The accuracy of the Hartree-Fock approximation for quantum dots

1992 J. Phys.: Condens. Matter 4 L623

(http://iopscience.iop.org/0953-8984/4/47/003)

J. Phys.: Condens. Matter 4 (1992) L623–L628. Printed in the UK

LETTER TO THE EDITOR

The accuracy of the Hartree–Fock approximation for quantum dots

N F Johnson† and M Reina

Departamento de Fisica, Universidad de Los Andes, Bogota, AA 4976, Colombia

Received 8 September 1992

Abstract. We investigate the accuracy of the Hartree–Fock (HF) and Hartree (H) approximations for calculating energies in an N-electron quantum dot. The HF and H energies calculated from an analytically solvable model Hamiltonian are compared to exact energy solutions. The HF and H approximations become less accurate with increasing number of electrons, decreasing magnetic field, increasing dot size and increasing electron–electron interaction strength. The dependence on electron number is contrary to the well known infinite electron gas result.

Current flexibility in semiconductor processing allows fabrication of zero-dimensional quantum dots (see, e.g., [1]) which are islands of quasi two-dimensional electrons formed by the additional confinement of a 2D electron gas within the plane. Each dot can contain anywhere from a few to hundreds of electrons [1]. A full understanding of the many recent optical and transport measurements on quantum dots requires detailed knowledge of the energies of the N-electron dot. However, the relevant energy scales, such as the single-electron confinement energy, the cyclotron energy, and the electron–electron interaction strength, are typically comparable making treatment of the many-body problem difficult. Apart from the computationally intensive approach of direct numerical diagonalization of the N-electron Hamiltonian [2], the Hartree [3] approximation has been used. However, the accuracy of standard many-body theories, which are often based on mean-field approximations (e.g. Hartree or Hartree–Fock), is unclear for few-electron systems.

This paper investigates the accuracy of the Hartree–Fock (HF) and Hartree (H) approximations for calculating the properties of an interacting N-electron gas in a quantum dot, subject to an external magnetic field. Algebraic expressions are obtained for HF and H energies of an analytically solvable model Hamiltonian of an N-electron dot [4, 5], and are compared to the exact result. We find that both HF and H approximations become less accurate with (i) increasing number of particles, (ii) decreasing magnetic field, (iii) increasing dot size and (iv) increasing electron–electron interaction strength. The dependence on electron number appears to be opposite to the result for an infinite 2D (see, e.g., [6]) or 3D (see, e.g., [7]) electron gas, where the

---

† Now at: Physics Department, Clarendon Laboratory, Oxford University, UK.

0953-8984/92/470623+06$07.50

HF approximation improves with increasing electron density. The resolution of this apparent contradiction lies in the non-linear N-dependence of the electron density in quantum dots with 'soft-wall' (parabolic [1]) confinement potentials. We finish by discussing the validity of Koopmans' theorem [8].

Our exactly solvable model Hamiltonian for an isolated 2D quantum dot in a perpendicular magnetic field $\boldsymbol{B}$ is given by [4, 5]

$$H = \frac{1}{2m^*}\sum_i \left(\boldsymbol{p}_i + \frac{e\boldsymbol{A}_i}{c}\right)^2 + \tfrac{1}{2}m^*\omega_0^2\sum_i |\boldsymbol{r}_i|^2 + \sum_{i<j} V(\boldsymbol{r}_i - \boldsymbol{r}_j) - g^*\mu_{\mathrm{B}}B\sum_i s_{i,z}. \tag{1}$$

The parabolic dot lies in the x–y plane and contains N interacting electrons with effective mass m^*, negative charge $-e$, g-factor g^*, spatial coordinates $\{\boldsymbol{r}_i\}$, and spin components $\{s_{i,z}\}$ along the z-axis. The momentum and vector potential associated with the ith particle are $\boldsymbol{p}_i$ and $\boldsymbol{A}_i$ respectively, and μ_{B} is the Bohr magneton. Many recent experimental studies of infrared absorption in semiconductor quantum dots [1] have shown the parabolic confining potential to be realistic, based on the generalized Kohn theorem [9]. The interaction potential is given by the first two terms in the Taylor series expansion of a cut-off Coulomb interaction

