A semiconducting nanotube quantum dot: effect of the form of the confinement potential.

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The states of a few (2-6) interacting electrons in a semiconducting carbon nanotube quantum dot are investigated for two extremes of the quantum dot confinement potential: harmonic and hard wall. The interacting electron states are calculated by exact diagonalisation of a single band, effective mass Hamiltonian, then the quantum dot addition energies and few particle densities are compared for the two types of confinement. By changing the harmonic confinement strength the system can be driven between the strongly and weakly correlated regimes, whereas for hard wall confinement the electrons are almost always strongly correlated. With hard wall or weak harmonic confinement the electrons localise and form Wigner crystal-like states.

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1 Introduction. Semiconducting nanotube (NT) quantum dots are technologically important. Recent advances include the first measurement of the addition energy of a semiconducting NT quantum dot [1] and the fabrication of a room temperature single electron transistor (SET) based on a semiconducting NT quantum dot [2]. However, although these novel 1D artificial atoms are important, their physics is not well understood. One of the major problems is that the nature of the quantum dot confinement potential is unknown and we address this problem here. We describe an effective mass approach for calculating the states of a few interacting electrons in a NT quantum dot and investigate the effect of the confinement potential on the dot addition energy.

We are interested in quantum dots similar to the dot in the NT SET device studied by Jarillo-Herrero *et al* [1]. In this device, a semiconducting tube is placed between two metallic contacts and separated from a back-gate by a thin insulating layer. The SET operation is controlled by a quantum dot which is formed near the tube centre where electrons may be confined electrostatically. The difficulty is that the exact form of this dot confinement is not understood. The total confinement will probably contain contributions from both the back-gate potential and the Schottky barriers at the interface between the nanotube and the contacts [1]. While the contribution from the back-gate will be approximately harmonic close to the tube centre, the Schottky barrier contribution is more difficult to estimate. Although the exact form of the confinement potential is unknown it is clear that it will lie somewhere between a smoothly varying harmonic potential and an abruptly varying hard wall potential. We compare the few particle charge density and addition energy for these two extremes of the NT quantum dot confinement and show that the confinement in the experimental system is more likely to be harmonic.

2 Calculation. We calculate the states of a few interacting electrons in a semiconducting NT quantum dot. We use a dot model based on the NT quantum dot studied experimentally by Jarillo-Herrero *et al* [1]. In this type of quantum dot electrons are confined at the centre of a nanotube of roughly a few hundred

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nanometres in length, L . Typically, the dot length scales are a few tens of nanometres and the dot energy scales are of the order of a few, to a few tens, of meV. We consider nanotubes with a band gap, E_g , much larger than this energy scale and a unit cell length, T , much smaller than this length scale. We can then ignore the excitations of the radial and angular tube sub-bands and the nanotube dot is effectively one dimensional. We describe the interacting electrons in this dot with the single band effective mass Hamiltonian,

$$H = \sum_i^N \frac{-\hbar^2}{2m^*} \frac{d^2}{dz_i^2} + U(z_i) + \sum_{i \neq j} V(|z_i - z_j|), \quad (1)$$

where N is the number of electrons, $U(z_i)$ is the single particle confinement potential and $V(|z_i - z_j|)$ is the effective Coulomb interaction. We calculate the effective mass, m^* , from a 4 orbital, 3rd nearest neighbour, tight binding calculation with the parameterisation of Porezag *et al* [3].

To calculate the few electron states we diagonalise the Hamiltonian (Eq. (1)) exactly in a basis of determinantal functions. We are interested in two types of single particle confinement potential, either hard wall ($U(z) = -V_o, |z| < \frac{L}{2}, 0$ otherwise) or harmonic ($U(z) = \frac{1}{2}m^*\omega^2 z^2$) confinement. In each case we expand the interacting few electron states in the determinantal basis that diagonalises the single electron part of the Hamiltonian. This ensures that the calculation is efficient and allows us to converge the ground state energy to within 0.1%. Typically we need to include approximately 10^5 Slater determinants in the expansion to obtain this level of convergence for 6 electrons.

We derive the effective Coulomb interaction between two electrons in the dot from the standard form of the interaction between charged particles on the surface of a cylinder. If we assume that the interaction cannot scatter electrons between the angular sub-bands of the tube then,

$$V(|z|) = \frac{e^2}{4\pi\epsilon_o\epsilon_r\sqrt{|z|^2 + 4R^2}} \frac{2}{\pi} \mathbf{K} \left(\frac{2R}{\sqrt{|z|^2 + 4R^2}} \right), \quad (2)$$

where $z = z_i - z_j$, R is the nanotube radius, ϵ_r is the relative permittivity of the nanotube, and $\mathbf{K}$ is an elliptic integral of the first kind. Close to $z = 0$ the interaction diverges like $\log(z)$ and, as expected, the interaction depends inversely on R : for larger tubes the effective Coulomb interaction is smaller.

The semiconducting nanotube in the Jarillo-Herrero device is a 35,0 zigzag tube. In the zigzag class of tubes the conduction band is doubly degenerate and we should include the effect of both bands in equation (1). However, in any real nanotube SET device, this degeneracy will be broken by, for example, the electric field across the nanotube, or a lack of rotational symmetry at the contacts. We have investigated the effect of the band splitting with a 2 band calculation [4] and we find that the ground state energies, and hence the addition energies are relatively insensitive to the splitting. At large splittings ($\approx \hbar\omega$) the results of the single band and 2 band calculations are identical. We know that the bands in the experimentally investigated nanotube must be significantly split because the measured addition energy oscillates with a period of 2 electrons. There is no evidence for any 4 electron periodicity which would signify the presence of a second degenerate, or nearly degenerate, band. The single band Hamiltonian (Eq. (1)) is therefore a good approximation to the Hamiltonian of the NT dot device investigated in the experiment.

