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A spin-density-functional study of quantum dots and rings

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Abstract

We present a spin-density-functional theoretical (SDFT) study of the electronic states in GaAs quantum dots embedded in AlGaAs substrates. The SDFT allows for a systematic study of the joint effects of confinement, Coulomb interactions and spin for realistic systems. We model the system as electrons confined in a finite cylindrical dot. The screening due to the gate electrodes is also taken into account. The method predicts the electron addition energy spectra that are in agreement with experiments. We also apply the SDFT to GaAs quantum rings and find that they too show shell structures in the additional energy spectra. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Quantum dots; Quantum rings; Mesoscopic physics; Addition energy

Recent progress in experimental techniques has made fabrication of quantum systems, such as quantum dots and quantum rings, in nanometer scales possible. Due to the small dimensions of the systems, energy level spacings are correspondingly large. This, together with the electron–electron interactions, makes the quantum dots show significant shell structures similar to those of atoms. The properties of the quantum dots strongly depend on the number of electrons, which can be controlled experimentally. Tarucha et al. [1] have first observed the atomic-like properties in the addition energy spectra by measuring Coulomb oscillations in a vertical quantum dot.

This subject has been intensively studied during recent years by theoretical approaches, such as exact diagonalization [2], Hartree–Fock approximation [3], and density-functional theory [4,5]. Since the vertical quantum dot is a very thin disk, the dots are usually treated as two-dimensional (2D) systems. However, it would be more realistic to consider them as three-dimensional (3D) models [6]. In this work we carried out 3D spin-density-functional studies of the electronic structures of the quantum dots. In addition, we extended the study to quantum rings, and also found the clear shell structures.

We consider a cylindrical quantum dot (GaAs) with height d , embedded in a larger cylindrical box (AlGaAs) with height L and radius R . The electrons are confined in the dot with a harmonic confining potential in the x - y plane and a vertical rectangular confining well along the z direction. The well thickness is small compared to the radial dimension, thus only the lowest subband in the z direction is filled.

In an ideal 2D parabolic dot, the single-particle energy levels are $E_{n,l} = (2n + |l| + 1)\hbar\omega_0$ with the radial quantum number n and angular quantum number l . These levels form a pronounced shell structure with the magic electron numbers (N) being 2, 6, 12, 20, 30, The degeneracy of the shells could be reduced by the electron–electron interactions. To study the joint effects of confinement, electron–electron interactions and spin on the electronic structure of a 3D N -electron dot system, we apply the spin-density-functional theory (SDFT) and treat the exchange–correlation part of the electron–electron interactions in the local-spin-density approximation [7]. The self-consistent single-particle Kohn–Sham equations are

$$\left[-\frac{\hbar^2}{2m^*} \nabla^2 + V_{\text{eff}}^\sigma(\mathbf{x}) \right] \psi_{i,\sigma}(\mathbf{x}) = \epsilon_{i,\sigma} \psi_{i,\sigma}(\mathbf{x}) \quad (1)$$

with σ being the spin index. The electron density is $n(\mathbf{x}) = \sum_\sigma n^\sigma(\mathbf{x})$ with $n^\sigma(\mathbf{x}) = \sum_{i=1}^{N_\sigma} |\psi_{i,\sigma}(\mathbf{x})|^2$. The effective Kohn–Sham potential $V_{\text{eff}}^\sigma(\mathbf{x})$ consists of

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the external confinement V_{ext} , the Hartree potential of the electrons $V_H[n(\mathbf{x})]$, and the functional derivative of the local exchange-correlation energy $E_{\text{xc}}[n^\uparrow, n^\downarrow] = \int d(\mathbf{x}) n(\mathbf{x}) \varepsilon_{\text{xc}}(n^\uparrow, n^\downarrow)$. The wave functions are expanded in a set of bases which are eigenfunctions of single particle in the cylindrical box with height L and radius R , i.e. $A_{lj} e^{-il\phi} J_l(k_{lj}r) \sin m\pi z/L$, where A_{lj} is the normalization factor, k_{lj} is the root of Bessel function $J_l(k_{lj}R) = 0$ and l is the angular quantum number. The size of the basis set used is about 800 with $l_{\text{max}} = 4$, $j_{\text{max}} = 17$ and $m_{\text{max}} = 12$. We include the screening due to the gate electrodes when calculating the Hartree potential. The effects of screening by the electrodes have been discussed by Hallam et al. [8]. In our model, the gate electrodes are considered to immediately surround the boundary of the system. Thus, we can solve the Poisson equation using the method of image charge, and the Hartree potential is