$$V(\boldsymbol{r}_i - \boldsymbol{r}_j) = 2V_0 - \tfrac{1}{2}m^*\Omega^2|\boldsymbol{r}_i - \boldsymbol{r}_j|^2 + \cdots \tag{2}$$

where V_0 and Ω are positive parameters which can be chosen to model dots of different sizes and materials [4]. The saturation of $V(\boldsymbol{r}_i - \boldsymbol{r}_j)$ for small electron separations models the effect of the finite electron wavefunction spread along the perpendicular (z) direction (see later discussion). The HF approximation proceeds [7, 10] by constructing a totally antisymmetric state from individual single-electron spin-orbital functions $\{\phi_i(\boldsymbol{x})\}$ by means of a Slater determinant. Following [10] we use $\boldsymbol{x}_i$ to denote the 4D manifold of space and spin coordinates of the ith electron; each spin-orbital function $\phi_i(\boldsymbol{x})$ corresponds to a product of a spatial wavefunction and a spinor. Acting with the Hamiltonian H on the Slater determinant, and minimizing with respect to the individual values of ϕ, yields the following set of N integro-differential equations ($i = 1, \ldots, N$):

$$H_i^{\mathrm{eff}}\phi_i(\boldsymbol{x}) = \epsilon_i\phi_i(\boldsymbol{x}) \tag{3}$$

where

$$H_i^{\mathrm{eff}} = H_0 + \sum_{j\neq i}\int \mathrm{d}\boldsymbol{x}'\,\phi_j^*(\boldsymbol{x}')V(\boldsymbol{x} - \boldsymbol{x}')(1 - P_{ij})\phi_j(\boldsymbol{x}') \tag{4}$$

with P_{ij} an operator which exchanges the subscripts i and j occurring to the right of it. The interaction $V(\boldsymbol{x} - \boldsymbol{x}')$ given in (2) is diagonal in the spinor basis, and

$$H_0 = (1/2m^*)(\boldsymbol{p} + e\boldsymbol{A}/c)^2 + \tfrac{1}{2}m^*\omega_0^2|\boldsymbol{r}|^2 - g^*\mu_{\mathrm{B}}Bs_z. \tag{5}$$

The total HF energy for the N-electron gas can be written as

$$E_{\mathrm{HF}} = \tfrac{1}{2}\sum_i(\epsilon_i + H_i) = \sum_i H_i + \sum_{j>i}(J_{ij} - K_{ij}) \tag{6}$$

where

$$H_i = \int \mathrm{d}\boldsymbol{x}\, \phi_i^*(\boldsymbol{x}) H_0 \phi_i(\boldsymbol{x}). \tag{7}$$

The direct and exchange integrals are given respectively by

$$J_{ij} = \int\int \mathrm{d}\boldsymbol{x}\, \mathrm{d}\boldsymbol{x}'\, \phi_i^*(\boldsymbol{x})\phi_j^*(\boldsymbol{x}')V(\boldsymbol{x}-\boldsymbol{x}')\phi_j(\boldsymbol{x}')\phi_i(\boldsymbol{x}) \tag{8}$$

$$K_{ij} = \int\int \mathrm{d}\boldsymbol{x}\, \mathrm{d}\boldsymbol{x}'\, \phi_i^*(\boldsymbol{x})\phi_j^*(\boldsymbol{x}')V(\boldsymbol{x}-\boldsymbol{x}')\phi_i(\boldsymbol{x}')\phi_j(\boldsymbol{x}). \tag{9}$$

The H energy E_{H} differs from E_{HF} through the absence of the exchange integrals ($K_{ij} = 0$ in equation (6)). In order to solve the self-consistent HF equations, we follow the standard method of Roothaan [10, 11] in which the spin orbitals ϕ are expressed as linear combinations in a given set of basis functions,

$$\phi_i(\boldsymbol{x}) = \sum_{\alpha=1}^{N'} c_{i\alpha}\chi_\alpha(\boldsymbol{x}). \tag{10}$$

