Charging Energy of a Quantum Dot in the Thomas-Fermi Approximation

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Quantum dots are nanometer scale semiconductor devices. Their small size leads to unique behavior different from that of macroscopic semiconductors. Our objective is to generalize the Thomas-Fermi method of atomic physics to understand the electronic structure of quantum dots. The problems we wish to investigate include calculation of the charging energy of a non-neutral quantum dot at zero magnetic field which is an important quantity measured by transport experiments. For atoms the Thomas-Fermi approximation can be proved to be exact in the limit where the atomic number tends to infinity. We wish to explore whether there is a similar limit in which the Thomas Fermi approximation is exact for quantum dots. A combination of analytic and numerical methods will be used in this project.

Introduction

A quantum dot, in the form we are interested, is a nanometer scale structure of semiconducting material. The nanometer size results in the confinement of motion of the system's electrons in all three spatial directions.

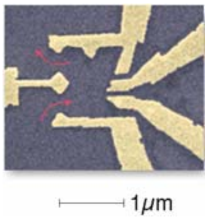

Fig. 1. In this case, electrons are confined vertically due to the interface between GaAs/AlGaAs, metallic gates confine the electrons laterally.

Picture taken from Marcus Lab Mesoscopic Physics

A quantum dot has a discrete quantized energy spectrum, much like an atom, only on a much smaller energy scale (~1 millielectron volt). The energy spectrum for a quantum dot is largely dependant upon the size and shape of the semiconductor nanostructure itself. Quantum confinement and the size of the nanocrystallites are responsible for the shift of the optical absorption threshold toward higher energies. As a consequence, quantum dots of the same material but differing sizes will emit different colors.

Fig. 2. Fluorescence induced by exposure to ultraviolet light in vials containing various sized Cadmium Selenide (CdSe) quantum dots.

Picture taken from Wikipedia article on Quantum Dots

Due to their optical properties quantum dots are being researched as a possible instrument in fluorescence spectroscopy, cutting edge electronics, diode lasers, amplifiers and biological sensors.

Quantum dots also have very important transport properties. The charging energy (energy required to gain/lose an extra electron) can be measured experimentally.

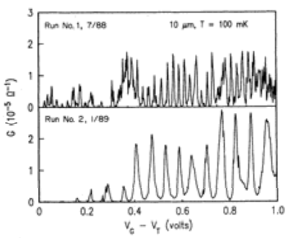

Fig. 3. In this graph M.A. Kastner et al measures the charging energy. The distance between peaks represents the voltage required to add an additional electron to the structure.

Graph taken from M.A. Kastner: A Single Electron Transistor

We wish to provide a means to calculate these values theoretically using the Thomas Fermi approximation.

Methods

The Kohn-Hohenberg theorem states that the energy of a system can be written down in terms of its electron density. The Thomas-Fermi equation is an approximation of *how* the energy depends on the electron density. This is done for an atom in Bethe and Jackiw's *Intermediate Quantum Mechanics*.

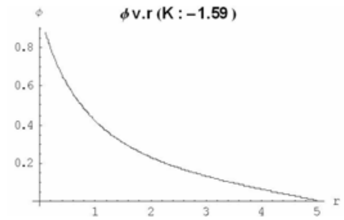

Fig. 4. The case in which Φ (which is a function of the electron density) asymptotically approaches zero as r approaches infinity corresponds to the atom.

We extend the Thomas Fermi equation to incorporate Quantum Dots. We solve the case of a neutral quantum dot ($Z=e^-$) and find that the electron density is uniformly smeared across the volume of the structure. This leads to the case of the non-neutral quantum dot ($Z=e^-+1$) and the following question:

How does an extra electron distribute itself over a quantum dot?

Results

We solve this problem by linearizing the Thomas-Fermi equation. The electron density is written as a sum of the uniform electron density for a neutral quantum dot plus a small correction that satisfies the linearized Thomas Fermi equation.

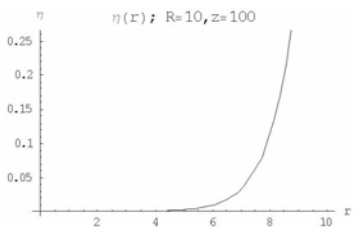

Fig. 5. The units of R are Bohr radii; this plot shows that the extra electron will position itself near the boundary of the spherical quantum dot.

An exact solution to this equation shows that for a large quantum dot the extra electron is confined largely to the surface; for a small quantum dot it is uniformly distributed over the volume of the dot. We find that the change in the electron density due to the extra electron simplifies in the limit that the dot is large or small compared to λ:

$$\delta\eta(r)_{small} = \frac{3}{4\pi R^3} \qquad \delta\eta(r)_{large} = [\frac{1}{2\pi R\lambda} e^{-\frac{R}{\lambda}}]\frac{1}{r}\sinh(\frac{r}{\lambda})$$

Eq. 1. The δη(r) approximations depend on the radius of the quantum dot (R) and λ

Here λ equals:

$$\lambda = (\frac{\pi^2}{144Z})^{1/6}\sqrt{R}$$

Results cont.

Next we calculate the charging energy which is the change in energy of the quantum dot due to the addition of an extra electron. We can compute this energy using the calculated change in electron density and the Thomas-Fermi equation which relates the energy of the quantum dot to the electron density. We find that the dominant interaction contribution to the charging energy is given by

$$E_{charging} = \frac{1}{4\pi\varepsilon_0}\frac{e^2}{R} S(R/\lambda)$$

Eq. 2. The charging energy of a non-neutral quantum dot

Fig. 6. shows a plot of S(R/λ) which we call the capacitance factor. In the limit that the quantum dot is very large (R >> λ) the capacitance factor is 1/2; for very small quantum dots (R << λ), it is 3/5. These values can be understood in terms of simple electrostatics as the potential energy of a spherical shell of charge and of a uniform ball of charge respectively. For very large and very small quantum dots the charging energy varies inversely with the radius of the quantum dot, the result expected from classical electrostatics. But we find that for quantum dots of intermediate size (R ~ λ) the Thomas-Fermi approximation shows the charging energy has a non-classical size dependence.

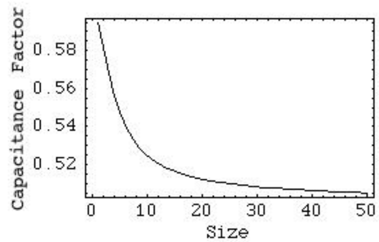

Fig.6. The capacitance factor for intermediate size quantum dots exhibiting non-classical size dependence.

Conclusion

The adaptation of the Thomas Fermi approximation for the neutral quantum dot led us to our predicted result: the electron density is uniform across the volume. In the case of the non-neutral quantum dot, we found that for a large quantum dot the extra electron is confined largely to the surface and for a small quantum dot it is uniformly distributed over the volume of the dot. We find that the charging energy of a quantum dot has a non-classical dependence on the size of the quantum dot. This is due to the emergence of a quantum length scale /λ which corresponds to the distance over which electric fields penetrate through the Thomas-Fermi electron liquid

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References

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2. Marcus Lab Mesoscopic Physics, http://marcuslab.harvard.edu/research.shtml (**Fig. 1.**)
3. Dr. D. Talapin, University of Hamburg, http://www.chemie.unihamburg.de/pc/Weller/ (**Fig. 2.**)
4. *The Single Electron Transistor*, M.A. Kastner, American Physical Society Vol. 64, No. 3, July **1992** (**Fig. 3.**)