$$V_H(\mathbf{x}) = \frac{e^2}{4\pi\epsilon_0\epsilon} \int_V n(\mathbf{x}') G(\mathbf{x}, \mathbf{x}') d\mathbf{x}' + \frac{1}{4\pi} \oint_S \left[G(\mathbf{x}, \mathbf{x}') \frac{\partial V_H(\mathbf{x}')}{\partial \mathbf{n}'} - V_H(\mathbf{x}') \frac{\partial G(\mathbf{x}, \mathbf{x}')}{\partial \mathbf{n}'} \right] d\mathbf{a}', \quad (2)$$

where the Green's function $G(\mathbf{x}, \mathbf{x}')$ was chosen to be zero at the cylinder boundary and satisfy $\nabla^2 G(\mathbf{x}, \mathbf{x}') = -4\pi\delta(\mathbf{x} - \mathbf{x}')$. To simplify the calculation, we also let the gate voltage on the boundary to be zero. The ground state energy of the system is then obtained from

$$E(N) = \sum_{\sigma} \sum_{i=1}^{N_{\sigma}} \varepsilon_{i,\sigma} - \frac{1}{2} \int d\mathbf{x} n(\mathbf{x}) V_H(\mathbf{x}) - \sum_{\sigma} \int d\mathbf{x} n(\mathbf{x}) \frac{\delta E_{\text{xc}}[n^\uparrow, n^\downarrow]}{\delta n_{\sigma}(\mathbf{x})} + E_{\text{xc}}[n^\uparrow, n^\downarrow]. \quad (3)$$

We have first calculated the addition energy of the quantum dot up to $N = 29$. The material parameters chosen are typical GaAs values, effective mass $m^* = 0.067m_e$, dielectric constant $\epsilon = 12.4$. The dot height is $d = 12$ nm, and the strength of the radial confinement is set to $\hbar\omega_0 = 4$ meV, corresponding to those of experiment [1]. The addition energy needed to put an extra electron in the dot is obtained from $\Delta\mu(N) = \mu(N+1) - \mu(N)$, while the Coulomb oscillation peaks occur at the chemical potential $\mu(N) = E(N) - E(N-1)$.

Fig. 1 (upper) shows the addition energy as a function of the electron number (N) for two different heights of the cylindrical box, $L = 48$ nm and $L = 84$ nm, and compares with the experiment. The radius of the cylindrical box, R , is 160 nm in all the cases considered in this paper. For the smaller L , the electrons are closer to the gate electrodes and the effects of screening are more significant. The addition energy is then lower due to the screening. For both cases, there are prominent peaks at $N = 2, 6, 12$, and 20, which correspond to the magic number that form the closed shells. There are also

Fig. 1. The addition energy $\Delta\mu$ (upper) and the total spin S (lower) as a function of the electron number N for the quantum dots of height $d = 12$ nm, radius $R = 250$ nm, and $\hbar\omega_0 = 4$ meV with $L = 48$ and 84 nm. The experimental $\Delta\mu$ is also shown for comparison.

subpeaks at $N = 4, 9$ both for $N = 16$ and for $L = 48$ nm, corresponding to the half-filled shells. However, when the number of electrons increases, the peak heights decrease and the subpeaks are suppressed. This is due to the electron–electron interactions that weaken the structure of $\Delta\mu(N)$ oscillations.