The set of HF integro-differential equations (equation (3)) can then be transformed into an equivalent set of algebraic equations for the coefficients c_{i_α} [10]. For a given basis set $\{\chi_\alpha\}$ an initial set of coefficients c_{i_α} must then be chosen, and the algebraic Roothaan equations [10] iterated to obtain self-consistency. In general this is a heavy computational task. A suitable choice for the set $\{\chi_\alpha\}$ are the single-electron states corresponding to an electron in a 2D parabolic potential subject to an external magnetic field [12]. With (r, θ) denoting the polar coordinates in the x–y plane we have the (unnormalized) spatial part of $\chi_\alpha(\boldsymbol{x})$ given by [12]

$$\chi_n^l(\boldsymbol{r}) = \exp(-\mathrm{i}l\theta)\exp(-[m^*\omega_0(B)/2\hbar]r^2)r^{|l|}L_n^{|l|}([m^*\omega_0(B)/\hbar]r^2) \tag{11}$$

where $L_n^l(x)$ are associated Laguerre polynomials; $\omega_0(B) = \sqrt{(\omega_0^2 + \omega_c^2/4)}$ and the cyclotron frequency $\omega_c = eB/m^*c$. We consider the external magnetic field to be sufficiently large that the electrons in the dot are spin polarized. In the InSb quantum dots of Sikorski and Merkt [1] where $\hbar\omega_0 = 7.5$ meV, a field as small as 3 T is sufficient to ensure spin polarization for $N = 4$ electrons. The magnetic field required for spin polarization also guarantees that all the electrons lie in the lowest Landau level ($n = 0$, $l \geqslant 0$) in the absence of electron–electron interactions [13]. For computational simplicity we employ the so-called minimum basis set [10, 11] where $N' = N$. The number of basis functions therefore equals the number of electrons in the dot. Later we will discuss relaxation of this constraint.

The self-consistent HF equations can now be solved analytically, yielding the HF total energy for N electrons

$$E_{\mathrm{HF}} = \tfrac{1}{2}N(N+1)\hbar\omega_0(B) - \tfrac{1}{4}N(N-1)(N+2)\hbar\Omega^2/\omega_0(B) \\ - \tfrac{1}{4}N(N-1+4\zeta)\hbar\omega_c + N(N-1)V_0 \tag{12}$$

where $\zeta = g^*m^*/4m_0$. Likewise the H energy is given by

$$E_{\mathrm{H}} = \frac{N(N+1)}{2}\hbar\omega_0(B) - \frac{N(N^2-1)}{4}\frac{\hbar\Omega^2}{\omega_0(B)} - \frac{N(N-1+4\zeta)}{4}\hbar\omega_c + N(N-1)V_0. \tag{13}$$

The corresponding exact energy solution of H is given by

$$E = \hbar\omega_0(B) + \tfrac{1}{2}(N-1)(N+2)\hbar\Omega_0(B) - \tfrac{1}{4}N(N-1+4\zeta)\hbar\omega_c + N(N-1)V_0 \tag{14}$$

where $\Omega_0(B) = \sqrt{(\omega_0^2(B) - N\Omega^2)}$. If the electron–electron interaction parameter Ω is small (i.e. the electron–electron interaction is nearly constant across the diameter of the electron gas), we have

$$\Omega_0(B) = \sqrt{\omega_0^2(B) - N\Omega^2} \approx \omega_0(B)(1 - N\Omega^2/2\omega_0^2(B)) \tag{15}$$

and the expression for the exact energy E now becomes equal to the HF energy E_{HF}. For general values of Ω, E_{HF} is larger than E since the variational HF procedure provides an upper bound to the energy. The difference between E_{HF} and E is the so-called correlation energy.

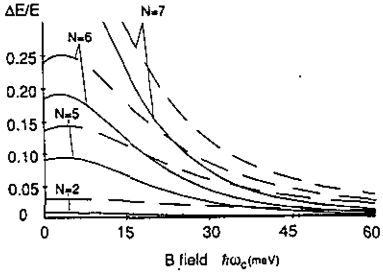

Figure 1. Fractional error $(E_{\mathrm{HF}} - E)/E$ of the HF approximation (solid line) as a function of the $\hbar\omega_c$ (magnetic field) for different N (electron number). Also shown is the fractional error for the H approximation (dashed line). The parameters used in equations (12)–(14) correspond to $\hbar\omega_0 = 15$ meV, $V_0 = 5$ meV, $\hbar\Omega = 5.6$ meV and $\zeta = 0.007$.