3 Results. We have calculated the few electron charge densities and the addition energies for quantum dots with both harmonic and hard wall confinement in a 35,0 zigzag nanotube of length $L = 270$ nm ($R = 1.37$ nm, $m^* = 0.0345$, $T = 0.426$ nm, and $E_g = 262.4$ meV). The NT dot addition energy is given by, $E_A(N) = E_{N+1} - 2E_N + E_{N-1}$, where E_N is the ground state energy of the N electron dot. The variation in E_A with N essentially depends on the ratio between the single particle confinement energy and the Coulomb interaction strength. In the weakly correlated regime the Coulomb interaction strength is small and the addition energy oscillates with N while, in the strongly correlated regime, the Coulomb interaction dominates and E_A varies smoothly with N .

Fig. 1 Addition energy against electron number. Diamonds: experimental results reproduced from Jarillo-Herrero *et al* [1]. Circles: calculated results with harmonic confinement parameters derived from the CI model ($\hbar\omega = 5.5$ meV, and $\epsilon_r = 1$). Triangles: calculated results with a hard wall confinement potential comparable to the harmonic confinement above, $V_o = \frac{1}{2}m^*\omega^2\frac{L^2}{2} = 124.7$ meV, $L = 270$ nm. $E_A(5)$ is not plotted for $\hbar\omega = 5.5$ meV, $\epsilon_r = 1$. With these parameters our best value for E_6 is only converged to $\approx 5\%$ with 2.9×10^5 Slater determinants in the expansion.

3.1 Constant interaction model. In the weakly correlated regime, quantum dots are often analysed qualitatively with the constant interaction (CI) model [1]. In this model the Coulomb interaction energy is constant, or slowly varying with N and, crucially, the single particle levels are assumed to fill up according to Hunds rule. The CI energy is then equal to $E_A(1)$, and the magnitude of the oscillations is related to the single particle level spacing. For example, in a quantum dot with harmonic confinement, $E_A(2) - E_A(1) = \hbar\omega$. In the Jarillo-Herrero device, the CI model constant interaction is large (≈ 45 meV) compared to $\hbar\omega \approx 5$ meV. From Fig. 1 it is clear that the CI model is unsatisfactory: the results of our full calculation with parameters derived from the CI model do not reproduce the experimentally observed features. This is because the interaction is so large. The interaction significantly mixes the single particle states and hence the assumption of simple, Hunds rule, shell filling breaks down.

3.2 Comparison between harmonic and hard wall confinement. To obtain addition energies with oscillations similar to those observed [1] we must increase the ratio of the single particle confinement energy to the Coulomb interaction strength from the value obtained with the CI model (Fig. 1). The Coulomb interaction is scaled by the tube dielectric constant, ϵ_r . The value of this constant is unknown but, typically, $1 < \epsilon_r < 7$ is assumed [5]. Here, we choose $\epsilon_r = 6$. Then, to obtain oscillations in $E_A(N)$ of approximately 5 meV we increase $\hbar\omega$ to 34.5 meV (Fig. 2). In this case the calculated $E_A(N)$ reproduces some of the features of the measured addition energy but decreases slightly too quickly with N . However, in the real system, the harmonic confinement is not constant but will increase with N . This is because, to put more electrons inside the dot, the back-gate potential must be raised and this will increase the harmonic confinement. If the insulating layer between tube and back-gate is relatively small (< 50 nm) the variation in harmonic confinement is significant and the rate at which E_A decreases with N will be much reduced.

From Fig. 2 it is clear that the electrons in both the harmonically confined dot ($\hbar\omega = 5.5$ meV) and the hard wall dot are too strongly correlated. To drive the hard wall system into the weakly correlated regime and hence obtain oscillations in E_A we need to reduce the dot width to less than 50 nm. With this type of confinement the dot width essentially controls the single particle level spacing and hence $E_A(N)$. The single particle level spacing is relatively insensitive to V_o . Instead V_o determines how many electrons will fit inside the dot. When $L = 50$ nm we need $V_o > 1$ eV to fit more than 5 electrons inside the dot.

In the strongly correlated limit the Coulomb interaction dominates, the electrons are forced as far apart as possible and tend to localise (Fig. 3). This electron localisation is, unsurprisingly, much more likely to occur with hard wall confinement when $U(z)$ is flat inside the dot. When the Coulomb interaction dominates, the electrons form Wigner crystal-like states and we can show that the 2 electron ground state in a dot with weak harmonic confinement is well described by a molecular approximation [4]. For example, with $\hbar\omega = 5.5$ meV and $\epsilon_r = 6$ the molecular approximation is accurate to 2% for E_2 .

Fig. 2 Addition energy against electron number. Boxes: harmonic confinement, $\hbar\omega = 34.5$ meV. Circles: harmonic confinement, $\hbar\omega = 5.5$ meV. Triangles: hard wall confinement, $V_o = 124.8$ meV, $L = 270$ nm.

Fig. 3 Left: 2 electron density. Right: 6 electron density. Solid line: harmonic confinement, $\hbar\omega = 5.5$ meV. Short dashed: harmonic confinement, $\hbar\omega = 34.5$ meV. Long dashed: hard wall $U(z)$, $V_o = 124.7$ meV, $L = 270$ nm.

In conclusion, we have described an effective mass approach to calculating the few (2-6) interacting electron states in a semiconducting nanotube quantum dot. We have investigated the form of the confinement potential in the NT dot and shown that, while some of the experimentally observed features in the addition energy may be reproduced with harmonic confinement, they cannot be explained if the quantum dot confinement is an abrupt potential step.

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