The occurrence of the subpeaks can be explained by Hund's rule. Fig. 1 (lower) shows the total spin as a function of electron number. We see that the electrons in general prefer to be in the highest possible total spin. Since the states of spin symmetry are spatial antisymmetry, the Coulomb repulsion energy between the electrons will be lowered significantly. This causes the addition energy to be relatively high when the shell is half filled. However, when $N > 22$, Hund's rule is violated and high-spin states are suppressed for the case of $L = 84$ nm though Hund's rule is still obeyed for $L = 48$ nm. The breakdown of Hund's rule has been found in 2D models [5] which is due to the nonparabolic effective potential caused by Coulomb interactions. Here incorporating the 3D models and the boundary effects, we see that the breakdown of Hund's rule disappears in the case of $L = 48$ nm. The different cylindrical height of the boundary may change the distributions of electrons slightly and deform the radial parabolic potential to different extents. For $L = 48$ nm, the Coulomb interactions are more reduced by the screening and the radial parabolic potential is less deformed, thus, Hund's rule still holds for large electron numbers.

We have extended the calculation to the case of quantum rings. The radial confinement of the quantum ring is of the form $\frac{1}{2}m^*\omega_0(r - r_0)^2$, where r_0 is the radius

Fig. 2. The addition energy $\Delta\mu$ (upper) and the total spin S (lower) as a function of the number of electrons N for the quantum ring for two different α values.

of the ring. The strength of the radial confinement can be determined by the dimensionless quantity [9] $\alpha = \omega_0 m^* \pi r_0^2 / \hbar$. When α is large ($\alpha > 20$), the electrons are laterally confined in a closely 1D ring, whose single-particle energy levels are $E_l = (\hbar^2 / 2m^* r_0^2) l^2$, where $l = 0, \pm 1, \pm 2, \dots$. The closed shells are formed at $N = 2, 6, 10, 14, 18, \dots$.

Here we calculate the addition energy and total spin for two different values of α , as shown in Fig. 2. For $\alpha = 1.0$, we take $\hbar\omega_0 = 12$ meV and $r_0 = 14$ nm, corresponding to the experiment in Ref. [10], and for $\alpha = 5.3$, $\hbar\omega_0 = 30$ meV and $r_0 = 20$ nm are taken for comparison. Fig. 2 shows that the first several peak heights of $\Delta\mu$, within $N < 6$ for $\alpha = 1.0$ and $N < 14$ for $\alpha = 5.3$, rise up as the number of electrons increases. This is due to the increasing energy level spacings in the single-particle picture. These peaks also fall exactly on the closed shells of an ideally narrow ring whereas those at larger N do not. When the number of electrons becomes larger, the electrons start to fill in higher energy levels which are identified not only by the angular quantum number l but also the radial one n , like that in the quantum dot.

Therefore, only the few lowest subbands belong to different angular momentum. If we turn down the value of α to make the ring wider, the upper subbands with nonzero n will come down in energy, and finally the addition energy spectra will approach that of the quantum dot when $\alpha = 0$. As to the spin configuration of the quantum ring, we see that the total spin also takes the maximum values allowed by the exclusion principle and becomes zero for closed shells. Hund's rule still holds for the quantum ring.

In summary, we have calculated the addition energy spectra for the quantum dots and rings using the SDFT. We consider realistic 3D models and reproduce the shell structures of the quantum dot observed in the experiment [1]. The effects of screening due to the gate electrodes are taken into account and are shown to play some important role in the addition energy spectra. Under certain boundary conditions, the electrons will fill the dot according to Hund's rule. We have shown that the quantum ring also has some shell structures, which should vary with the width of the ring.

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References

- [1] S. Tarucha, et al., Phys. Rev. Lett. 77 (1996) 3613.
- [2] S. Tarucha, et al., Physica E 3 (1998) 112.
- [3] H. Tamura, Physica B 249–251 (1998) 210.
- [4] M. Koskinen, M. Manninen, S.M. Reimann, Phys. Rev. Lett. 79 (1997) 1389.
- [5] K. Hirose, N.S. Wingreen, Phys. Rev. B 59 (1999) 4604.
- [6] M. Rontani, F. Rossi, F. Manghi, E. Molinari, Phys. Rev. B 59 (1999) 10165.
- [7] S.H. Vosko, L. Wilk, M. Nusair, Can. J. Phys. 58 (1980) 1200.
- [8] L.D. Hallam, J. Weis, P.A. Maksym, Phys. Rev. B 53 (1996) 1452.
- [9] T. Chakraborty, P. Pietiläinen, Phys. Rev. B 50 (1994) 8460.
- [10] A. Lorke, et al., Phys. Rev. Lett. 84 (2000) 2223.