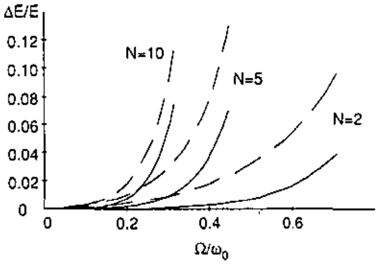

Figure 2. Fractional error $(E_{\mathrm{HF}} - E)/E$ of the HF approximation (solid line) as a function of the ratio of Ω (electron–electron interaction) to ω_0 (dot confinement) for different N (electron number). Also shown is the fractional error for the H approximation (dashed line). Dot parameters are as in figure 1, and $\hbar\omega_c = 2\hbar\omega_0 = 30$ meV.

Figure 1 shows the fractional error $(E_{\mathrm{HF}} - E)/E$ of the HF approximation (solid line) as a function of the magnetic field for different values of N, the number of electrons inside a quantum dot. Also shown is the fractional error $(E_{\mathrm{H}} - E)/E$ for the H approximation (dashed line). Figure 2 shows the same quantities as a function of Ω/ω_0, the ratio of the electron–electron interaction parameter Ω to the confinement ω_0. We first comment on the behaviour in figures 1 and 2 for fixed N. As expected E_{HF} and E_{H} are always greater than E. In addition E_{HF} represents a better approximation to the exact energy E than does E_{H}. In figure 1, the fractional error $\Delta E/E$ at fixed N for both the HF and H approximations shows a slight initial increase with magnetic field, before decreasing steadily as the magnetic field is further increased. The close agreement between E_{HF} and E_{H} at high magnetic field follows from the shrinkage of the individual electron wavefunctions, which leads to a decreased importance of the Pauli exclusion principle and hence of exchange effects. It is interesting to note that an increase in the magnetic field will increase the effective confinement $\omega_0(B)$, and reduce the size of the electron gas droplet. The effective density of the electron gas, defined as N/A where A is the area of the droplet, will therefore increase as the magnetic field increases. This overall improvement of HF and H with increasing density agrees with the trend for an infinite 2D [6] or 3D [7] electron gas. Figure 2 shows that both the HF and H approximations become worse as the electron–electron interaction parameter Ω increases. The electron–electron interaction is now varying more rapidly with electron separation, and the mean-field description of the electrons is breaking down (i.e. correlation effects are becoming more important). Figure 2 also shows that both HF and H approximations become worse with decreasing ω_0. A decrease in ω_0 is equivalent to an increase in dot size, enabling the electron gas droplet to expand and hence reduce its effective density. Once again we have the same density dependence as for the infinite electron gas. The most surprising feature from figures 1 and 2 is that both HF and H approximations become *worse* as the number of electrons N is increased, which seems contrary to the infinite gas result for increasing densities. The explanation is that the 'soft' walls of the parabolic dot potential allow the electron gas to spread out as N is increased. The effect on the effective electron gas density N/A as N increases is therefore non-trivial. When $\Omega \approx 0$ the area A of the electron droplet is essentially proportional to N, hence the electron gas density is independent of N. As Ω is increased, the droplet size A actually increases faster than N, implying a decrease in density. We note that for $(\Omega/\omega_0) > N^{-1/2}$ in figure 2, states with higher angular momentum become lower in energy than the state with energy E considered until now [14]. The minimal basis set chosen in the present calculation is therefore no longer suitable, and we have consequently truncated the curves in figure 2 at $(\Omega/\omega_0) = N^{-1/2}$. The accuracy of the HF approximation for these states will be analysed elsewhere.

