

Calculation of the few-electron states in semiconductor carbon nanotube quantum dots by exact diagonalisation

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Abstract. The 3 and 4-electron states of a gated semiconducting carbon nanotube quantum dot are calculated by exact diagonalisation of a modified effective mass Hamiltonian. A typical nanotube quantum dot is examined and the few-electron states are Wigner molecule-like. The exact diagonalisation method and the rate of convergence of the calculation are discussed.

1. Introduction

In a gated semiconductor nanotube (SNT) quantum dot, electrons or holes are confined electrostatically. The occupancy of this type of dot can be precisely controlled and, importantly, a semiconducting nanotube dot can be completely emptied of electrons. This means that SNT quantum dots are excellent systems in which to study the physics of a few interacting particles.

Like traditional semiconductor quantum dots, SNT dots behave like artificial atoms but there are some differences that make semiconducting carbon nanotube dots unique. First, the SNT bandstructure is unusual, primarily because nanotubes are rolled graphene sheets and the nanotube bandstructure is derived from that of graphene. Second, the geometry of SNT dots is also unusual: the nanotube length is very much greater than its diameter and so SNT dots are quasi-1 dimensional objects. In Ref. [1] we demonstrated that the electronic correlation was large in a large proportion of all physically accessible SNT quantum dots. Taken together with the tube geometry, this means that SNT dots are ideal systems in which to observe quasi-1D Wigner (or all-electron) molecules.

In our approach, we describe the few particle states in a SNT quantum dot with a 1 dimensional, 2 band, effective mass Hamiltonian [1]. However, because the electron correlation is strong and the few-particle states are Wigner molecule-like, care must be taken in the calculation of the electron states: exact diagonalisation, or configuration interaction (CI), techniques must be used to include the correlation effects correctly. In this paper we give an overview of our effective mass description of SNT dots and then discuss the exact diagonalisation scheme in detail. We examine the convergence of the few-electron states and discuss a way of truncating the full CI basis which improves the convergence of the few-electron energy.

2. Effective mass description of a nanotube dot

Our dot model is based on the type of semiconducting nanotube quantum dot that is typically studied experimentally [2]. In this type of device, a long ($\sim \mu\text{m}$) nanotube is contacted between source and drain electrodes and separated from a back-gate by a thin insulating layer. The quantum dot potential is generated electrostatically by applying a voltage to the back-gate and, in our model, this confinement potential varies parabolically along the length of the tube.

The confined states of a typical SNT dot are broad and may extend over a few hundred nanometres (of the order of a few hundred thousand atoms). However, although the states are broad, their extent is much less than the typical source-drain separation and so we assume our dot is completely isolated. The energy scale of a SNT dot is typically of the order of a few to ten meV, much less than the typical SNT band gap (~ 1 eV). So, given that the dot length scale is also much larger than the SNT unit cell length, the few-particle states in the dot can be described with an effective mass theory. However, there are new features which must be added to the theory to take account of the unusual nanotube bandstructure.

Nanotubes are rolled graphene sheets, and in each SNT there are two equivalent conduction bands with minima equally spaced either side of $k = 0$. The two conduction bands arise from the equivalence of the K and K' points in the graphene Brillouin zone and there is an additional complication because the dispersion near to each of these points in the graphene zone is linear. In Ref. [1] we demonstrated how to deal with both these effects by expanding the SNT dot states in terms of the two-component Bloch wavefunctions of graphene. The net result is a 1D, 2-band effective mass Hamiltonian with a modified Coulomb interaction, U , that contains all of the effects of the nanotube bandstructure and the details of the nanotube geometry,

$$H = \sum_{i=1}^N \left[\frac{-\hbar^2}{2m^*} \frac{d^2}{dz_i^2} + \frac{m^* \omega^2}{2} z_i^2 \right] + \frac{\rho/R}{2\epsilon_r} \sum_{i \neq j} U(z_i - z_j, R). \quad (1)$$

Here N is the number of electrons, $\hbar\omega$ is the parabolic confinement energy, R is the SNT radius, ϵ_r is the dielectric constant, m^* is the effective mass, $\rho = e^2/(4\pi\epsilon_o)$ and

$$U(z, R) = \frac{1}{4}(2u(z, R) + u(z - 3R, R) + u(z + 3R, R)). \quad (2)$$

$u(z, R)$ is derived from the standard form of the interaction between charged particles on the surface of a cylinder,

$$u(z, R) = \frac{2R}{\pi\sqrt{z^2 + 4R^2}} \mathbf{K} \left(\frac{2R}{\sqrt{z^2 + 4R^2}} \right), \quad (3)$$

and \mathbf{K} is an elliptic integral of the first kind which diverges like $\log(z/R)$ as $z \rightarrow 0$.

