the Nth single-particle wave function $\phi_N^0(\mathbf{r})$ of a noninteracting disordered dot. The Thomas-Fermi wave vector is $q_0 = (2 \pi e^2/\kappa)(dn/d\mu) = 1/a_B^*$. It is found that the fluctuation $\delta U^{TF} = \sqrt{\langle (U_{N,N+1}^{TF})^2 \rangle - \langle U_{N,N+1}^{TF} \rangle^2}$ is always considerably smaller than the noninteracting level-spacing fluctuation $\delta \Delta_0$ up to at least $r_s \simeq 5$. The total fluctuation estimated as $\delta \Delta^{TF} = \sqrt{(\delta \Delta_0)^2 + (\delta U^{TF})^2}$ is shown in Fig. 2(b) by crosses. We see that the fluctuations in the Thomas-Fermi screening model agree well with the DFT results with no free parameters. This supports the picture[9] that the addition-energy fluctuation arises from two quasiparticles above a filled Fermi sea interacting via a *screened* Coulomb potential.

Within this picture, the increase of the fluctuation of $U_{N,N+1}$ with increasing interaction strength leads naturally to greater symmetry of the distribution of addition energies. Numerically, we find that that the distribution of $U_{N,N+1}^{TF}$ has a symmetric Gaussian form. Hence, in agreement with Eq. (5), we observe that the addition-energy distribution function $P(\Delta)$ is always extremely well described by the convolution of a Wigner-Dyson distribution for level spacings $\Delta \epsilon$,

$$P_{\mathrm{WD}}(\Delta \epsilon) = \frac{\pi}{2} \frac{\Delta \epsilon}{\langle \Delta_0 \rangle^2} e^{-\pi \Delta \epsilon^2/(4 \langle \Delta_0 \rangle^2)} \tag{8}$$

with a Gaussian distribution for interaction energies $U_{N,N+1}$,

$$P_{\text{Gauss}}(U_{N,N+1}) = \frac{1}{\sqrt{2\pi}\delta U} e^{-(U_{N,N+1} - \langle\Delta\rangle + \langle\Delta_0\rangle)^2/[2(\delta U)^2]}.$$

(9)

The result for the distribution of addition energies is

$$P(\Delta) = \int \int d\Delta\epsilon \, dU_{N,N+1} \, P_{\text{WD}}(\Delta\epsilon) P_{\text{Gauss}}(U_{N,N+1})$$

$$\times \delta(\Delta\epsilon + U_{N,N+1} - \Delta)$$

$$= \frac{1}{2} \sqrt{\frac{\pi}{2}} \frac{\delta U}{\alpha\langle\Delta_0\rangle^2} e^{-\pi\tilde{\Delta}^2/(4\alpha\langle\Delta_0\rangle^2)}$$

$$\times \left\{ e^{-\tilde{\Delta}^2/[2\alpha(\delta U)^2]} + \sqrt{\frac{\pi}{2\alpha}} \frac{\tilde{\Delta}}{\delta U} \right.$$

$$\times \left. \left[1 + \text{erf}\left(\frac{\tilde{\Delta}}{2\alpha(\delta U)^2} \right) \right] \right\}.$$

(10)

Here $\alpha = \pi\delta U^2/(2\langle\Delta_0\rangle^2) + 1$ and $\tilde{\Delta} = \Delta - \langle\Delta\rangle + \langle\Delta_0\rangle$, where $\langle\Delta\rangle$ is the center of the distribution and δU is a fitting parameter giving the width of the fluctuations of $U_{N,N+1}$. In the noninteracting case, $\delta U = 0$ so that $P(\Delta) = P_{\text{WD}}(\Delta)$ as expected. In the other limit, $P(\Delta)$ becomes nearly symmetric for sufficiently large δU. In Fig. 1, we show $P(\Delta)$ given by Eq. (10) as a dashed line. It is seen that the DFT distribution is described very well by Eq. (10) with the best fit value of $\delta U = 0.13$ meV very close to the value $\delta U_{N,N+1}^{TF} = 0.10$ meV estimated from the Thomas-Fermi screened Coulomb interaction between two electrons at the Fermi surface.