The HF solution presented above can be improved upon by increasing the size N' of the minimal basis set. In particular, states from higher Landau levels ($n > 0$, $l \geqslant 0$; $n \geqslant 0$, $l < 0$) should be included. For $N > 2$ the subsequent HF (Roothaan) algebraic equations cannot easily be solved analytically. However, for $N = 2$ electrons in the singlet state, we have managed to obtain analytic solutions including basis states from *all* Landau levels (i.e. N' is infinite). The resulting HF energy is given by

$$E_{\mathrm{HF}} = 2\hbar\sqrt{\omega_0^2(B) - \Omega^2} + 2V_0 \tag{16}$$

while the exact energy is

$$E = \hbar\omega_0(B) + \hbar\sqrt{\omega_0^2(B) - 2\Omega^2} + 2V_0. \tag{17}$$

Performing a series expansion of E_{HF} in equation (16) yields terms to *all* orders of the electron–electron interaction parameter Ω^2. This is in contrast to the minimal basis solutions presented earlier which only accounted for the electron–electron interaction to order Ω^2. However, the coefficients in the expansion of E_{HF} still only agree with those in the expansion of E to order Ω^2. Even though we have gone beyond the minimal basis set solution, we still have the results that the HF approximation becomes worse with decreasing magnetic field, increasing dot size (i.e. decreasing ω_0), and increasing electron–electron interaction Ω.

The above HF energies allow us to comment on the validity of Koopmans' theorem [8] for quantum dot systems. Within the HF approximation, the ionization energy associated with removing the Nth electron from the dot has magnitude $I_N = E_{\rm HF}(N) - E_{\rm HF}(N-1)$ where $E_{\rm HF}(N)$ is the HF total energy for N electrons given in equation (12). Koopmans' theorem states that $I_N = \epsilon_N$ where ϵ_N is the HF one-electron energy for orbital N given by equation (3). However, it is straightforward to show from equation (12) that

$$I_N = \epsilon_N + N\Omega^2/2\omega_0(B). \tag{18}$$

As is the case for the HF approximation itself, Koopmans' theorem therefore loses its validity with increasing electron number N, decreasing magnetic field, increasing dot size and increasing electron–electron interaction Ω. Note that the energy I_N is of direct physical importance in quantum dot tunnelling measurements, since it is essentially the energy of the conductance peak arising from an Nth electron tunnelling on to a dot containing $N-1$ electrons.

Finally we emphasize that the true form of the electron–electron interaction $V(\boldsymbol{r}_i - \boldsymbol{r}_j)$ in experimental dots is unknown; it will differ from the Coulomb form due to image charges in adjacent layers and gates, and the finite extent of the electron wavefunction in the z-direction. Indeed $V(\boldsymbol{r}_i - \boldsymbol{r}_j)$ may not actually be translationally invariant. Although the simple model form used in this paper is not the most realistic, it has allowed us to investigate *analytically* the accuracy of two standard many-body theories in quantum dot systems. Any improvement upon the present results by employing a more realistic form for the interaction is certain to require heavy numerical computation.

We thank L Quiroga for useful discussions. This work was supported by COLCIENCIAS (Colombia), the Comite de Investigaciones de la Universidad de Los Andes (Colombia), and by St John's College, Cambridge (UK).

References

[1] Tewordt M, Law V, Kelly M, Newbury R, Pepper M, Peacock D, Frost J, Ritchie D and Jones G 1990 *J. Phys.: Condens. Matter* **2** 8969
Meurer B, Heitmann D and Ploog K 1992 *Phys. Rev. Lett.* **68** 1371
[2] Maksym P A and Chakraborty T 1990 *Phys. Rev. Lett.* **65** 108
[3] Kumar A, Laux S and Stern F 1990 *Phys. Rev.* B **42** 5166
[4] Johnson N F and Payne M C 1991 *Phys. Rev. Lett.* **67** 1157; 1992 *Phys. Rev.* B **45** 3819
[5] Johnson N F 1992 *J. Phys.: Condens. Matter* **4** L555
[6] Ando T, Fowler A B and Stern F 1982 *Rev. Mod. Phys.* **54** 437
[7] Gross E, Runge E and Heinonen O 1991 *Many-Particle Theory* (New York: Adam Hilger)
[8] Koopmans T A 1933 *Physica* **1** 104
[9] Brey L, Johnson N F and Halperin B I 1989 *Phys. Rev.* B **40** 10647
[10] Blinder S M 1965 *Am. J. Phys.* **33** 431
[11] Roothaan C C J 1951 *Rev. Mod. Phys.* **23** 69
[12] Fock V 1928 *Z. Phys.* **47** 446
[13] Johnson N F and Reina M unpublished
[14] Johnson N F 1992 *Phys. Rev.* B **46** 2636