Eq. (2) gives the *intra*band effective interaction. We ignore the *inter*band interaction because its effect is small [3, 4]. In this case the band index acts like a pseudospin and our Hamiltonian is block diagonal in both the total electron spin and the pseudospin.

3. Exact diagonalisation

To calculate the N particle energies and wavefunctions we must solve the Schrödinger equation, $H\Psi(z_1 \dots z_N) = E\Psi(z_1 \dots z_N)$. We expand the N particle states, $\Psi(z_1 \dots z_N)$, as a sum of Slater determinants, $\Phi_i(z_1 \dots z_N)$, constructed from the single particle states of the system,

$$\Psi(z_1 \dots z_N) = \sum_i a_i \Phi_i(z_1 \dots z_N), \quad (4)$$

where each $\Phi_i(z_1 \dots z_n)$ is a normalised eigenfunction of the total spin, S and its z component S_z . In principle, this expansion is exact. However, in reality, the basis set can never be truly complete

and must be truncated. In our calculation we use a slight variation of the full configuration interaction scheme in which *all* the possible determinants that can be generated from a given finite single particle basis set are included in the expansion. The size of our finite basis is then determined by a single parameter, a maximum quantum number n , and we can systematically converge our calculation of the few particle states simply by increasing n .

To solve the Schrödinger equation numerically we first transform it into a standard matrix eigenvalue equation by left multiplying with one of the determinants from the expansion in Eq. (4) and integrating over all the $z_1 \dots z_N$ spatial coordinates. We have

$$\sum_i (\langle \Phi_j | H | \Phi_i \rangle - E \delta_{ij}) a_i = 0,$$

or, in matrix notation,

$$(\mathbf{H} - E\mathbf{I}) \mathbf{a} = 0,$$

where \mathbf{I} is the unit matrix. To find the N particle energies and the expansion coefficients, a_i , we diagonalise this Hamiltonian matrix numerically. The calculation time scales as the size of the matrix cubed and, to reduce this time, we block the Hamiltonian matrix according to the total pseudospin and the symmetry of each N particle state along the nanotube.

To reduce the size of the calculation we also slightly modify the full CI scheme. Our basis functions span a multi-dimensional Hilbert space, but we find that states from the 'corners' of this space do not contribute significantly to the few-electron wavefunctions. So, if

$$\Phi_i(z_1 \dots z_N) = \frac{1}{\sqrt{N!}} \begin{vmatrix} \phi_{m_{i1}}(z_1) & \phi_{m_{i2}}(z_1) & \dots & \phi_{m_{iN}}(z_1) \\ \phi_{m_{i1}}(z_2) & \phi_{m_{i2}}(z_2) & \dots & \phi_{m_{iN}}(z_2) \\ \vdots & \vdots & \ddots & \vdots \\ \phi_{m_{i1}}(z_N) & \phi_{m_{i2}}(z_N) & \dots & \phi_{m_{iN}}(z_N) \end{vmatrix}, \quad (5)$$

we find a_i to be negligible if all the m_{ij} quantum numbers are large. We therefore neglect determinants from these 'corners' and use a 'spherical cut off' to truncate the basis when $\left(\sum_{j=1}^N m_{ij}^2\right)^{1/2} > n$, as opposed to the usual 'cuboid cut-off' from the full CI scheme in which the basis is truncated when $m_{ij} > n$. The total number of determinants in our expansion at a given maximum quantum number, $M(n)$, depends on the truncation scheme, and we compare the convergence of the ground state energy in each of these two schemes in section 4.

4. Results and discussion

In Ref. [1] we found that the correlation was large in the majority of NT dots, and that all 2-electron states were Wigner molecule-like when $\frac{m^*}{\hbar^2} \left(\frac{\rho/\epsilon_r}{\hbar\omega}\right)^2 \geq 2.8$. In this work, we therefore focus on a typical strongly correlated SNT dot ($\hbar\omega = 5$ meV, $\epsilon_r = 2.0$) and we examine the 3 and 4 electron states. We choose a 35,0 nanotube with $R = 1.37$ nm and $m^* = 0.034$ as this is the type of tube studied experimentally in Ref. [2]. However, within the effective mass approximation, the details of the chirality are unimportant and our results will be similar for any SNT with similar R and m^* . In the 35,0 NT dot the N -electron ground state energies, E_N , (converged to within 0.1 meV) are $E_3 = 76.4$ meV and $E_4 = 145.1$ meV. Fig. 1 shows the 3 and 4 electron density as a function of z . The states are clearly Wigner molecule-like with a strong peak in the density corresponding to each electron: each electron is almost localised on its own lattice site and the probability of tunnelling between lattice sites is small.