To test whether the distribution of addition energies is well described by the sum of noninteracting level spacings and a symmetric distribution due to interactions, we propose to compare the third moment of the distribution $P(\Delta)$ with and without a magnetic field $B_\perp$ normal to the plane of the dot. Since the interaction part, coming from the screened Coulomb interaction in our picture, is symmetric, it does not contribute to the third moment of $P(\Delta)$. Therefore, the ratio $\langle(\Delta - \langle\Delta\rangle)^3\rangle_{B_\perp \neq 0}/\langle(\Delta - \langle\Delta\rangle)^3\rangle_{B_\perp = 0}$ should take the value $(2 - 5\pi/8)/(2 - 6/\pi) \simeq 0.405$, which applies to level spacings taken from a Gaussian orthogonal ensemble ($B_\perp = 0$) and a Gaussian unitary ensemble ($B_\perp \neq 0$).[2] Since our results apply only to the case of spin-polarized electrons, it is necessary to apply a large magnetic field in the plane of the dots, or to spin polarize the nuclei.[23] The result can also be tested numerically, e.g., by exact diagonalization studies as in Ref. 3.

Existing diagonalization studies for spin polarized electrons on small lattices find addition-energy fluctuations $\delta\Delta \simeq 0.15e^2/C$.[3] For comparison, the Coulomb contribution to the fluctuations found by DFT are much smaller, $\delta\Delta \simeq 0.03e^2/C$ at $r_s \simeq 2$. This difference may be attributed to differences in the strength of disorder; while the dimensionless conductance in our dots is $g = 2-4$, we estimate g

$= 0.1 - 0.3$ in Ref. 3. Various theoretical estimates give fluctuations $\delta\Delta \propto \langle\Delta_0\rangle/g$ (Ref. 24) or $\delta\Delta \propto \langle\Delta_0\rangle/\sqrt{g}$.[9] In either case, the discrepancy between exact diagonalization and DFT can be attributed to the order of magnitude difference in the dimensionless conductance g in the samples studied. The experiments on GaAs (Refs. 3–5) have $r_s \sim 1$ and $g > 1$, and thus fall more closely in the range of interaction strengths and dimensionless conductance treated in this paper.

Recently, several calculations[12–14] based on the self-consistent Hartree-Fock (SCHF) equations have found large fluctuations, up to $\delta\Delta \simeq 0.2e^2/C$, in the same range of r_s we consider. In one case,[13] the dimensionless conductance is estimated to be $g \gg 1$, a regime where DFT predicts fluctuations an order of magnitude smaller. As pointed out by Walker *et al.*,[12] the exchange interaction in the SCHF equations is unscreened. To test whether the lack of exchange screening in the SCHF approach could be responsible for the discrepancy with DFT, we have calculated the unscreened exchange interaction between two electrons near the Fermi surface in our dots

$$U_{N,N+1}^{\text{exch}} = \frac{e^2}{\kappa} \int \int \frac{\phi_N^{0*}(\mathbf{r})\phi_N^0(\mathbf{r}')\phi_{N+1}^{0*}(\mathbf{r}')\phi_{N+1}^0(\mathbf{r})}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} \, d\mathbf{r}'.$$

(11)

In Fig. 2(b), we have plotted as open circles the fluctuations taken by summing the unscreened exchange interaction (11) with the noninteracting level spacing.[25] It is clear that for $r_s > 1$, the unscreened exchange interaction noticeably overestimates the addition-energy fluctuations. In contrast, density-functional theory correctly accounts for screening within the electron gas, including exchange interactions.[26] These results suggest that the unscreened exchange interaction in the SCHF approach may generally lead to an overestimate of the addition-energy fluctuations.

In this paper, we have neglected external screening by gates or electrodes. This simplification should be valid as long as the distance to external conductors is larger than the diameter of the dot. In the opposite limit, it is essential to consider external screening, but this may be done by a simple modification of the $1/r$ potential between electrons.

In conclusion, we have studied the electronic states of spin-polarized, disordered quantum dots using density-functional theory and investigated the fluctuation of the ground-state energies. We have found that interactions change the mean addition energy but not the second moment much. Electron-electron interactions increase the fluctuation of addition energies by no more than 25%, up to $r_s \simeq 5$, even though the average addition energy is increased by a factor of 10. The addition energy is well approximated as the sum of the noninteracting level spacing and the screened Coulomb interaction between two electrons at the Fermi surface. Hence the distribution of addition energies is the convolution of a Wigner-Dyson distribution of level spacings with a Gaussian distribution of interaction energies. Since the latter is symmetric, it does not contribute to the third moment of the addition-energy distribution. The third moment is there-

fore predicted to decrease by a universal factor of 0.405 on application of a magnetic field that transforms the dot from the Gaussian orthogonal to the Gaussian unitary ensemble. For quantum dots having larger numbers of electrons, whether spin polarized or not, we anticipate that the decrease of the screened Coulomb interaction-energy fluctuations oc-

curs as fast as the decrease of the level spacing and thus the present results are also applicable.

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