To investigate the convergence of the 3 and 4-electron ground state energy and to compare the 'spherical' and 'cuboid' schemes for truncating the basis set we calculate E_N as a function of n , the maximum single particle quantum number that we include in our basis set. We assume

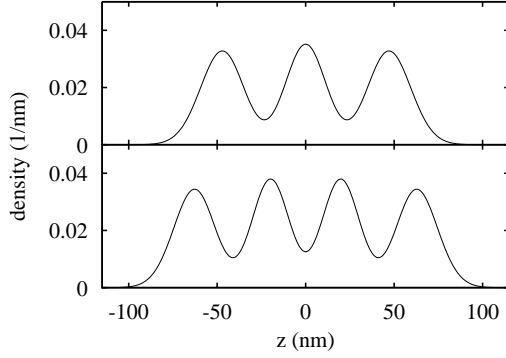


Figure 1. Ground state few-electron densities in a (35,0) SNT quantum dot ($R = 1.37$ nm, $m^* = 0.034$, $\hbar\omega = 5.0$ meV, $\epsilon_r = 2.0$). Top: 3 electron number density. Bottom: 4 electron number density.

that the lowest energy we obtain in our calculations (using the largest number of basis states, M_{max}) is converged. Then $\Delta E_N = E_N(M(n)) - E_N(M_{max})$ gives a measure of the convergence of the ground state energy for each n .

In Fig. 2 we plot ΔE_N as a function of $M(n)$. It is immediately clear that a large number of determinants are needed to give an accurate value for E_N and this is another indication that the electron correlation in SNT dots is important. For example, to converge the ground state energies to within 0.1 meV we need roughly $M = 150$ Slater determinants for 3 electrons and about $M = 3000$ for 4 electrons. It is also apparent that truncating the CI basis with the

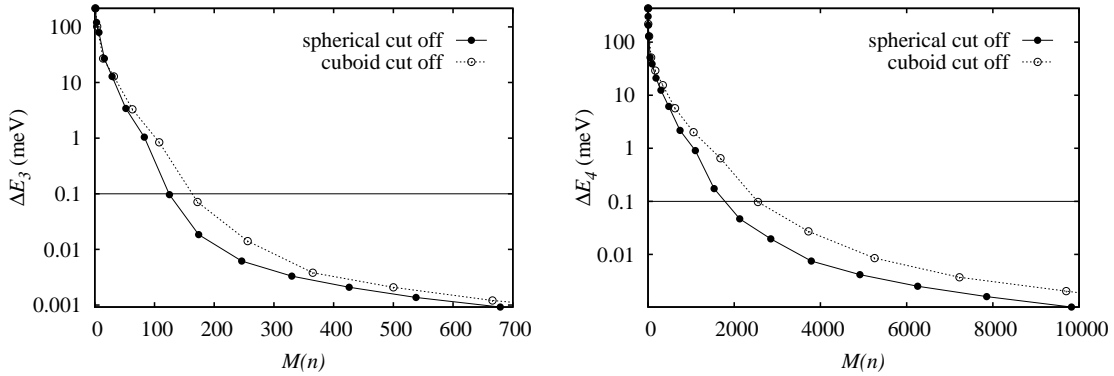


Figure 2. Convergence of the N -electron ground state energy as a function of the number of Slater determinants in the expansion. Left: 3 electrons. Right: 4 electrons. Each point indicates a different value of n in the calculation.

spherical cut off gives an advantage over the full CI method, and this advantage increases with increasing N . For example, to obtain $\Delta E_N = 0.1$ meV with 3 electrons we need $M = 124$ Slater determinants with the spherical cut-off and $M = 164$ with the cuboid cut off. So, using the spherical cut off reduces the computer time by a factor of roughly $(124/164)^3 = 0.57$. Similarly, for 4-electrons the calculation time is reduced by a factor $(1815/2568)^3 = 0.35$.

In conclusion, we have outlined our effective mass description of the N -electron states in SNT quantum dots and demonstrated that many Slater determinants are needed in an expansion of the states to get accurate ground state energies. Also, significant advantages can be obtained using a 'spherical' quantum number cut off to truncate the basis set.

References

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